Biogeosciences Discussions

Nitrogen balance of a boreal Scots pine forest

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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. The most important components of N pools and fluxes were measured in a boreal Scots pine stand in Hyytiälä, Southern Finland. The measurement 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 micro-catchments. N was accumulating to the system with 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 input in deposition. Total outputs were $0.4 \, kg \, N \, ha^{-1} \, yr^{-1}$, the most important outputs being N_2O emission to the atmosphere and organic N flux in drainage flow. Nitrogen uptake and retranslocation were as important sources of N for plant growth. Most of the uptaken 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.

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, N fixation and fertilization, outputs include N losses in gaseous emissions and drainage flow, and 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 (Mälkönen, 1974). The increased N deposition in turn affects the whole N cycling in forest ecosystems.

In boreal forests, the inputs of N via atmospheric deposition and N_2 fixation, are relatively small, at maximum around $10\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$ (Flechard et al., 2011; Syri et al., 2004; Mustajärvi et al., 2008) and $0.5–3.5\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$ (DeLuca et al., 2002, 2008; Zackrisson et al., 2004), respectively. In these systems, the N outputs are reported to be small, both ammonium (NH_4^+-N), nitrate (NO_3^--N) and dissolved organic nitrogen (DON) via leaching into groundwater (Kubin, 1998), or through volatilization of oxidized nitrogen (N_2O, NO_x) into the atmosphere (Pilegaard et al., 2006; Pihlatie et al., 2007; Maljanen et al., 2010). 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 Dise, 2004; Hattenschwiler and Vitousek, 2000) and biomass of the growing trees.

According to the current knowledge, plants can take N either in mineral (NH_4^+ or NO_3^-) or in 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, the availability of plant-available-N is less than the consumption potential of the plants, because forests in general benefit from N fertilization (Saarsalmi and Mälkönen, 2001). In boreal upland forests, this results from the cool climate and the chemical composition of soil organic matter. Plants can re-use 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 recognized by several studies, the role and magnitude of organic N inputs and outputs to 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 onto ecosystems (Neff et al., 2002), and up to 80 % of the total N lost as runoff (Mustajärvi et al., 2008). The uptake of N from soil to trees directly as amino acids seems evident (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 during the years from 2006 to 2010. We further calculate the individual sources of N for the plants, and 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) and (2010), respectively.

2 Materials and methods

2.1 Measurement site

Measurements were conducted at a Scots pine stand at 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 during 1971–2000 were 3.3 °C and 713 mm, respectively (Drebs et al., 2002). The stand is even-aged forest and it is regenerated by sowing after clear-cut, prescribed burning and soil preparation in 1962. The measurement station was established in 1995. The stand has been partly thinned in January–March 2002 (Vesala et al., 2005).

The dominant tree species is Scots pine, covering 93 % of the stem basal area in the mini-catchments. 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 understorey are 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*, *Vaccinium vitis-idaea* and in the ground layer the mosses *Dicranum polysetum* and *Pleurozium schreberi* (Kulmala et al., 2008).

2.2 Nitrogen pools

2.2.1 Total and non-soluble nitrogen in soil

The pool of N bound to soil matrix (the non-soluble soil N pool) was calculated by subtracting extractable and soil water N pools from the total soil N pool.

Soil total 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/F/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 2 mm sieve and ground before the analysis. The C- and N-contents of each sample were analyzed using elemental CN-analyzer (LECO, Leco Corporation, St. Joseph, MI, USA).

The N storage was calculated for each layer and for each pit by using and layer-specific average N concentration, layer depth, soil density and stone fraction. The total soil N pool was calculated as the sum of N pools of all layers.

2.2.2 Extractable nitrogen in soil

Soil extractable nitrogen pool of 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 Westman soil auger (Westman, 1995) 1 to 3 times per year (8 times in total) during 2006–2009. Fresh soil samples were extracted with 2 M KCl for 2 h, 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 in soil particle surfaces were estimated by subtracting N pools in soil water in uppermost 0.3 m depth from the measured values of extractable N pools.

2.2.3 Nitrogen in soil water

The pool of NH_4^+-N, NO_3^--N and N_{org} in soil water was calculated by measuring the concentrations in every soil layer, and multiplying measured concentrations with soil horizon specific soil water storage. Layer specific soil water storage was calculated based on time-domain-reflectometry (TDR) 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 weekly to fortnight 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 as long as an amount of 250 ml at minimum was reached 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 amount 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 "Laboratory water analyses".

2.2.4 Aboveground biomass nitrogen

To estimate N pools in aboveground biomass, the diameter and height of every tree in the catchment was measured at 1.3 m height. Regression functions described in

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Repola (2008, 2009) were used to model the dry weight of wood, stem bark, needles, leaves and alive and dead branches. The dry weights of the biomass classes were then multiplied with the representative N concentration, presented in Table 1. The data of coniferous needles used in the regression were collected in autumn, when part of the needles were already shed (Repola, 2009). Therefore the model gives an underestimation for the maximum foliage pool. 60 % of the annual needle litter fall was added to the estimation, as based on litter fall measurements on site, this presents the amount of needle litter fall occurring during the autumn.

Annual biomass N pool change (ΔB; $\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$) calculations were based on the difference in the biomass classes from 2003 and 2008. We consider the model results of wood and stem bark biomass increases to be relatively reliable, but the biomass change in branches and foliage only indicative.

2.3 Nitrogen transport in water

2.3.1 Sampling and maintenance

Bulk deposition, throughfall and stemflow waters were collected to canisters, which were changed monthly in winter and once a fortnight in summer, or whenever they were getting full. All 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), and rinsed with tap water three times and finally twice with deionized water. Throughfall collectors were cleaned daily from needles, pollen and other dirt with brush, deionized water and wash cloth. No anti-microbial substances were used in the canisters. 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 sub-samples were pooled separately each into one sample (3) pH and electric conductivity were measured, (4) precipitation, troughfall, stemflow samples were filtered with a vacuum driven filtering system (Millipore) using 0.45 μm membrane filters (Millipore) (5) samples were bottled and stored in −17 °C until further analysis.

Ammonium (NH_4^+), nitrate (NO_3^-) and organic nitrogen (N_{tot}) 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 11 732:2005 (FIA), SFS-EN ISO 13 395:1997: SFS-EN ISO 11 905-1:1998 (FIA), respectively). Detection limits for NH_4^+-N, NO_3^--N, and N_{tot} were 0.03, 0.001 and 0.1 mg dm^{-3}, respectively. Organic N (N_{org}) was determined using total N concentrations in the samples as follows

$$[N_{org}] = [N_{tot}] − [NH_4^+\text{-N}] − [NO_3^-\text{-N}]. \tag{1}$$

The filtering of the soil extracts with Whatman 40 filter paper (8 μm) does not remove all the particulate N. Therefore, we recognize that the N_{org} includes both dissolved organic nitrogen (DON) 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 (NH_4^+-N, NO_3^--N, N_{org}) in stemflow (S_c; mg N m^{-2} day^{-1}) in

the forest was calculated as follows

$$S_c = \frac{1}{4} \sum_{i=1}^{4} c_{ci} V_{ci} \frac{A_{b\,tot}}{A_{b\,i} \cdot t \cdot (889 + 301)\,\mathrm{m}^2}, \tag{2}$$

where c refers to chemical forms of N i refers to a measured tree, 4 is the number measured of trees, C_c is the concentration of N ($\mathrm{mg\,dm}^{-3}$) in stem flow water in different chemical forms ($\mathrm{NO_3^-}$-N, $\mathrm{NH_4^+}$-N, $\mathrm{N_{org}}$), V_c is the volume of collected stem flow water, $A_{b\,i}$ is the basal area of the trees from where the stem flow was measured, $A_{b\,tot}$ the total basal area of the trees in the catchment and t is the length of collection period (in days).

2.3.4 Drainage flow

The study area consisting of two mini-catchments (C1, 889 m^2 and C2, 301 m^2) receive water only from precipitation since they locate on the top of a hill (Fig. 1). The soil is Haplic podzol on glacial till (FAO-UNESCO-ISRIC, 1988), overlaying homogeneous granite bedrock at an average of 0.6 m depth. The lowest soil layer on the bedrock has a high silt fraction rendering the watershed water tight and allowing the closing of site water balance. The soil penetrating radar measurements during the establishment of the site in 1994 showed that there were no major vertical cracks on the bedrock and the water flows in the soil along the direction of the slope and there outflow occurs only via the outlets in the weirs as reported in Ilvesniemi et al. (2010).

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 on a daily basis from the outlet of the weir. The daily sum of water flow was multiplied with the concentrations of different chemical forms of N ($\mathrm{NH_4^+}$-N, $\mathrm{NO_3^-}$-N, $\mathrm{N_{org}}$) to get the daily N fluxes separately. When the concentrations were under the detection limit, we used half of the detection limit as measured value. We also calculated the lower and upper values for the N flux in

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drainage flow by assuming that these concentrations were either zero, or the detection limit, respectively. The lower and upper values were used to calculate 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 dry deposition was modeled in Flechard et al. (2011).

2.4.1 Bulk deposition

Different N components (NO_3^-, NH_4^+, N_{org}) in the bulk N deposition above the forest canopy 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, snow fall 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 NO_3^-, NH_4^+ and N_{org} with precipitation and dividing with collection time. The precipitation was measured optically by 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, effective water collecting area (A_t) being $0.385\,m^2$. Throughfall waters were collected into insulated canisters installed below the mid-point of each collector. In winter, when precipitation

was dominated by snowfall, the throughfall collectors were replaced by circular canisters ($0.2\,\text{m}^2$).

Throughfall rates (T_c; $\text{mg}\,\text{N}\,\text{m}^{-2}\,\text{day}^{-1}$) were calculated as follows

$$T_c = \frac{c_{tc}V_t}{A_t t}, \tag{3}$$

where subscript c refers to the concentration of N stored in different chemical forms (NO_3^-, NH_4^+ or N_{org}), c_{tc} is concentration of the compound in water sample ($\text{mg}\,\text{dm}^{-3}$), V_t is the volume of the water sample (dm^3), A_t is area of the collector and t is length of the collection period (days).

2.4.3 Estimating wet deposition from bulk deposition measurement

The bulk deposition measurement gives an underestimation of total (wet + dry) deposition, and 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 that 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 dry deposition, but only that depositing on the bulk deposition collector, not of the whole ecosystem. As a result of the model we got that wet deposition was 57 % of the bulk deposition measurement, and to calculate wet deposition, bulk deposition amount was multiplied with this number. The model is described briefly in (Korhonen et al., 2011).

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 dry deposition. When this dry deposition data was used in conjunction with the measured bulk deposition data from this study, the

modelled 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_2O)

The fluxes of nitrous oxide (N_2O) were measured with one automatic and six manual static chambers. The automatic chamber and four of the manual chambers were placed in the two catchment areas, whereas two manual chambers located outside the catchment (Fig. 1). The automatic chamber was made of stainless-steel ($0.40 \times 0.80 \times 0.32$ m: width × length × height), it 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 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 N_2O in the gas samples were analyzed by a gas chromatograph equipped with an electron capture detector. N_2O fluxes were calculated by linear regression method. Non-linearity of the concentration change over chamber closures was tested. Due to measuring N_2O 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 N_2O fluxes.

2.5.2 Nitrogen oxide (NO)

Flux of NO from the soil was measured by three automatic dynamic flow-through chambers during a short campaign in 15 July–30 October 2011. The chamber-system

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consisted of three transparent chambers similar as the automatic N_2O chamber, and fluorinated ethylene–propylene (FEP) 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 $4.1\,dm^3\,min^{-1}$ to a chemilumiscence analyser (TEI 42S, Thermo Environmental Instruments, Philadelphia, PA, USA). At the time of sampling, compensation air from the above canopy atmosphere was directed into the chambers at a rate of $4.5\,dm^3\,min^{-1}$.

2.6 Internal cycling

2.6.1 Litter fall

Litter fall was measured monthly from 2006 to 2010 using 20 circular litter collectors (à $0.2\,m^2$) installed systematically on the two catchment areas (Fig. 1) as described in Ilvesniemi et al. (2009). The litter collectors were emptied once a month, the litter was dried in 60 °C for 24 h and weighed. Dried litter was then separated into needles, leaves, bark, branches, seeds (including cones) and the rest. Each compartment was weighed, ground and pooled. Carbon and N concentrations were measured from the pooled samples by elemental CN-analyzer (LECO, Leco Corporation, St. Joseph, MI, USA). The N concentrations of litter fractions are presented in Table 2. Litter of larger branch litter was collected into 20 frames ($0.5 \times 1.0\,m$) 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; $gN\,m^{-2}\,yr^{-1}$).

$$L = \frac{1}{20} \sum_{i=1}^{20} (m_{ci} c_c) \frac{1}{A_L t}, \tag{4}$$

where i refers to the number of litter collector or branch frame, c refers to different biomass compartments, m_{ci} is the mass of collected litter compartment (g), C_c is N concentration ($mgN\,g^{-1}$), A_L is the area of the collector and t is 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

$$R = \frac{1.49 m_b c_g - m_b c_b}{A_L t_r}, \tag{5}$$

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 N of green needles ($12\,mg\,g^{-1}$; Palmroth and Hari, 2001), A_L is the area of 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 in senescence (S; $kg\,N\,ha^{-1}\,yr^{-1}$) was calculated as the sum of litter fall and retranslocation. For all other biomass classes 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 the values for N loss in 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,

$$\Delta N_s + \Delta N_b = D_w + D_d - E_{N_2O} - E_{NO} - D_f, \tag{6}$$

where ΔN_s is the change of N pools in the soil, ΔN_b is the change of N pool in above-ground biomass, D_w is wet N deposition, D_d is dry N deposition, E_{N_2O} and E_{NO} are N losses in N_2O and NO emissions, respectively, and D_f is N loss in drainage flow.

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

$$Y_i = \Delta B_i + S_i, \tag{7}$$

where ΔB_i is the change of N in biomass and S_i is the senescence in the biomass class, both in $kg\,N\,ha^{-1}\,yr^{-1}$ and subscript i refers to aboveground biomass class (nee-
dles, leaves, branches, bark, wood).

The total amount of N used by plants (Y_{tot}; $kg\,N\,ha^{-1}\,yr^{-1}$) was calculated as follows

$$Y_{tot} = \sum_{i=1}^{5} Y_i, \tag{8}$$

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

$$U = B_{tot} - R, \tag{9}$$

where B_{tot} is N use by plants and R is retranslocation of N.

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

$$R_d = U + E_{tot} + D_f - D_{tot}, \tag{10}$$

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

The change of soil non-soluble soil N pool (ΔN_{som}; $kg\,N\,ha^{-1}\,yr^{-1}$) was calculated as follows,

$$\Delta N_{SOM} = L - R_d, \tag{11}$$

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

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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}). Vast majority of the soil N was bound to soil matrix (2050 kg N ha^{-1}). Organic layer (litter and humus) and the uppermost 0.2 m mineral soil (A and B layers) 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 in the order of 1 mg g^{-1} in A- and B-horizons, and in the order of 0.1 mg g^{-1}, in C1- and C2-horizons. The calculated soil non-soluble N accumulation rate (ΔN_{som}) was −1.2 kg N ha^{-1} yr^{-1}, suggesting decrease in soil N pool. However, the estimation is not different from 0, when taking the error margin into account.

Extractable N pool in 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. Of this N almost all (98.9 %) was in organic form (N_{org}), 26.5 kg N ha^{-1}. Vast majority of the mineral N was ammonium-N (NH$_4^+$-N; 0.31 kg N ha^{-1}; 99.4 %), 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, majority of it was in organic form (0.66 kg N ha^{-1}). Ammonia and NO$_3^-$ pools in 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 is 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 ratio between nitrate and nitrite (NO$_3^-$: NO$_2^-$) in drainage water measurement was 9.3 and 7.4, respectively.

3.2 Biomass nitrogen pools

Nitrogen stored in aboveground tree biomass in 2008 was $210 \, \mathrm{kg\,N\,ha^{-1}}$, which is 9 % of the total N in the ecosystem. Nitrogen was distributed quite evenly to foliage ($73 \, \mathrm{kg\,N\,ha^{-1}}$), branches ($58 \, \mathrm{kg\,N\,ha^{-1}}$), wood ($49 \, \mathrm{kg\,N\,ha^{-1}}$) and bark ($24 \, \mathrm{kg\,N\,ha^{-1}}$). $12 \, \mathrm{kg\,N\,ha^{-1}}$ of the N pool stored in branches were estimated to be in dead branches.

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

3.3 Atmospheric N deposition and throughfall

Most of the total deposition ($7.4 \, \mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$) occurred in the form of dry deposition ($4.6 \, \mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$). Most of the deposition was in mineral form, but organic deposition contributed over one fourth of the total deposition. During years 2006–2010 the annual measured bulk N deposition varied from 4.0 to $6.3 \, \mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$, the mean bulk N deposition being $4.9 \, \mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$. The distribution of the deposition is described in Table 4.

The measured throughfall of N was $2.9 \, \mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$, which consisted mostly of N_{org} and NO_3^--N, 1.4 and $1.1 \, \mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$, respectively. The measured N flux in stemflow, $0.1 \, \mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$, was very low compared to throughfall, and consisted mainly of N_{org}. The measured concentrations of NH_4^+, NO_3^- were 25 % to 90 % higher in bulk deposition than in throughfall, but the concentration of N_{org} was on average 33 % higher in throughfall than in bulk deposition. However, the measured throughfall flux was on average lower than the measured bulk deposition for mineral N and N_{org}.

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$_{\mathrm{org}}$, 0.12 kg N ha^{-1} yr^{-1}. Amount 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 $\pm 50\,\%$, and for the organic N up to $\pm 90\,\%$. 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. 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 0.3 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.

3.5 Nitrogen balance

The inputs to the system were one order of magnitude higher than the outputs (Fig. 2). The total N accumulation was 7 kg N ha^{-1} yr^{-1}. Dry deposition was higher than wet deposition, but they were in the same order of magnitude. Approximately three fourths of the N lost from the system was in form of gaseous N$_2$O-N emissions and one third as N$_{\mathrm{org}}$ in drainage flow. Nitrous oxide (N$_2$O) emission to N deposition ratio was 0.038 and N$_2$O : NO emission ratio was approximately 25.

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3.6 Internal nitrogen cycling

3.6.1 Litter fall

During the years 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 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 foliage. Nitrogen retranslocation was higher than the N flux in the aboveground litter fall and approximately.

Nitrogen flux in litter fall of branches, bark, cones and unidentified litter was assumed to present the N loss in senescence. The senescence of needles and leaves was estimated to be 30, and 3.9 kg N ha^{-1} yr^{-1}, respectively. The total senescence was 43 kg N ha^{-1} yr^{-1}, which is approximately one fifth of the aboveground biomass N pool (210 kg N ha^{-1}).

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 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 to branches, wood and bark were 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, $0.35\,kg\,N\,ha^{-1}\,yr^{-1}$. Relatively large amount, $2.4\,kg\,ha^{-1}\,yr^{-1}$, of unidentified litter fall was measured, and this was interpreted as to grow the unidentified 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 the rate of $7\,kg\,N\,ha^{-1}\,yr^{-1}$. Internal cycling of N within the forest is 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 uptaken N originates from decomposition of organic matter. The atmospheric N deposition was about one third of the total N uptake. This means that the release from the 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. We did not measure N-fixation in this study, but based on literature it is the third-most important source of N for plants, even if it was as high as $4\,kg\,N\,ha^{-1}\,yr^{-1}$ as reported by DeLuca et al. (2008).

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. Therefore, we conclude that N release in decomposition is at least partly dependent on the amount of litter fall. As the atmospheric N deposition increases the plant growth and thus also litter fall, we argue further that this effect accumulates over time. Therefore, based on N balance, 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 retranslocation rate and release rate of N from decomposition, and (3) the indirect effect

accumulates over time. In addition, it is long time 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, since when more carbon is allocated to the root system less carbon is available for the foliage growth. Large maintenance costs of large root system can be reduced by higher N availability and successively large growth enhancements can be achieved with increasing N input (Saarsalmi and Mälkönen, 2001; Hyvönen et al., 2008).

Internal cycling and pools of N at Hyytiälä were systematically slightly higher than those for a similar 35-yr-old Scots pine forest in Mekrijärvi, Southeast Finland (Helmisaari, 1995). The only major difference was in the annual aboveground litter fall, which in our study was approximately as high as the as annual retranslocation, but in study of Helmisaari (1995) annual litter fall was only half of the annual retranslocation.

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 fast. Nitrogen allocated to structural biomass (wood, branches, bark) cycled much slower, the residence time (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 millennia.

4.2 Nitrogen pools

Berg and Dise (2004) estimated that N has been accumulating to north-Scandinavian forest soils at the rate between 3.0 to 3.5 kg N ha^{-1} yr^{-1}, but we estimate that soil N pool is slightly decreasing, though the estimation is not significantly different from zero. The study of Berg and Dise (2004) concerns longer time-scale (hundreds of years) and different period than our study. The difference between the accumulation rates can be attributed to this because, the N deposition rates are higher in the period of this study, and because our study only covers a small part of the succession of boreal forests. In this study practically all of the accumulating N is assimilated to biomass, while the soil N pool seems to remain constant. This is contrary to the results of Berg and Dise (2004),

who show N accumulation to soil. However, it is noteworthy that our estimation is for a young forest, in which biomass accumulation is known to be strong. In old forests biomass net accumulation is slower, because the biomass cannot increase indefinitely. This can be observed as increased senescence, and thus increased flux from living
5 biomass to soil. Although N accumulation to soil does not necessary occur in time-scales of years, but it happens in millennial scale.

Our estimations of biomass pools only consider aboveground tree biomass. Based on 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 ground veg-
10 etation was 4760 kg DM ha^{-1} as reported for the study site by Ilvesniemi et al. (2009). Coarse and fine root N contents of 0.92 mg g^{-1} and 4.79 mg g^{-1}, respectively were reported for a 47-yr old Scots pine stand near the SMEAR II station by Mälkönen (1974). Helmisaari et al. (2007) reported Scots pine fine root N concentration of 7.7 mg g^{-1}. Using these values, we get that N pool in coarse roots was 13 and in fine root pool was
15 from 24 to 37 kg N ha^{-1}. Kulmala et al. (2008) reported total ground vegetation distribution by species or by plant groups at the study site, total above ground vegetation biomass being 1240 kg ha^{-1} at the study site. Using this plant species distribution and respectable N concentrations presented by Mälkönen (1974), we get an estimate of 14 kg N ha^{-1} in the aboveground ground vegetation. As follows the total biomass N in
20 the ecosystem is approximately 265 kg N ha^{-1}. Roots and aboveground ground vegetation contribute about 16 % and 5 % of the total biomass N pool, respectively.

Based on the biomass equations presented by Repola (2009, 2008), we estimated that foliage N pool was increasing. The forest was partly thinned in early 2002 (Vesala et al., 2005). We did not observe any trend in needle litter fall in time series from 2003
25 to 2012 that would support the foliage N pool growth, although there was an increasing trend in leaf litter fall. However, before the thinning, from 1998 to 2001, the needle litter fall was 36 % ($\pm$11 %; standard error) higher than after the thinning. Therefore, we assume that the foliage and branch mass probably is still recovering from the thinning and thus increasing.

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 inputs, which is much smaller than those in Central European forest ecosystems with high N deposition (e.g. Kreutzer et al., 2009). Kreutzer et al. (2009) showed that in N-saturated spruce forest (Höglwald) in Southern Germany in total over 80 % of the N deposition was lost in form of NO_3^- leaching and N_2O and NO emissions.

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

N_2 fixation and N_2 emissions are two unknowns in the N balance of 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 boreal forest floor. However, due to the small coverage of Alder and Birch trees in our measurement site, we estimate that their role in bringing N into the forest is minimal. Also, based on our measurements of N_2 fixation by forest floor mosses at SMEAR II stand (unpublished data), we estimate that moss-related N_2 fixation is negligible at Hyytiälä measurement site.

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.

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 into this Scots pine forest at a rate of $7\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$, which coincides with the N accumulation rate to aboveground biomass. This means that soil N pool was 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_2O and NO were higher than the N flow in drainage in the form of N_{org}, NH_4^+ or NO_3^-. Uncertainties remain in the estimation of input via N_2 fixation and output via N_2 emissions from the soil.

Although boreal forests are considered as N limited, these ecosystems are not deprived of N as such, but 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 plants. Annually, the plant uptake rate of N originating from decomposition is at least $18\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$. The fraction of uptaken N originating directly from the atmospheric deposition was up to 30 %. Internal nutrient cycling of N plays very important role, as half of the N used in 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_2O emission, while NO emission was extremely small.

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Table 1. Nitrogen concentrations of aboveground biomass classes used for biomass nitrogen pool calculations. The wood concentration was measured in Juupajoki, near the measurement site and Norway spruce needles concentration was a typical value for Norway spruce in Finland.

Biomass class	N (mg g^{-1})	Description	Reference
Wood	0.72	literature	Mälkönen, 1974
Scots pine needles	12	measured on site	Palmroth and Hari, 2001
Norway spruce needles	11	literature	Braekke et al., 1998
Broadleaved species	24	literature	Berg and McClaugherty, 2003
Bark	4.2	measured on site	Litter trap data (this study)
Branches	1.3	measured on site	Litter trap data (this study)

Table 2. Content of nitrogen in annual litter fall, nitrogen contents and carbon-to-nitrogen-ratios in different litter fractions for years from 2006 to 2008. Cones represent all material distinguished as reproductive material but mostly consists of cones. Uncertainty is standard error of the annual means.

	needles	leaves	large branches	small branches	bark	cones	other	total
C $\mathrm{mg\,g^{-1}}$	512 ± 3	486 ± 4	499 ± 0.2	494 ± 2	492 ± 6	471 ± 3	503 ± 3	502 ± 3
N $\mathrm{mg\,g^{-1}}$	4.9 ± 0.3	8.8 ± 0.7	4.7 ± 0.06	4.4 ± 0.3	3.7 ± 0.17	2.1 ± 0.2	12 ± 0.6	5.1 ± 0.3
C : N	104 ± 7	55 ± 5	105 ± 1	112.6 ± 7	134 ± 6	221 ± 18	41 ± 2	99 ± 6

Table 3. Nitrogen concentrations, nitrogen pools and carbon to nitrogen ratios in different physical soil horizons. The average depth of the mineral soil is 0.59 m.

	O	A	B	C1	C2
Horizon thickness (m)	0.05	0.041	0.16	0.19	0.20
N concentration (mg g^{-1})	13	1.2	1.0	0.17	0.053
N storage (kg N ha^{-1})*	710	240	860	190	75
C : N	28	33	23	36	19

* $1\,\text{ha} = 10\,000\,\text{m}^2$.

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Table 4. The measured atmospheric annual bulk N deposition, estimated annual wet N deposition and modeled annual dry N deposition (all in $kg\,N\,ha^{-1}\,yr^{-1}$).

	NH_4^+	NO_3^-	N_{org}	Total
Measured bulk deposition	1.3	2.1	1.5	4.9
Estimated wet deposition	0.7	1.2	0.9	2.8
Modelled/estimated dry deposition	1.0*	2.5*	1.1	4.6
Estimated total deposition	1.7	3.7	2	7.4
Measured throughfall	0.5	1.1	1.4	3.0

* Flechard et al. (2011). NH_4^+ and NH_3 are combined as NH_4^+ and NO_3^-, NO_2 and HNO_3 are combined as NO_3^-. Organic N deposition was not included in the study.
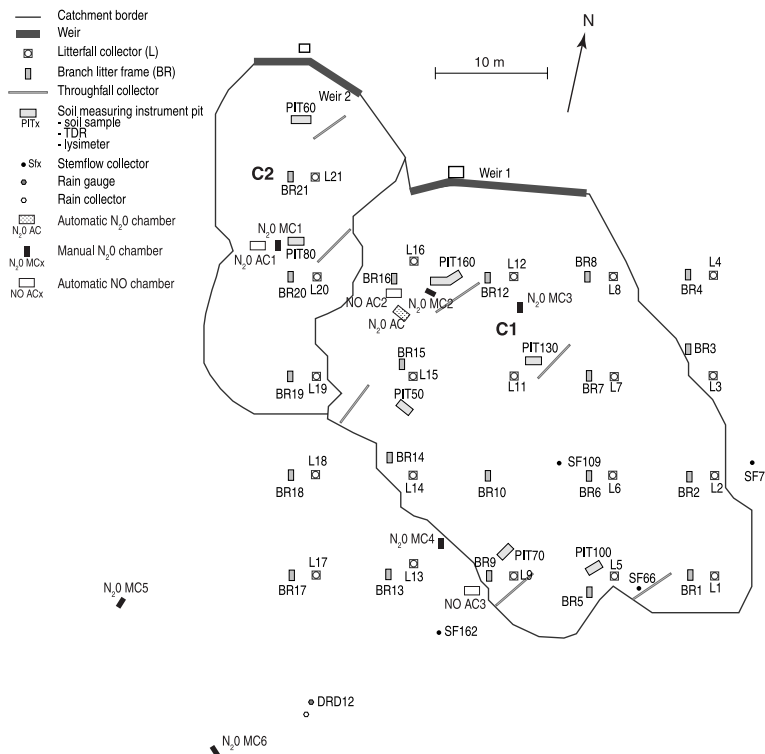
Fig. 1. Map of the measuring site indicating the sampling design for measuring nitrogen fluxes in precipitation, throughfall, stemflow, litter fall, drainage flow, NO- and N_2O-emissions and nitrogen pools in the soil water. The two catchment areas (C1, C2) are marked in the map with line, and the drainage flow is directed to the two weirs on top of the map. The height and diameter at 1.3 m of every tree in the watershed areas were measured for biomass inventory.

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Nitrogen balance of a boreal Scots pine forest

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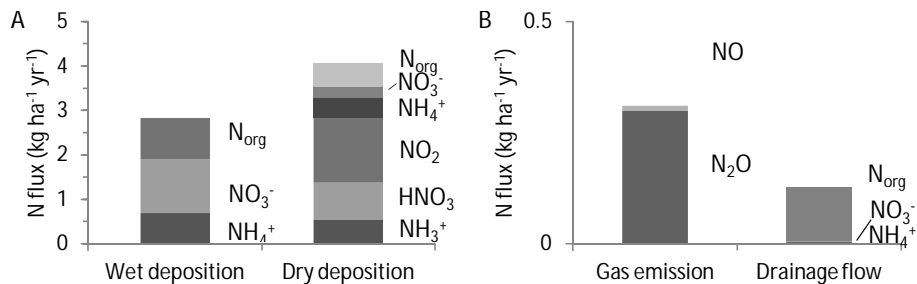
Fig. 2. Inputs **(A)** and outputs **(B)** of N in boreal Scots pine forest in Hyytiälä. Note the different y-scale in the images.
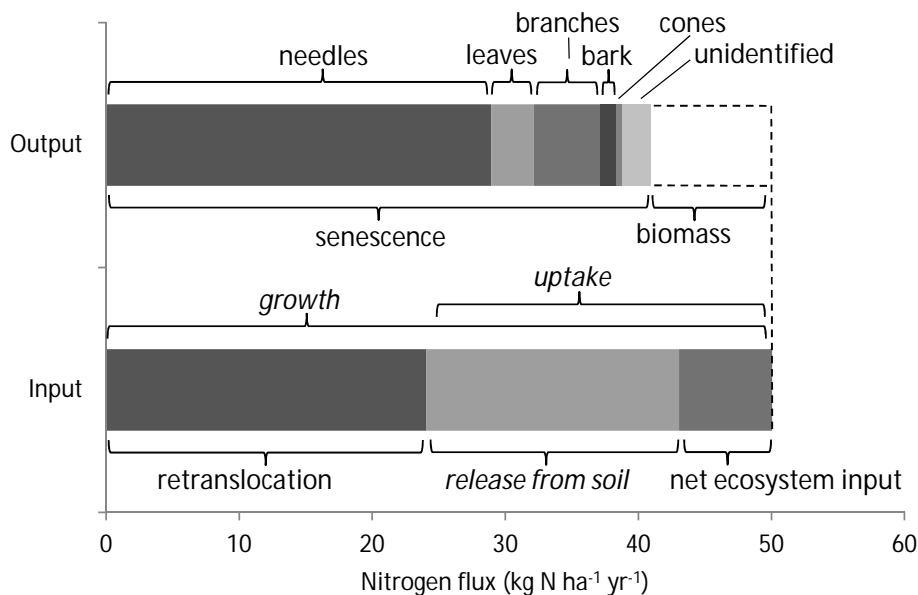
Fig. 3. Nitrogen balance of aboveground part of the trees in boreal Scots pine forest in Hyytiälä. Release from soil is the minimum amount of N uptake from soil, if all N from the atmospheric deposition was immediately available for plants. The variables marked in bold are direct measurements and variables in italics are calculated from the other variables as follows, Growth = Senescence + Biomass increment, Uptake = Growth – Retranslocation, Release from soil = Uptake – Net ecosystem input, Net ecosystem input = Deposition – gaseous emissions – drainage flow.
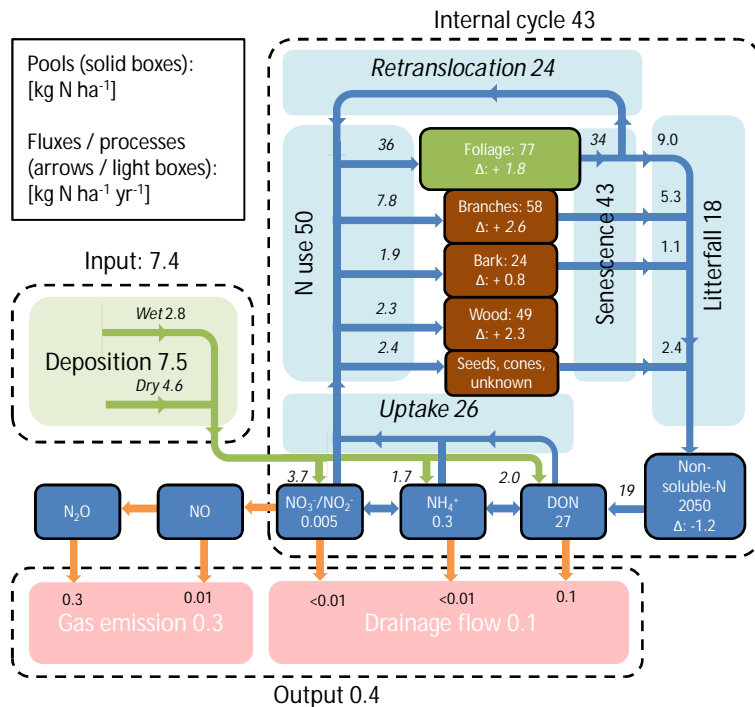
Fig. 4. Flow chart of nitrogen cycling in Hyytiälä Scots pine forest. The solid boxes are pools and arrows are fluxes. The light boxes overlapping the arrows are processes. The numbers at light boxes are total fluxes of the processes. Inputs, outputs and internal cycling of nitrogen are separated by dashed line and arrows (processes) are colored accordingly. The values originating from direct measurements are in bold. Values which are considered indirect measurements or which are otherwise more uncertain, are in italics.