Nitrogen balance of a boreal Scots pine forest

J. F. J. Korhonen¹, M. Pihlatie¹, J. Pumpanian², H. Aaltonen², P. Hari², J. Levula³, A.-J. Kieloaho¹, E. Nikinmaa², T. Vesala¹, and H. Ilvesniemi⁴

¹Department of Physics, University Of Helsinki, P.O. Box 48, 00014, Helsinki, Finland
²Department of Forest Sciences, University Of Helsinki, P.O. Box 27, 00014, Helsinki, Finland
³Hyytiälä Forestry Field Station, Hyytiäläntie 124, 35500, Korkeakoski, Finland
⁴Finnish Forest Research Institute, Vantaa Unit, P.O. Box 18, 01301, Vantaa, Finland

Correspondence to: J. F. J. Korhonen (janne.fj.korhonen@helsinki.fi)

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Abstract. The productivity of boreal forests is considered to be limited by low nitrogen (N) availability. Increased atmospheric N deposition has altered the functioning and N cycling of these N-sensitive ecosystems by increasing the availability of reactive nitrogen. The most important components of N pools and fluxes were measured in a boreal Scots pine stand in Hyytiälä, Southern Finland. The measurements at the site allowed direct estimations of nutrient pools in the soil and biomass, inputs from the atmosphere and outputs as drainage flow and gaseous losses from two microcatchments. N was accumulating in the system, mainly in woody biomass, at a rate of 7 kg N ha⁻¹ yr⁻¹. Nitrogen input as atmospheric deposition was 7.4 kg N ha⁻¹ yr⁻¹. Dry deposition and organic N in wet deposition contributed over half of the inputs in deposition. Total outputs were 0.4 kg N ha⁻¹ yr⁻¹, the most important outputs being N₂O emission to the atmosphere and organic N flux in drainage flow. Nitrogen uptake and retranslocation were equally important sources of N for plant growth. Most of the assimilated N originated from decomposition of organic matter, and the fraction of N that could originate directly from deposition was about 30%. In conclusion, atmospheric N deposition fertilizes the site considerably, but there are no signs of N saturation. Further research is needed to estimate soil N₂ fluxes (emission and fixation), which may amount up to several kg N ha⁻¹ yr⁻¹.

1 Introduction

Anthropogenic emissions of reactive nitrogen (N) have markedly increased the atmospheric N deposition to forests, especially around industrialized regions (Pinho et al., 2012; Gruber and Galloway, 2008; Galloway et al., 2003). There is, however, a large spatial variability in the effects by N deposition (Magnani et al., 2007; Fischer et al., 2010). Forest ecosystems with slow N cycling and low or moderate atmospheric N deposition are called nitrogen limited, as their productivity is enhanced by the increased N inputs. Forest ecosystems with high deposition rates receive N in excess, which increases the outputs markedly. In such a case the ecosystems are called nitrogen saturated (Aber et al., 1998). Here we divide N cycling in forests into inputs, outputs and internal cycling. Inputs include atmospheric N deposition, fixation, and fertilization. Outputs include N losses in gaseous emissions and drainage flow. Internal cycling includes all the processes where N is transported within the ecosystem. The inputs and outputs of N to and from undisturbed forest ecosystems are small, and the internal cycling dominates the N flow (Målkönen, 1974). The increased N deposition in turn affects the whole N-cycling process in forest ecosystems.

In boreal forests, the inputs of N via atmospheric deposition and N₂ fixation are relatively small, at maximum around 10 kg N ha⁻¹ yr⁻¹ (Flechard et al., 2011; Syrjälä et al., 2004; Mustajärvi et al., 2008) and 0.1–3.5 kg N ha⁻¹ yr⁻¹ (DeLuca et al., 2002, 2008; Zackrisson et al., 2004, 2009), respectively. In these systems, the N outputs are reported to be small, both via leaching of ammonium (NH₄⁺-N), nitrate
(NO$_3$-N) and dissolved organic nitrogen (DON) into groundwater (Kubin, 1998), and through volatilization of oxidized nitrogen (N$_2$O, NO$_x$) into the atmosphere (Pilegaard et al., 2006; Pihlatur et al., 2007; Maljanen et al., 2010). The soil emissions of N$_2$ are highly uncertain as there are no measurements available from boreal ecosystems. In general, the total N inputs and outputs in boreal forests are markedly less than those in more N-affected Central European forest ecosystems (Flechard et al., 2011; Holland et al., 2005). In boreal forests, N is reported to accumulate into the soil (Berg and Difie, 2004; Hattenschwiler and Vitousek, 2000) and biomass of growing trees.

According to the current knowledge, plants can take up N either in mineral (NH$_4^+$ or NO$_3^-$) or amino acid forms (e.g. Kielland et al., 2006). Nitrogen uptake by plants is affected by the availability of these compounds, released via decomposition, but also by the atmospheric N deposition and N$_2$ fixation. Even though there are large pools of N in the boreal ecosystems, there is less N available for plant uptake than plants are able to consume. This is concluded from the fact that N fertilization tends to increase the productivity of boreal forests (Saarsalmi and Mälkönen, 2001). In boreal upland forests, the low N availability results from the cool climate and the chemical composition of soil organic matter. However, plants can reuse N efficiently because it is a mobile nutrient. A major fraction of the N that is being lost in senescing plant tissue can be resorbed and retranslocated when new tissue is grown.

As several studies recognize, the role and magnitude of organic N inputs to and outputs from the ecosystems and direct plant uptake has largely been overlooked (Kielland et al., 2007; Neff et al., 2002; Mustajärvi et al., 2008). Recent studies show that organic N can contribute as much as 30% of the total N deposition into ecosystems (Neff et al., 2002), and up to 80% of the total N lost as runoff (Mustajärvi et al., 2008). It seems evident that trees uptake N from soil directly as amino acids (Jones and Kielland, 2002; Kielland et al., 2007).

Here we present a measurement-based N budget of a boreal Scots pine forest in Southern Finland. We show the N budget based on a comprehensive data series covering the inputs, outputs, pools, and internal cycling of N within the forest ecosystem from 2006 to 2010. We further calculate the individual sources of N for the plants and the total N atmospheric deposition. The study compliments the long-term measurements of carbon and water balances of the site, which are presented in Ilvesniemi et al. (2009, 2010), respectively.

### 2 Materials and methods

#### 2.1 Measurement site

Measurements were conducted at a Scots pine stand at the SMEAR II station in Hyytiälä (Hari and Kulmala, 2005), Southern Finland (61°51’ N, 24°17’ E). The mean annual air temperature and precipitation at Hyytiälä from 1971 to 2000 were 3.3°C and 713 mm, respectively (Drebs et al., 2002). The stand is an even-aged forest, and it was regenerated by sowing after clear-cutting, prescribed burning, and soil preparation in 1962. The measurement station was established in 1995. The stand was partially thinned from January to March 2002 (Vesala et al., 2005).

The dominant tree species is Scots pine, covering 93% of the stem basal area in the study area. Of the other tree species, mainly in the understorey, the most important are Norway spruce (Picea abies) and Silver birch (Betula pendula), contributing 2.6% and 1.1% of the total basal area, respectively. Other species in the understorey include rowan (Sorbus aucuparia), Grey alder (Alnus incana), Goat willow (Salix caprea) and Eurasian aspen (Populus tremula). The dominant species in the field layer are the shrubs Vaccinium myrtillus and Vaccinium vitis-idaea, and in the ground layer the mosses Dicranum polysetum and Pleurozium schreberi (Kulmala et al., 2008).

The soil at the site is Haplic podzol on glacial till (FAO-UNESCO-ISRIC, 1988), overlaying homogeneous granite bedrock at an average of 0.6 m depth. The study area is defined as the area inside the borders of two mini-catchments (C1, 889 m$^2$, and C2, 301 m$^2$) that receive water only from precipitation, since they are located at the top of a small hill (Fig. 1). The mini-catchment borders were mapped based on bedrock topography, and measured by soil penetrating radar in 1994. Two concrete weirs were built to guide the lateral outflow of water to outlets of the two weirs. The radar measurement showed that there were no major vertical cracks in the bedrock. The lowest soil layer on the bedrock has a high silt fraction, rendering the watershed water tight. Thus water flows in the soil along the direction of the slope and outflow occurs only via the outlets in the weirs as reported in Ilvesniemi et al. (2010).

#### 2.2 Nitrogen pools

##### 2.2.1 Total and non-soluble nitrogen in soil

The pool of N bound to the soil matrix (the non-soluble soil N pool) was calculated by subtracting extractable and soil water N pools from the total soil N pool. The total soil N pool was determined from soil samples collected from different soil horizons in 1995. The samples were taken using steel cylinders (148 cm$^3$ volume, 6 cm diameter) from each morphologic soil horizon (L/H, eluvial E horizon, illuvial B horizon and parent material C horizon).
from the vertical face of 5 soil pits. The samples were air dried at 60°C, sieved through a 2 mm sieve, and ground before the analysis. The C and N contents of each sample were analyzed using an elemental CN analyzer (LECO, Leco Corporation, St. Joseph, MI, USA).

The N pool was calculated for each soil horizon and for each pit by using a horizon-specific average for each of the following: N concentration, horizon depth, soil density and stone fraction. The N pool for each soil horizon was calculated as the average of the N pools of the specific horizon in each pit. The total soil N pool was calculated as the sum of N pools of all horizons.

2.2.2 Extractable nitrogen in soil

The extractable pools of soil ammonium (NH$_4^+$-N), nitrate (NO$_3^-$-N) and organic nitrogen (N$_{org}$) were determined from samples taken from litter- and humus-layers and from mineral soil at 0–0.1 m and 0.1–0.3 m depths. The samples were collected with a Westman soil auger (Westman, 1995) 1 to 3 times per year (8 times in total) from 2006 to 2009. Fresh soil samples were extracted with 2 M KCl for 2 h, and the extracts were filtered through Whatman 40 filter papers and frozen until analysis. Dissolved NH$_4^+$, NO$_3^-$ and N$_{org}$ in the extracts were analyzed by flow-injection spectrometry, as described in Sect. 2.3.2. Nitrogen pools on soil particle surfaces were estimated by subtracting N pools in soil water in the uppermost 0.3 m depth from the measured values of extractable N pools.

2.2.3 Nitrogen in soil water

The pools of NH$_4^+$-N, NO$_3^-$-N and N$_{org}$ in soil water were calculated by measuring the concentrations in every soil layer, and multiplying the measured concentrations with soil water storage specific to each soil horizon. Layer specific soil water pool was calculated based on time-domain reflectometry measurements, as described in Ilvesniemi et al. (2010). The concentrations of NH$_4^+$-N, NO$_3^-$-N and N$_{org}$ were measured in soil water samples obtained with suction cup lysimeters. Suction cups were installed at 7 locations (pits) and at each location in every soil horizon. Samples were collected in weekly to fortnightly intervals during the periods when the soil was not frozen. At the time of sampling, a suction pump (∼400 mbar) was applied to the tubes connected with suction cups in different soil depths. Water was sampled when either the water volume reached a minimum of 250 mL, or after 7 to 34 h of collecting. During the summer months when the soil was relatively dry, even the 34-h collection did not provide sufficient amounts of water from all locations. To determine the ratio between nitrate and nitrite (NO$_3^-$ : NO$_2^-$) in soil water, we used data measured in 1997. Nitrate (NO$_3^-$) and nitrite (NO$_2^-$) concentrations were measured colorimetrically from the drainage flow water by a nitrate reduction tube with cadmium column (Dorich and Nelson, 1984). The water analysis is explained in Sect. 2.3.2.

2.2.4 Aboveground biomass nitrogen

To estimate N pools in aboveground biomass, the diameter at 1.3 m height and height of every tree in the catchment were measured. Regression functions described in Repola (2008, 2009) were used to model the dry weight of wood, stems, bark, needles, leaves, roots, alive branches, and dead branches. The dry weights of the biomass classes were then multiplied with the representative N concentrations, presented in Table 1. The data of coniferous needles used in the regression were collected in the autumn, when part of the needles were already shed (Repola, 2009). Therefore the model gives an underestimation of the maximum foliage pool. As follows, 60% of the annual needle litter fall was added to the estimation, as based on the litter fall measurements on the site. This 60% represents the amount of needle litter fall occurring during the autumn.

Annual biomass N pool change (ΔB; kg N ha$^{-1}$ yr$^{-1}$) calculations were based on the difference between the biomass classes from 2003 and 2008. We consider the model results for the increase of wood and bark biomass to be relatively reliable, but we consider the biomass change in branches and foliage to be only suggestive, because the needle mass is usually assumed not to increase after the canopy has been closed.

2.3 Nitrogen transport in water

2.3.1 Sampling and maintenance

Bulk deposition, throughfall and stemflow waters were collected in canisters, which were changed monthly during the winter and once a fortnight during the summer, or whenever they were getting full. All of the canisters were always changed at the same time. The canisters were washed with hot water and Deconex® laboratory cleaning detergent (Borer Chemie AG, Zuchwil, Switzerland), rinsed with tap water three times, and finally rinsed twice with deionized water. Throughfall collectors were cleaned daily using a brush, deionized water, and washcloth to remove needles, pollen and other dirt. No anti-microbial substances were used in the canisters or the throughfall collectors. The water collecting system is described in more detail by Ilvesniemi et al. (2010).

2.3.2 Laboratory water analyses

The water samples (precipitation, throughfall, stemflow, soil water and drainage flow) were analyzed as follows: (1) the amount of collected water was measured by weighing (1 kg ≈ 1 dm$^3$), (2) precipitation, throughfall and stemflow subsamples were each pooled into one sample (3) pH and electric conductivity were measured, (4) precipitation, throughfall, and stemflow samples were filtered with a vacuum-driven filtering system (Millipore) using 0.45 μm
membrane filters (Millipore), and (5) samples were bottled and stored at \(-17\,^\circ\text{C}\) until further analysis.

Ammonium (\(\text{NH}_4^+\)), nitrate (\(\text{NO}_3^-\)) and organic nitrogen (\(\text{N}_{\text{org}}\)) were measured from all the water samples and the soil extracts by flow-injection spectrometry at the Finnish Forest Research Institute, Vantaa Unit (modified ISO 11732:2005 (FIA), SFS-EN ISO 13395:1997: SFS-EN ISO 11905-1:1998 (FIA), respectively). Detection limits for \(\text{NH}_4^+,\) \(\text{NO}_3^-\), and \(\text{N}_{\text{org}}\) were 0.03, 0.001 and 0.1 mg dm\(^{-3}\), respectively. Organic N (\(\text{N}_{\text{org}}\)) was determined using total N concentrations in the samples as follows:

\[
[\text{N}_{\text{org}}] = [\text{N}_{\text{tot}}] - [\text{NH}_4^+ - \text{N}] - [\text{NO}_3^- - \text{N}].
\] (1)

The filtering of the soil extracts with Whatman 40 filter paper (8 \(\mu\)m) does not remove all the particulate N. Therefore, we recognize that the \(\text{N}_{\text{org}}\) includes both dissolved organic nitrogen and some particulate N.

### 2.3.3 Stemflow

Stemflow was measured from 2006 to 2009 during snow-free periods from 4 trees by directing stemflow water into insulated canisters. For that purpose cleaved silicon rubber tubes (diameter 25 mm) were attached around the trees. The N flux rate in separate chemical N forms (\(\text{NH}_4^+,\) \(\text{NO}_3^-,\) \(\text{N}_{\text{org}}\)) in stemflow (\(S_c\); mg N m\(^{-2}\) day\(^{-1}\)) in the forest was calculated as follows:

\[
S_c = \frac{1}{4} \sum_{i=1}^{4} \frac{A_{\text{bi}}}{A_{\text{ci}}} \frac{C_{\text{ci}} V_{\text{ci}}}{(A_{\text{C1}} + A_{\text{C2}})} t^{-1},
\] (2)

where “\(c\)” refers to the chemical forms of N, \(i\) refers to a measured tree, 4 is the number of measured trees, \(C_{\text{ci}}\) is the concentration of N (mg dm\(^{-3}\)) in the stem flow water in different chemical forms (\(\text{NO}_3^-,\) \(\text{NH}_4^+,\) \(\text{N}_{\text{org}}\)), \(V_{\text{ci}}\) is the volume of the collected stem flow water (dm\(^3\)), \(A_{\text{bi}}\) is the stem cross section area of the trees from where the stem flow water was measured, \(A_{\text{ci}}\) is the total stem cross section area of the trees in the catchments (m\(^2\); \(C1 + C2\), \(A_{\text{C1}}\) and \(A_{\text{C2}}\) are the areas of the two micro-catchment areas (m\(^2\)) described in Sect. 2.1, and \(t\) is the length of the collection period (in days).

### 2.3.4 Drainage flow

As described in Sect. 2.1, the study site is defined as the area inside two micro-catchments, and the outflow water is directed to the two weirs. The water flow through the weirs was measured automatically with a flow meter (Schlumberger Aquatic, Schlumberger Water Services, Paris, France), and when flow existed, water was sampled for chemical analysis in a daily basis from the outlet of the weirs. The daily sum of water flow was multiplied with the concentrations of different chemical forms of N (\(\text{NH}_4^+,\) \(\text{NO}_3^-\), \(\text{N}_{\text{org}}\)) to get the daily N fluxes separately. When the concentrations were under the detection limit, we used half of the detection limit as the measured value. We also calculated the lower and the upper values for the N flux in the drainage flow by assuming that the lower value was zero and that the upper value was the detection limit. The lower and upper values were used to calculate the uncertainty for the average flux. The drainage flow measurements are explained in more detail in Ilvesniemi et al. (2010).

### 2.4 Atmospheric N deposition

The total annual N deposition to the site was calculated as the sum of estimated wet and dry deposition. Wet deposition was estimated from the measured bulk deposition data, and the values for dry deposition were taken from Flechard et al. (2011).

#### 2.4.1 Bulk deposition

Different N components (\(\text{NO}_3^-,\) \(\text{NH}_4^+,\) \(\text{N}_{\text{org}}\)) in the bulk N deposition were sampled in a tower above the forest canopy using two rain water collectors made of polyethylene funnels (0.13 m\(^2\) in area; Plastex Oy, Lohja, Finland). In the winter, snowfall was collected into circular canisters (0.2 m\(^2\) in area). The canisters were changed monthly in winter and once a fortnight in summer, or whenever they were getting full. No anti-microbial substances were used in the canisters. The bulk deposition rate was calculated by multiplying measured concentrations of \(\text{NO}_3^-,\) \(\text{NH}_4^+\) and \(\text{N}_{\text{org}}\) by precipitation and dividing by collection time. The precipitation was measured optically with a DRD12 rain detector (Vaisala Oyj, Helsinki, Finland). The water collecting system is described in more detail by Ilvesniemi et al. (2010).

#### 2.4.2 Throughfall

Throughfall water was collected using seven rectangular rainwater collectors installed below the forest canopy at approximately 0.5 m height from soil surface. The collectors were made of stainless steel and were 4 m long and 0.1 m wide, with effective water collecting area \(A_t\) being 0.385 m\(^2\). Throughfall waters were collected in insulated canisters installed below the midpoint of each collector. During the winter, when precipitation was dominated by snowfall, the throughfall collectors were replaced with circular canisters (0.2 m\(^2\)).

Throughfall rates \((T_c\); mg N m\(^{-2}\) d\(^{-1}\)) were calculated as follows:

\[
T_c = \frac{c_{\text{tc}} V_t}{A_t t},
\] (3)

where subscript “\(c\)” refers to the concentration of N stored in different chemical forms (\(\text{NO}_3^-,\) \(\text{NH}_4^+\) or \(\text{N}_{\text{org}}\)), \(c_{\text{tc}}\) is the concentration of the compound in the water sample (mg dm\(^{-3}\)), \(V_t\) is the volume of the water sample (dm\(^3\)), \(A_t\) is area of the collector, and \(t\) is the length of the collection period (days).
2.4.3 Estimating wet deposition from bulk deposition measurement

The bulk deposition measurement gives an underestimation of the total (wet + dry) deposition, and an overestimation to the wet deposition. This is due to the fact that in the bulk deposition measurement, some but not all of the dry deposition is included. A simple model to fractionate the bulk deposition into wet and dry deposition was formulated based on the amount of precipitation and the period of time during which the dry deposition could have occurred. As the result, the model gives an estimate of wet nutrient deposition of the ecosystem. The model also gives an estimation of the dry deposition, but only of the deposition on the bulk deposition collector, not the deposition of the whole ecosystem. As a result of the model, we get that the wet deposition was 57% of the measured bulk deposition. To calculate the total wet deposition at the site, the amount of bulk deposition was multiplied by this number (57%). The model is described briefly in Korhonen et al. (2012).

2.4.4 Dry deposition

The mean of the dry deposition of four models presented in Flechard et al. (2011) for Hyytiälä were used as an estimate of the dry deposition. When this dry deposition data was used in conjunction with the measured bulk deposition data from this study, the modeled aerosol particle NH$_4^+$ and gaseous NH$_3$ deposition were coupled with the measured NH$_4^+$ deposition, and modeled aerosol particle NO$_3^-$ and gaseous NO$_2$ and HNO$_3$ deposition were coupled with the measured NO$_3^-$ deposition.

2.5 Gaseous emissions

2.5.1 Nitrous oxide (N$_2$O)

The fluxes of nitrous oxide (N$_2$O) were measured with one automatic and six manual static chambers. The automatic chamber and four of the manual chambers were located in the two catchment areas, whereas two manual chambers were located outside the catchment (Fig. 1). The automatic chamber was made of stainless steel ($0.40 \times 0.80 \times 0.32$ m: width $\times$ length $\times$ height), and was equipped with two fans and a thermocouple for chamber air temperature measurement. The chamber was automatically closed 1–4 times per day for 60 min. During each enclosure a minimum of 4 gas samples were withdrawn from the headspace by a custom-made autosampler (MaSa, Pohja-Metallityöpaja, Juupajoki, Finland). The manual chambers were made of stainless-steel
(0.29 × 0.40 × 0.24), and they were equipped with a fan and a sample port in the middle of the chamber (Pihlatie et al., 2007).

The manual chamber measurements were conducted on a weekly basis during summer months and monthly in the winter as described by Pihlatie et al. (2007). The concentrations of N₂O in the gas samples were analyzed by a gas chromatograph equipped with an electron capture detector. N₂O fluxes were calculated by linear regression method. Non-linearity of the concentration change over chamber closures was tested. Due to measuring N₂O fluxes close to the detection limit (see Pihlatie et al., 2007), we chose to use the linear regression method as the more robust calculation method for the N₂O fluxes.

### 2.5.2 Nitrogen oxide (NO)

Flux of NO from the soil was measured using three automatic dynamic flow-through chambers during a short campaign from 15 July to 30 October 2011. The chamber system consisted of three transparent chambers similar to the automatic N₂O chamber, and with fluorinated ethylene–propylene film as the transparent wall material. The operation of the chambers was automated; each chamber was closed for 15 min once every three hours. Sample air was drawn from the chambers at a rate of 4.1 dm³·min⁻¹ into a chemiluminescence analyser (TEI 42S, Thermo Environmental Instruments, Philadelphia, PA, USA). The measurement principle for the soil NO flux was similar to that of the shoot NO₃ flux described in Raivonen et al. (2003), except that an empty chamber was not used as a reference chamber. At the time of the sampling, compensation air from the above canopy atmosphere was directed into the chambers at a rate of 4.5 dm³·min⁻¹. Soil flux was calculated using a flux calculation method for flow through chambers as described in Kolari et al. (2012).

### 2.6 Internal cycling

#### 2.6.1 Litter fall

Litter fall was measured monthly from 2006 to 2010 using 20 circular litter collectors (0.2 m² each) installed systematically on two catchment areas (Fig. 1) as described in Ilvesniemi et al. (2009). The litter collectors were emptied once a month, dried at 60 °C for 24 h, and weighed. Dried litter was then separated into needles, leaves, bark, branches, seeds (including cones), and remaining material. Each compartment was weighed, ground, and pooled. Carbon and N concentrations were measured from the pooled samples by elemental CN analyzer (vario Max CN, Elementar Analysensysteme GmbH, Hanau, Germany). The N concentrations of litter fractions are presented in Table 2. Larger branch litter was collected into 20 frames (0.5 × 1.0 m) lying on the ground. The branches were collected once a year and treated similarly as the other litter. The N flux in litter fall (L; g N m⁻² yr⁻¹) is

\[
L = \frac{1}{20} \sum_{i=1}^{20} \left( m_{ci} C_c \right) \frac{1}{A_L t},
\]

where \( i \) refers to the number of the litter collectors or the branch frames, “c” refers to different biomass compartments, \( m_{ci} \) is the mass of collected litter compartment (g), \( C_c \) is N concentration (mg N g⁻¹), \( A_L \) is the area of the collector, and \( t \) is the length of collection period (in days).

#### 2.6.2 Nitrogen retranslocation and senescence

Based on Helmisaari (1992), retranslocation of N (\( R \); kg N ha⁻¹ yr⁻¹) was calculated as follows:

\[
R = \frac{1.49 m_b c_g - m_b c_b}{A_L t_r},
\]

where \( m_b \) is the mass of (brown) foliage litter collected by litter traps per year (g), \( c_b \) is the N concentration of that litter
(mg g\(^{-1}\)), \(c_g\) is N concentration of green needles (12 mg g\(^{-1}\); Palmroth and Hari, 2001). \(A_L\) is the area of the litter collector, \(t_r\) is the litter collection time (1 yr), 1.49 is the relative mass of a single green needle to a single brown needle, calculated as an average from the three plots from various years (\(n = 11\)) presented by Helmisaari (1992). This number was applied to all tree species.

Plant N loss during senescence (\(S\); kg N ha\(^{-1}\) yr\(^{-1}\)) was calculated as the sum of litter fall and retranslocation. For all biomass classes other than foliage, retranslocation was assumed to be negligible. We assumed that the pool of dead plant material attached to the trees does not change, and thus that the values for N loss in the litter fall represent the N loss in senescence.

### 2.7 Variables based on mass balance calculations

The total N balance of the ecosystem (\(\Delta N_s + \Delta N_b\); kg N ha\(^{-1}\) yr\(^{-1}\)) was calculated as follows:

\[
\Delta N_s + \Delta N_b = D_w + D_d - E_{N_2O} - E_{NO} - D_t,
\]

where \(\Delta N_s\) is the change of N pools in the soil, \(\Delta N_b\) is the change of N pool in aboveground biomass, \(D_w\) is the wet N deposition, \(D_d\) is the dry N deposition, \(E_{N_2O}\) and \(E_{NO}\) are the N losses in N\(_2\)O and NO emissions, respectively, and \(D_t\) is N loss in the drainage flow.

The amount of N used for growth (\(Y_i\); kg N ha\(^{-1}\) yr\(^{-1}\)) was calculated as follows:

\[
Y_i = \Delta B_i + S_i,
\]

where \(\Delta B_i\) is the change of N in biomass and \(S_i\) is the senescence in the biomass class, both measured in kg N ha\(^{-1}\) yr\(^{-1}\) and where subscript \(i\) refers to the aboveground biomass class (needles, leaves, branches, bark, wood). The total amount of N used by plants (\(B_{tot}\); kg N ha\(^{-1}\) yr\(^{-1}\)) was calculated as the sum of N used for each individual aboveground biomass class.

Nitrogen uptake by plants (\(U\); kg N ha\(^{-1}\) yr\(^{-1}\)) was calculated as follows:

\[
U = B_{tot} - R,
\]

where \(B_{tot}\) is N use by plants and \(R\) is the retranslocation of N.

Net release of N from the decomposition (\(R_d\); kg N ha\(^{-1}\) yr\(^{-1}\)) was calculated by assuming that the pool of plant-available-N is constant in a time scale of a couple of years as follows:

\[
R_d = U + E_{tot} + D_t - D_{tot},
\]

where \(U\) is N uptake by plants, \(E_{tot}\) is the gas emission of N (\(E_{N_2O} + E_{NO}\)), \(D_t\) is the drainage flow and \(D_{tot}\) is the total deposition (\(D_w + D_d\)), all in kg N ha\(^{-1}\) yr\(^{-1}\).

### Table 3. Nitrogen concentrations, nitrogen pools and carbon-to-nitrogen ratios in different physical soil horizons (O, A, B, C1 and C2).

<table>
<thead>
<tr>
<th>Horizon thickness (m)</th>
<th>O</th>
<th>A</th>
<th>B</th>
<th>C1</th>
<th>C2</th>
</tr>
</thead>
<tbody>
<tr>
<td>N concentration (mg g(^{-1}))</td>
<td>0.05</td>
<td>0.041</td>
<td>0.16</td>
<td>0.19</td>
<td>0.20</td>
</tr>
<tr>
<td>N pool (kg N ha(^{-1}) yr(^{-1}))</td>
<td>710</td>
<td>240</td>
<td>860</td>
<td>190</td>
<td>75</td>
</tr>
<tr>
<td>C : N</td>
<td>28</td>
<td>33</td>
<td>23</td>
<td>36</td>
<td>19</td>
</tr>
</tbody>
</table>

\(c = 1\) ha = 10 000 m\(^2\).

The change of the non-soluble soil N pool (\(\Delta N_{som}\); kg N ha\(^{-1}\) yr\(^{-1}\)) was calculated as follows:

\[
\Delta N_{som} = L - R_d,
\]

where \(L\) is N flux in the litter fall and \(R_d\) is the net release of N from the decomposition, both in kg N ha\(^{-1}\) yr\(^{-1}\).

### 3 Results

#### 3.1 Soil nitrogen pools

The total N pool in the soil was 2070 kg N ha\(^{-1}\) (1 ha\(^{-1}\) = 10 000 m\(^2\)). The vast majority of the soil N was bound to the soil matrix (2050 kg N ha\(^{-1}\)). The organic layer (litter and humus) and the uppermost 0.2 m mineral soil horizons (A and B horizons) contained 710 and 1100 kg N ha\(^{-1}\), respectively, which comprised 87% of the total soil N (Table 3). The highest N concentration in the soil was in the organic layer, 12.9 mg g\(^{-1}\) of soil. In the mineral soil the N concentration was on the order of 1 mg g\(^{-1}\) in the A and B horizons, and on the order of 0.1 mg g\(^{-1}\), in the C1 and C2 horizons. The calculated soil non-soluble N accumulation rate (\(\Delta N_{som}\)) was \(-1 \) kg N ha\(^{-1}\) yr\(^{-1}\), suggesting a slight decrease in the soil N pool. However, the estimation does not differ from 0, taking into account the error margin (\(\pm 0.8 \) kg N ha\(^{-1}\) yr\(^{-1}\)).

The extractable N pool in the organic layer and the topmost 0.30 m of the mineral soil (26.8 kg N ha\(^{-1}\)) was small compared to the total soil N pool. Of the extractable soil N almost all (98.9%) was in organic form (N\(_{org}\)), 26.5 kg N ha\(^{-1}\). The vast majority of the mineral N was ammonium-N (NH\(_4\)-N; 0.31 kg N ha\(^{-1}\)), and only a minor fraction was nitrate-N (NO\(_3\)-N; 2 g N ha\(^{-1}\); 0.6%). The nitrate-N estimation is relatively uncertain.

N pool in soil water was 0.70 kg N ha\(^{-1}\), and similar to the extractable N, the majority of it was in organic form (0.66 kg N ha\(^{-1}\)). Ammonia and NO\(_3\) pools in the soil water were approximately 30 and 3 g N ha\(^{-1}\), respectively. Nitrate concentrations were typically under the detection limit, and thus the extractable and especially soil water NO\(_3\) pool sizes are uncertain. A more reliable estimation was obtained from the extracted NO\(_3\) concentration, which includes both
NO$_3^-$-N in soil particle surfaces and in soil water. Based on the measurements in 1997, the median and mean ratios between nitrate and nitrite (NO$_3^-$: NO$_2^-$) in the drainage water measurements were 9.3 and 7.4, respectively.

### 3.2 Biomass nitrogen pools

Nitrogen stored in the aboveground tree biomass in 2008 was 210 kg N ha$^{-1}$, which was 9% of the total N in the ecosystem. Nitrogen was distributed quite evenly to foliage (77 kg N ha$^{-1}$), branches (58 kg N ha$^{-1}$), wood (49 kg N ha$^{-1}$) and bark (24 kg N ha$^{-1}$). Of the branch N pool, 12 kg N ha$^{-1}$ was estimated to be stored in dead branches.

The total N accumulation to aboveground biomass was 7.4 kg N ha$^{-1}$ yr$^{-1}$. The wood and bark biomass increase were 2.3 and 0.8 kg N ha$^{-1}$ yr$^{-1}$, respectively. The more uncertain estimates for foliage and branch biomass accumulation were 1.8 and 2.6 kg N ha$^{-1}$ yr$^{-1}$, respectively.

### 3.4 Drainage flow and gaseous emissions

Annual N flux from the ecosystem via drainage flow varied between 0.04 and 0.23 kg N ha$^{-1}$ yr$^{-1}$ and was on average 0.13 kg N ha$^{-1}$ yr$^{-1}$. The N flux in drainage flow was dominated by N$_{org}$, on average 0.12 kg N ha$^{-1}$ yr$^{-1}$. The average flux of mineral N in drainage flow was very low, 0.005 kg N ha$^{-1}$ yr$^{-1}$ and 0.002 kg N ha$^{-1}$ yr$^{-1}$ for NH$_4^+$ and NO$_3^-$, respectively. The uncertainty for the mineral N values is approximately ±50%, and for the organic N up to ±90%. The uncertainty is primarily caused by the fact that the fluxes were very small and most of the time the N concentrations were below the detection limit. Therefore, it is more likely that our estimate of the drainage flow is an overestimation than an underestimation.

Both N$_2$O and NO were emitted from the soil and NO$_2$ was deposited into the soil, however, the fluxes were very small. Annual cumulative soil N$_2$O emission averaged to 0.2 kg N ha$^{-1}$ yr$^{-1}$. Measuring NO emission and NO$_2$ deposition from/to the soil was challenging because of the small fluxes. During the campaign in the autumn period 2011, measured NO-N emission was around 0.01 kg N ha$^{-1}$ yr$^{-1}$ and NO$_2$-N deposition was even smaller.

### 3.6 Internal nitrogen cycling

#### 3.6.1 Litter fall

From 2006 to 2010, the amount of N flux in annual aboveground litter fall from trees varied from 14 to 22 kg N ha$^{-1}$ yr$^{-1}$, being on average 18 kg N ha$^{-1}$ yr$^{-1}$. Half of the N flux in the aboveground litter fall was in needles and leaves, 8.0 and 1.0 kg N ha$^{-1}$ yr$^{-1}$, respectively. Branches contributed about one fourth of the N in aboveground litter fall, 5.3 kg N ha$^{-1}$ yr$^{-1}$. Nitrogen flux in the litter fall of bark, reproductive matter and unidentified matter were 1.1, 0.35 and 2.1 kg N ha$^{-1}$ yr$^{-1}$, respectively.

#### 3.6.2 Senescence and retranslocation

Nitrogen retranslocation from needles and leaves was estimated to be 21 kg N ha$^{-1}$ yr$^{-1}$ and 2.8 kg N ha$^{-1}$ yr$^{-1}$, respectively. This was 73% of the initial amount of N in the
The error bar of the drainage flow is the standard error of mean annual drainage flow of years 2006–2010.

model input parameters are not included. The error bar of the gas emission is the standard error of mean of average flux between the chambers. The error bar of the dry deposition represents the standard deviation between the four models used in Flechard et al. (2011) and the uncertainty of estimating \( \text{N}_\text{org} \) in precipitation from bulk deposition measurement. The error bar of the wet deposition is based on the standard error of mean of the annual precipitation amount and on the uncertainty of estimating N concentration in precipitation from bulk deposition measurement. The error bar of the gas emission is the standard error of mean of average flux between the chambers. The error bar of the drainage flow is the standard error of mean annual drainage flow of years 2006–2010.

Fig. 2. Inputs (A) and outputs (B) of N in boreal Scots pine forest in Hyytiälä. Note the different y-scale in the images. The error bar of the wet deposition is based on standard error of mean of the annual precipitation amount and on the uncertainty of estimating N concentration in precipitation from bulk deposition measurement. The error bar of the dry deposition represents the standard deviation between the four models used in Flechard et al. (2011) and the uncertainty of estimating \( \text{N}_\text{org} \). Systematic uncertainty in the models and uncertainty in the model input parameters are not included. The error bar of the gas emission is the standard error of mean of average flux between the chambers. The error bar of the drainage flow is the standard error of mean annual drainage flow of years 2006–2010.

3.6.3 N use by plants

Estimated N use for growth was 50 kg N ha\(^{-1}\) yr\(^{-1}\) (Fig. 3). Nitrogen uptake and retranslocation were as important sources for the N use, 26 and 24 kg N ha\(^{-1}\) yr\(^{-1}\), respectively. Nitrogen uptake comprised 19 and 7 kg N ha\(^{-1}\) yr\(^{-1}\) of net N release from decomposition and deposition, respectively.

Most of the used N, 36 kg N ha\(^{-1}\) yr\(^{-1}\), was allocated to the foliage. The amount of N used to grow branches, wood and bark was 7.8, 2.3 and 1.9 kg N ha\(^{-1}\) yr\(^{-1}\), respectively. The amount of N used to grow cones, seeds and flowers was relatively low, 0.35 kg N ha\(^{-1}\) yr\(^{-1}\). A relatively large amount, 2.1 kg ha\(^{-1}\) yr\(^{-1}\), of unidentified litter fall was measured. An equivalent amount was interpreted to be used by trees to grow an unknown biomass fraction.

4 Discussion

4.1 Nitrogen balance and internal nitrogen cycling

Overall N cycling at Hyytiälä Scots pine forest is presented in Fig. 4. The outputs of N from the system are very small, and N is accumulating to the system at a rate of 7 kg N ha\(^{-1}\) yr\(^{-1}\). Internal cycling of N within the forest is a very important source of N for the plants in this N limited ecosystem. Nitrogen retranslocation and N uptake are equally important N sources. Most of the assimilated N originates from the decomposition of organic matter. The atmospheric N deposition was about one third of the total N uptake. This means that release during decomposition is the main origin of N for the plant uptake, but also that N deposition has clearly increased the total N uptake, boosting the plant growth and productivity. Internal cycling and pools of N at Hyytiälä were systematically slightly higher than those of a similar 35-yr-old Scots pine forest in Mekrijärvi, southeast Finland (Helmisaari, 1995). Overall, the results of these studies agree very well.

The amount of N released from decomposition annually is approximately the same as the amount of N released to the soil in litter fall. We hypothesize that a considerable part of the N released in decomposition originates from fresh litter, which naturally contains more easily decomposable fractions than old litter does. Therefore, we conclude that the N release in the decomposition is at least partly dependent on the amount of litter fall. As the atmospheric N deposition increases the plant growth, and thus also the litter fall, we argue further that this effect accumulates over time. Therefore, based on the N balance, the N deposition increases plant productivity in three ways: (1) it directly increases the availability of plant-available N, (2) it indirectly increases the availability of N by increasing the rate of retranslocation and the release of N from the decomposition, and (3) the indirect effect accumulates over time. In addition, it has long been known that N availability affects the leaf-to-fine-root ratio (Helmisaari et al., 2007; Ericsson, 1995), as hypothesized in the functional balance concept (Brouwer, 1962; Davidson, 1969). A low leaf-to-fine-root ratio reduces plant growth, because of the fact that when more carbon is allocated to the root system, less carbon is available for the foliage growth. The large maintenance costs of a large root system can be reduced by higher N availability, and successively...
large growth enhancements can be achieved by increasing N input (Saarssalmi and Mäkelänen, 2001; Hyvönen et al., 2008).

Over two thirds of the N available was used to grow new foliage. The residence time of foliage was 2.6 yr and thus most of the allocated N was cycling quickly. Nitrogen allocated to structural biomass (wood, branches, bark) cycled much more slowly, the residence times (pool size/annual senescence) being 12 and 21 yr for branches and bark, respectively. The N allocated to wood was immobilized for up to half of a millennium.

4.2 Nitrogen pools

We estimate that the soil N pool at Hyytiälä Scots pine forest is staying rather constant. Berg and Dise (2004) estimated that N has been accumulating in north-Scandinavian forest soils at a rate between 3.0 to 3.5 kg N ha\(^{-1}\) yr\(^{-1}\). This estimate is not different from our result, taking into account the uncertainty. It is worth noting that their estimation mostly considers preindustrial time and full succession of forests, whereas Helmisaari et al. (2007) reported a Scots pine fine root N concentration of 7.7 mg g\(^{-1}\). Using these values, we got that the N pool in coarse roots was 13 kg N ha\(^{-1}\) and in the fine root pool varied from 24 to 37 kg N ha\(^{-1}\). Kulmala et al. (2008) reported that the total aboveground forest floor biomass at the study site was 1240 kg ha\(^{-1}\) and the corresponding N concentrations presented by Mäkelänen (1974), we get an estimate of 14 kg N ha\(^{-1}\) in the aboveground forest floor at the SMEAR II station. As follows, the total biomass N in the ecosystem was approximately 265 kg N ha\(^{-1}\). Roots and aboveground ground vegetation contributed about 16 % and 5 % of the total biomass N pool, respectively.

Our estimations of biomass pools only consider the aboveground tree biomass. Based on the regression functions in Repola (2009), the coarse root (> 1 cm) biomass was 14 100 kg dry mass (DM) ha\(^{-1}\). A fine root biomass including fine roots of the ground vegetation was 4760 kg DM ha\(^{-1}\) as reported for the study site in Ilvesniemi et al. (2009). Mäkelänen (1974) reported coarse and fine root N contents of 0.92 mg g\(^{-1}\) and 4.79 mg g\(^{-1}\), respectively, for a 47-yr-old Scots pine stand near the SMEAR II station, whereas Helmisaari et al. (2007) reported a Scots pine fine root N concentration of 7.7 mg g\(^{-1}\). Using these values, we got that the N pool in coarse roots was 13 kg N ha\(^{-1}\) and in the fine root pool varied from 24 to 37 kg N ha\(^{-1}\). Kulmala et al. (2008) reported that the total aboveground forest floor biomass at the study site was 1240 kg ha\(^{-1}\). Using the plant species distribution reported by Kulmala et al. (2008) and the corresponding N concentrations presented by Mäkelänen (1974), we get an estimate of 14 kg N ha\(^{-1}\) in the aboveground forest floor at the SMEAR II station. As follows, the total biomass N in the ecosystem was approximately 265 kg N ha\(^{-1}\). Roots and aboveground ground vegetation contributed about 16 % and 5 % of the total biomass N pool, respectively.

Based on the biomass equations presented by Repola (2009, 2008), we estimated that the foliage and coarse root N pool was increasing. The forest was partly thinned in early 2002 (Vesala et al., 2005). We did not observe any trend in the needle litter fall in the time series from 2003 to 2012, which would support the foliage N pool growth, although there was an increasing trend in the leaf litter fall. However, before the thinning, from 1998 to 2001, the needle litter fall was 36 % (±11 %; standard error) higher than that after the thinning. Therefore, we assume that the foliage and branch mass are probably still recovering from the thinning, and
thus increasing. The increase in coarse root biomass given by biomass equations was 0.7 kg N ha\(^{-1}\) yr\(^{-1}\), but it is very difficult to estimate the validity of this value.

### 4.3 Inputs and outputs

The total losses of N from our study site in the form of N leaching or N-oxide emissions were approximately 5% of the N inputs, which is much smaller proportion than those measured in Central European forest ecosystems with high N deposition (e.g. Kreutzer et al., 2009). Kreutzer et al. (2009) showed that in an N-saturated spruce forest (Högwald) in Southern Germany, over 80% of the N deposition was lost in the form of NO\(_3^-\) leaching and N\(_2\)O and NO emissions.

At Hyytiälä, most of the N leaching was in the form of organic nitrogen (N\(_{\text{org}}\)). This is in line with results of Mustajärvi et al. (2008), who found that DON (N\(_{\text{org}}\) in our study) was the dominant N species in the percolation water of 16 pine and spruce forests in Finland. Mustajärvi et al. (2008) also found that the runoff of DON was larger than the input of DON into the forest canopy via atmospheric deposition. We found that the input of N\(_{\text{org}}\) into the forest was higher than the output in the runoff, which may be due to the fact that the measurement sites and the methods were different. Overall, our findings show that N\(_{\text{org}}\) is an important component of the N cycling in boreal forests.

N\(_2\) fixation and N\(_2\) emissions are two unknowns in the N balance of the Hyytiälä forest site. Symbiotic N\(_2\) fixing bacteria in the root system of alder and birch trees (Rönkkö et al., 1993; Smolander, 1990), and in association with common feather mosses (DeLuca et al., 2002, 2008; Zackrisson et al., 2004), have a potential to bring significant amounts of N into the boreal forest floor. However, due to the small coverage of alder and birch trees at our measurement site, we estimate that their role in bringing N into the forest is minimal. Also, based on our laboratory measurements of N\(_2\) fixation by forest floor mosses at the SMEAR II stand (unpublished data), we estimate that moss-related N\(_2\) fixation at Hyytiälä Scots pine forest is at the lower range of that (0.1 to 4 kg N ha\(^{-1}\) yr\(^{-1}\)) reported for boreal forests by DeLuca et al. (2008, 2002) and Zackrisson et al. (2009). Personal communication with Maija Salemaa (Finnish Forest Research Institute) also supports the findings that N fixation by forest floor mosses in southern Finland is much smaller than that of the forest floor mosses in northern Finland.

Measuring N\(_2\) emission in the field is currently not possible, as the exchange of N\(_2\) cannot be resolved from the high background concentration of N\(_2\) in the air. There is no field measurement data available on N\(_2\) emissions of boreal ecosystems, whereas laboratory measurements indicate high N\(_2\) emissions (10–120 kg N ha\(^{-1}\) yr\(^{-1}\)) from temperate forest soils (Dannenmann et al., 2008; Butterbach-Bahl et al., 2002). Based on these results, we can expect that N\(_2\) is also emitted from the soil at our measurement site; however, the rates remain unresolved. If the rates of N\(_2\) emissions are significant, they may reduce the estimated N accumulation rates in the forest, which underlines the importance of quantifying these N exchange rates. Using a N\(_2\)O:N\(_2\) emission ratio of 0.15 for N saturated Norway spruce forest (Butterbach-Bahl et al., 2002), we estimate that the N\(_2\) emission is on the order of 1 kg N ha\(^{-1}\) yr\(^{-1}\).

### 5 Conclusions

The main inputs, outputs, internal cycling and pools of N in a Scots pine forest at the SMEAR II station were quantified. Our measurements show that N is accumulating in this Scots pine forest at a rate of 7 kg N ha\(^{-1}\) yr\(^{-1}\). Most of the N accumulates to the aboveground biomass, whereas the soil N pool is close to a steady state. The largest external N input into the forest was atmospheric deposition. Outputs from the system were very small and emissions to the atmosphere in the form of N\(_2\)O and NO were higher than the N flow in drainage in the form of N\(_{\text{org}}\), NH\(_4^+\) or NO\(_3^-\). High uncertainties remain in the quantification of the input via N\(_2\) fixation and especially in the output via N\(_2\) emissions from the soil.

Boreal forests are considered to be N limited; though these ecosystems are not deprived of N, the large organic N pool in the soil is not directly available for plant uptake. We estimated that the release of N from litter decomposition and retranslocation are the main sources of N for the plants. Annually, the plant uptake rate of N originating from the decomposition is at least 18 kg N ha\(^{-1}\) yr\(^{-1}\). The fraction of assimilated N originating directly from the atmospheric deposition was up to 30%. The main source of N for plant use is internal cycling, demonstrated by that half of the N used for growth originated from retranslocation.

Organic N deposition in the studied forest was an important component of the N balance, and N losses in drainage flow were mostly in the form of organic N. The largest output of N from the system was N\(_2\)O emission, while NO emission was extremely small.

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