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Effects of climate change and nitrogen deposition on groundwater

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Authors
Yuki Fujita, Bernard Voortman, Jan-Philip M. Witte

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More information
Dr. Yuki Fujita
T 030 60 69 707
E Yuki.Fujita@kwrwater.nl

Postbus 1072
3430 BB Nieuwegein
The Netherlands

T +31 (0)30 60 69 511
F +31 (0)30 60 61 165
E info@kwrwater.nl
I www.kwrwater.nl

Watercycle Research Institute

BTO *Managementsamenvatting*

PROBE-3 ingezet om effecten van klimaatverandering en stikstofdepositie op grondwater te bepalen

Auteurs: dr. Yuki Fujita, MSc. Bernard Voortman, prof. dr. ir. Jan-Philip M. Witte

Om te onderzoeken hoe klimaatverandering en atmosferische stikstofdepositie de kwantiteit en kwaliteit van het grondwater kunnen beïnvloeden, heeft KWR het ecosysteemmodel PROBE-3 ontwikkeld, dat de ontwikkeling van bodem, vegetatie, en water onder een veranderend klimaat kan nabootsen. In dit project is PROBE-3 geschikt gemaakt en toegepast op een grondwateronafhankelijk heidesysteem. Voor dit systeem berekent het model meer grondwateraanvulling onder zowel en gematigde (G_L) als het droge (W_H) scenario van het KNMI (2050). De nitraatuitspoeling kan zowel hoger als lager uitvallen, afhankelijk van de bodem- en planteneigenschappen van het ecosysteem. Een hoog atmosferische stikstofdepositieniveau zal de stikstofuitspoeling verhogen, vooral als het ecosysteem al verzadigd is met stikstof.

Het heide-ecosysteem van de Hoge Veluwe. Een van de lysimeters ligt onder de sensoren (links op de foto). Bodem, bodemvocht, en regenwatermonsters zijn genomen in de drie aangrenzende locaties (rondom de regenvangers).

Belang: voorspellen van toekomstig grondwater

Veranderingen in klimaat en atmosferische stikstofdepositie kunnen de kwantiteit en kwaliteit van het grondwater beïnvloeden, wat directe gevolgen heeft voor de drinkwatervoorziening in Nederland. Met behulp van een klimaatrobuust model kan op deze veranderingen worden geanticipeerd.

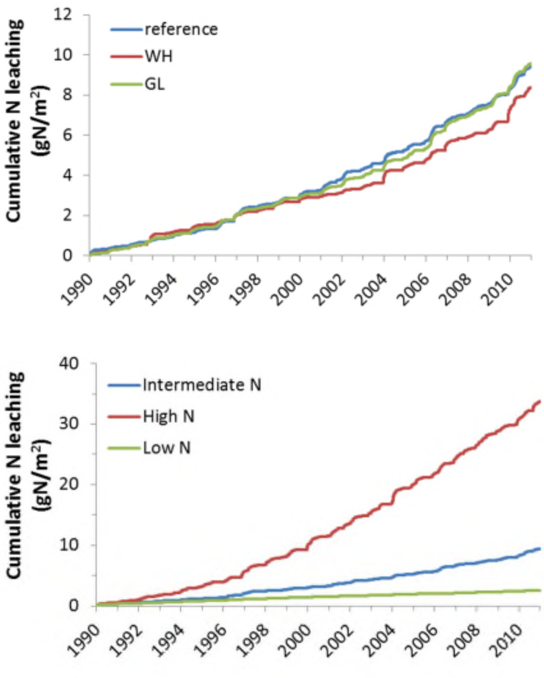
Aanpak: modelaanpassingen, veldmetingen, en scenariostudies

Het procesmatige ecosysteemmodel PROBE-3 is aangepast zodat het niet alleen kruidachtige ecosystemen kan simuleren, maar ook houtachtige. Daarnaast is een intensieve veldmeetcampagne uitgevoerd in een grondwateronafhankelijk droge-heidesysteem van het nationale park De Hoge Veluwe om grondwateraanvulling, stikstofopslag en stikstoffluxen in bodem en vegetatie te bepalen. Vervolgens, na de parametrisatie (karakteristieke systeemeigenschappen omzetten naar modelparameters), op basis van de veldmetingen, zijn verschillende scenario's van klimaat en atmosferische stikstofdepositie gesimuleerd. Daarmee zijn de gevolgen voor de grondwateraanvulling en de stikstofuitspoeling bepaald.

Resultaten: grondwateraanvullingen en stikstofuitspoeling in droge-heidesystemen

Uit de veldmetingen bleek dat het huidig niveau van stikstofuitspoeling van de heide erg laag was, veel lager dan de atmosferische depositie. Dit wijst op een hoge stikstofretentie van het ecosysteem.

De scenariostudie wees uit dat onder het klimaatscenario W_H (2050) de grondwateraanvulling zal verhogen, voornamelijk als gevolg van een verhoogde aanvulling in de winter. Ons model voorspelde gemiddeld een licht verminderde stikstofuitspoeling onder dit toekomstige klimaatscenario, maar het verschil tussen de scenario's was klein en in enkele jaren was er zelfs meer uitspoeling. Reden daarvoor is dat de stikstofuitspoeling een 'eindproduct' is van verschillende interactieprocessen, die zowel positief als negatief kunnen reageren op klimaatverandering.
Het model voorspelde een hogere stikstofuitspoeling bij een hoger niveau van atmosferische stikstofdepositie.



Gesimuleerde cumulatieve stikstofuitspoeling van droge heide onder klimaatscenario's (boven) en stikstofdepositiescenario (onder).

Implementatie: behoefte van reductie van atmosferische stikstofdepositie

Onze studie toonde aan dat het behouden van een lage stikstofdepositie van < ca. 2-3 g/m^2/jaar belangrijk is om het niveau van de stikstofuitspoeling uit de droge heide laag te houden. Dit komt min of meer overeen met de gepubliceerde kritische depositiewaarde van stikstof voor Nederlandse droge heiden (1.5 g/m^2/jaar). Het voorspelde uitspoelingsniveau bleek echter erg gevoelig voor een aantal eigenschappen van het ecosysteem. Om de stikstofuitspoeling beter te voorspellen, is het noodzakelijk het model verder te verbeteren, o.a. na validatie aan uitgebreide datasets van klimaatmanipulatie-experimenten, bij voorkeur op verschillende soorten ecosystemen.

Rapport

Dit onderzoek is beschreven in het rapport Effects of climate change and nitrogen deposition on groundwater *(BTO 2018.081).*

Summary

Background and research aims

Future climate and atmospheric deposition will affect quantity and quality of groundwater, which has direct impacts on drinking water provision in the Netherlands. Ecosystems play important roles therein, as they transform rainwater to groundwater in the thin surface layer where all kinds of interactions occur between plants, soil, and water.

This study aimed at better understanding the effects of future climate and nitrogen (N) deposition scenarios on groundwater quantity and quality (in particular N leaching), by combining the knowledge obtained from process-based modeling of the ecosystem and field measurements of lysimeter leachates, soil, and vegetation.

Methods

First, an intensive field measurement campaign was conducted on lysimeter leachate, soil, and vegetation to quantify the groundwater discharge, N pools, and N fluxes in the heathland ecosystem of the national park De Hoge Veluwe. Subsequently, PROBE3, a process-based model to simulate dynamics of soil, water, and vegetation, was adjusted so that it is now ready to simulate not only herbaceous ecosystems but also woody ecosystems. The model was then validated with the groundwater recharge, N leaching, and soil and vegetation pools obtained from the field campaign. The validated model was used to test different future scenarios of climate and N deposition on groundwater recharge and N leaching.

Results

Our field measurements revealed that the level of N leaching from our heath sites was very low (approximately 0.1 g N/m^2 in 2017), which was less than 10 % of the N input by atmospheric N deposition. The most of leached N was in the form of dissolved organic N (DON), which contrasts to the relatively high input of dissolved inorganic N (DIN; N-NH_4 N-NO_3 and N-NO_2) via atmospheric deposition all year round. This implies that the heath sites retain N via plant uptake, microbial immobilization, and adsorption to soil, acting as an N sink.

Our model predicted the drainage of lysimeter very well. The model performance was poorer for soil and vegetation, although the model predicted N leaching and N accumulation in the same order of magnitude as the measurements. In particular, the model failed in predicting low leaching rates of DIN in winter, possibly due to the lack of detailed soil adsorption processes in the model.

The scenario study of future climate indicated that future climate scenario W_H (i.e. high increase in mean temperature and high influence of air circulation change) will increase groundwater recharge from the heath ecosystems, mainly due to increased recharge in winter. Our model predicted slightly decreased N leaching rates under future climate, but the difference between scenarios was small and even reversed for some years. This is because

the climate effects on N leaching depend on the balance between multiple (and possibly counteracting) impacts on plant growth, soil accumulation, and soil nutrient transformation. These impacts are also highly sensitive to a number of ecosystem properties (e.g. plant, soil microbial, and geochemical properties), making the uncertainty of the model results very high.

The model predicted that a higher level of atmospheric N deposition led to a higher DIN leaching rate. The predicted DON leaching rates hardly differed between different scenarios of N deposition.

Implications

Our study showed that increased atmospheric N deposition will increase N leaching. The heath ecosystems retain a large amount of N, and can act as an N sink until a certain level of N deposition level (which does not saturate N of the system). Thus, in order to reduce N leaching, it is important to continue the effort of reducing the level of atmospheric N deposition.

Predicted future climate conditions will increase groundwater recharge, but their effects on N leaching are uncertain and can be either positive or negative. Many processes are involved in determining the N leaching rates, and these processes were influenced by a number of site-specific ecosystem properties. Thus, it is not possible to deduce a generic strategy for climate adaptation to reduce N leaching in future. To enable site-specific predictions of N leaching under future climate, it is necessary to improve process-based models by validating it with extensive datasets of climate manipulation experiments, preferably on different types of ecosystems.

Contents

1 Introduction

Sandy areas with deep groundwater tables, like ice-pushed ridges and coastal dunes, are of prime importance to the drinking water supply of the Netherlands, because they contain vast amounts of fresh groundwater. This groundwater is continuously replenished with rainwater that has not evaporated, but percolated out of the root zone instead. In this transformation process from rainwater to groundwater, the vegetation and top soil play a pivotal role: it is in this shallow layer where water is taken up by roots, where organic matter is formed and decomposed, and where nitrogen (N) is taken up, denitrified, or leached to the groundwater below. This system of soil, vegetation, and water thus acts as the skin of the earth: an active green filter between rainwater and groundwater.

Future climate, which is expected to be warmer and with more extreme drought and rainfall events, will affect this skin. These changes will have consequences for ecosystem services that infiltration areas provide to drinking water companies: groundwater recharge, reduction of N leaching to groundwater, and biodiversity. On top of that, atmospheric N deposition will also have a direct impact on vegetation and soil development, and therefore on ecosystem services. However, there are large uncertainties in future levels of atmospheric N deposition since it depends on governmental policy on national and European levels as well as N-emitting economical activities (e.g. agriculture, industry, traffic). Since we have rarely experienced the (combinations of) new climate conditions and N deposition levels, it is not possible to predict future groundwater solely from empirical relationships obtained from the past. To properly predict the effects of future conditions on ecosystem services, process-based modelling is a powerful tool.

For this reason, we recently developed a one-dimensional process-based model of ecosystem services, PROBE-3, which dynamically couples soil, water, and vegetation in groundwater-independent ecosystems (Fujita et al., 2016; Witte et al., 2017). PROBE-3 was applied to a coastal dune ecosystem, and it successfully simulated soil and vegetation development during a long-term dune succession. However, the plausibility of the model predictions of groundwater recharge and N leaching has not been evaluated yet. The recently installed precision lysimeters at the Hoge Veluwe provide an excellent opportunity to test and improve PROBE-3, since all the water fluxes of these devices are accurately known and do only have to be sampled for chemical analyses. In this way, the effect of future climate and changing atmospheric N deposition on groundwater recharge and N leaching can be examined by use of the validated PROBE-3 model. The results of the model scenario studies will help drinking water companies to gain insight in the future status of groundwater recharge, and to search for strategies to deal with climate change.

In this study, we aimed at improving our knowledge of effects of future conditions (in terms of climate and atmospheric N deposition) on groundwater quantity and quality, by applying the PROBE-3 model for the heathland ecosystem of the national park De Hoge Veluwe. To this end, we carried out an intensive field measurement campaign on soil, water, and vegetation to quantify the groundwater discharge, N pools, and N fluxes in this ecosystem (Chapter 2). We also adjusted the PROBE-3 model for the heathland ecosystem (Chapter 3) and validated its results with the groundwater recharge, N leaching, and soil and vegetation pools obtained from the field campaign (Chapter 4). Subsequently, the validated model was used to test different future scenarios on groundwater recharge and N leaching, in terms of climate change and atmospheric N deposition (Chapter 4). Finally, the insights from the field measurements and model scenario studies were translated into suggestions for drinking water companies to re-evaluate their strategies to deal with future changes (Chapter 5).

2 Field measurements

2.1 Study sites

This study was conducted in a heathland ecosystems at De Hoge Veluwe National Park, an ice-pushed ridge with active drift sands in the centre of the Netherlands (Figure 1). The heath vegetation is predominated by *Calluna vulgaris*, with regular occurrence of *Erica tetralix*. Grass species (mainly *Molinia caerulea*) occur only sporadically. The ground is in some places covered by moss species *Hypnum cupressiforme*; in other places without any cover. The site is groundwater independent.

Six precision lysimeters (three with free drainage system, three with a soil suction plate that mimics the water content of the surroundings) have been installed in the same location (Figure 1). The area is enclosed with a fence to ensure that there is no influence of (large) herbivores on the vegetation.

Figure 1. location of the lysimeters at the hoge veluwe, located in the centre of the Netherland (52° 3'5.86"N, 5°49'27.42"O).

2.2 Soil and water measurements

2.2.1 Lysimeter leachate collection

Leachates from three free-drainage lysimeters (Lysimeter 1, 2, and 5; 50 cm depth) were collected monthly from December 2016 to December 2017. Note that the 20 L reservoir of the lysimeters could not be completely emptied (because the outlet of the reservoir is located a little above the bottom of the reservoir), leaving always ca. 300-1500 ml of water in the reservoir after pumping. This means that the concentration of the leachate is slightly influenced by its concentration in the previous months. The leachate was kept in a freezer prior to lab analysis (§ 2.2.4).

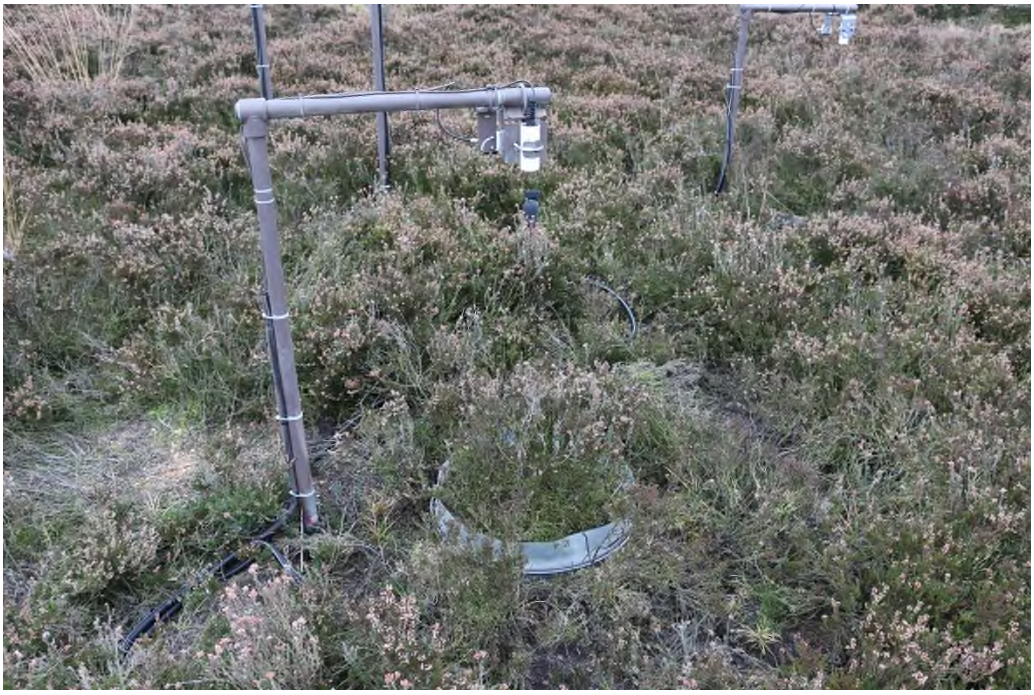


Figure 2. Precision lysimeters installed in the study site.

2.2.2 Soil pore water collection

Soil moisture samplers with porous cup (MacroRhizons, Rhizosphere) were installed at three depths (10, 25, and 50 cm) at three locations adjacent to the lysimeters in December 2016 (Figure 3). Soil pore water was collected monthly from January to December 2017. The first samples after the installation were discarded to avoid effects of disturbance on N concentrations. In dry months, some rhizons failed to collect enough water to analyze. The water samples were kept in a freezer prior to lab analysis (§ 2.2.4).

Figure 3. Soil water sampler 'macrorhizons' (left) and their installation in the field (right).

2.2.3 Rain water collection

Rain samplers with a diameter of 20 cm (RS200, METER Group AG) were installed at three locations adjacent to the lysimeters in July 2017 (Figure 4). A bird scarecrow ring was attached to each fennel to avoid contaminations by bird droppings. The bottles to store rain water were buried at a depth of 40 cm, so that the temperature of collected rain water was kept approximately at soil temperature. Rain water was collected monthly from August to December 2017. The filters were coverd by algae in September and October, causing possible contamination of the sampled water in these months. The samples were kept in a freezer prior to lab analysis (§ 2.2.4).

Figure 4. Rain water sampler.

2.2.4 Lab analysis of water samples

All water samples (i.e. lysimeter leachates, soil pore water, and rain water) were analysed for N-NH_4 , N-NOx (N-NO_2 plus N-NO_3), total dissolved N (Nts), P-PO_4, dissolved inorganic carbon (IC), and total dissolved carbon (TC) with Segmented Flow Analyser (SFA). Dissolved organic N (DON) was calculated as: Nts – N- NH_4 – N-NOx. Dissolved organic carbon (DOC) was calculated as: TC - IC.

2.2.5 Soil bulk measurement

Soil samples were collected at three depths (ca. 7.5 - 12.5, 22.5 - 27.5, and 42.5 - 47.5 cm) for three locations close to the soil pore water sampling site. The soil was analysed for the following properties: pH in 0.01M $CaCl_2$ extracts; N- NH_4 , N-NOx, Nts P-PO_4, and TC in 0.01M $CaCl_2$ extracts measured with SFA; N- NH_4 and N-NOx in 1M KCl extracts measured with SFA; total C and total N measured with elemental analyser; total P measured with SFA after H_2SO_4-H_2O_2-Se destruction; and Al, Fe, and P in oxalate extracts measured with ICP-AES. Note that KCl extraction was carried out on dried soil due to logistical constrains. Other extractions were done on fresh soils.

DON in soil pore water was calculated as in §2.2.4, using the concentrations in $CaCl_2$ extracts.

$CaCl_2$-extractable fraction of N is more or less equivalent to the freely available N in soil pore water. Since KCl is a stronger extract than $CaCl_2$, the KCl-extractable N can be considered (approximately) as the adsorbed fraction of inorganic N. Therefore, we considered N-NH_4_$CaCl_2$ and N-NOx_$CaCl_2$ as free dissolved inorganic N, and N-NH_4_KCl - N-NH_4_CaCl2 and N-NOx_KCl - N-NOx_$CaCl_2$ as adsorbed inorganic N.

2.3 Plant measurements

The above-ground biomass of heath vegetation in a frame of 50 cm x 50 cm was harvested monthly from April to November 2017. Three replicas were taken in August and October; for the rest of the months only one sample was taken. Dry weight of the above-ground biomass was estimated after drying at 65 ˚C for two days. Percentages of dead leaves were visually estimated every month in the field.

A few individuals of *Calluna* were selected and separated into ‘canopy’ (which include leaves and thin branches, until the height where green leaves are attached) and ‘branches’. Living leaves and dead lives were separately sampled for three times (in June, July and August 2017). Roots of 0 – 20 cm depth were collected in June 2017 with a metal core of diameter 19 mm. The roots were carefully washed with water to remove soils. Five parts of plants (i.e. canopy, branches, living leaves, dead lives and roots (N=3 for each)) were dried at 65 ˚C for 2 days.

Plant litter was collected monthly in three litter traps of diameter 12 cm per location, from March to November 2017.

After grinding, concentrations of C and N in each part of plants were determined with CN element analyser. Furthermore, concentrations of phosphorus (P) and N were determined with SFA after digesting with H_2SO_4 - H_2O_2 - Se.

Figure 5. Calluna vegetation with 50 cm x 50 cm frame.

2.4 Calculation of N pools and N fluxes

The amount of N stored in different pools of the heath ecosystems was calculated as follows. The N pools in heath canopy and branches were estimated by multiplying their biomass (average values of 8-times measurements between April and November 2017) with N concentrations in these parts (average values of June, July, and August 2017). Litter N pool was calculated by multiplying cumulative litter fall for 12 months with N concentration in dead leaves (average value of June, July, and August 2017). Root N pool was approximated by multiplying root biomass measured in June 2017 with N concentration in the roots. Soil N pools were estimated separately for three depths (0-20 cm, 20-30 cm, and 30-50 cm), based on soil measurements in July 2017 at the depths of 7.5-12.5 cm, 20-30 cm, and 40-50 cm respectively.

Inflow of N to the ecosystem via atmospheric N deposition was estimated by multiplying annual precipitation of 2017 at Deelen weather station (916.2 mm) with the average N concentrations in rain water samples collected between August and December 2017. Other possible pathways of N influx are symbiotic and asymbiotic N fixation, but these processes were not measured in this study.

Outflow of N via leaching was estimated by multiplying the amount of drainage from the three free-drainage lysimeters with the N concentrations in the leachate samples, for each of the 12 measurement periods (which had approximately monthly intervals). Measurements of lysimeter drainage were sometimes temporarily missing due to technical errors. For those periods, the missing drainage data was obtained by using average daily values of the other two lysimeters. Since the leachate collection was conducted for the period of December 10th 2016 - December 17th 2017, the cumulative N leaching measured for this specific period was used as annual estimate of N leaching.

Other possible pathways of N outflow are denitrification and grazing by animals (possibly by insects and small animals only, because the area was fenced to prevent herbivory by large animals). These pathways were not measured, but are expected to be of minor importance.

3 Modelling heath ecosystems

PROBE-3, the process-based model to simulate ecosystem dynamics, consists of modules of vegetation, soil, and hydrology (Fujita et al. 2016).

The vegetation and soil modules are based on the existing model CENTURY. We used the modified version of the CENTURY model of Fujita *et al.* (2016), which was tested for herbaceous ecosystems in the Netherlands. A heathland, however, is not an herbaceous ecosystem, but an ecosystem dominated by dwarf shrubs. In order to simulate heathlands, we added a forest compartment, based on the forest submodule of CENTURY ver. 4 (Metherell *et al*., 1993, Kirschbaum, 1999) (§3.1). Furthermore, we modified the N leaching processes of CENTURY to reflect relatively high leaching rates of dissolved organic N (DON) which we observed in our study site (§3.2).

The hydrology module was reconstructed using Hydrus 1D (Šimůnek *et al*., 2008) (§3.3). Soil moisture and soil temperature simulated by Hydrus 1D was used as input data in the vegetation and soil modules. Unlike in the version of Fujita et al. (2016), the feedback from vegetation and soil modules to hydrology module was not included.

3.1 Forest module

To simulate forest ecosystems, three state variables of woody litter pools are added: C_9 (Dead fine branches), C_{10} (Dead large wood), and C_{11} (Dead coarse roots). They are decomposed in the same manner as the surface structural pool (for C_9 and C_{10}) or belowground structural pool (for C_{11}). Furthermore, five pools of living tree biomass are added: leaves ($TreeC_0$), fine roots (<2mm diameter) ($TreeC_1$), fine branches (<10cm diameter) ($TreeC_2$), large woods ($TreeC_3$), and coarse roots ($TreeC_4$). See Figure 6 for state variables of C and flows between them in the CENTURY model.

Equations of dynamics of tree biomass and woody litter decomposition were taken from CENTURY ver. 4 (see Appendix V for detailed description of these equations). For simplicity, we assumed that there is no growth of herbaceous plants under the heath canopy.

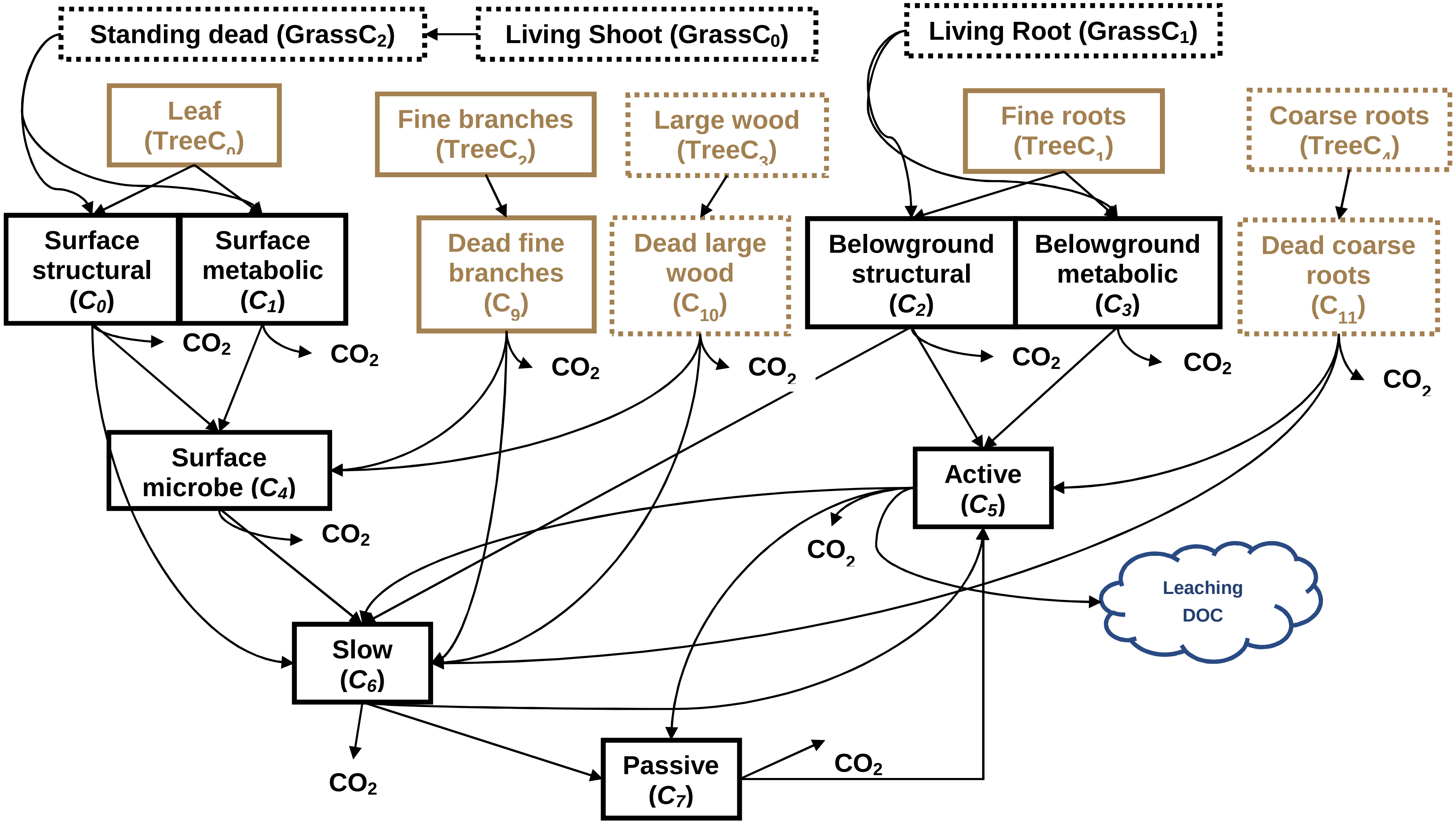


Figure 6. State variables of C in the CENTURY model. Arrows represent C flow rates. Brown boxes are forest-specific state variables. State variables in dotted boxes are not used in this study.

3.2 N leaching

In the original CENTURY model, leaching of DON and dissolved organic carbon (DOC) are computed as a fraction of decomposed active pools, which is a function of sand content and amount of leached water. We did not use the equation of CENTURY, as it leads to higher leaching of DOC and DON in warm months when decomposition rate of organic matter is high, whereas in our study sites in the Hoge Veluwe such seasonality was not observed. Instead, we assume that a part of DOC (e.g. biologically recalcitrant DOC, which is not easily taken up by plants and microbes) is prone to leaching. The amount of leached DOC was calculated by multiplying the concentrations of the recalcitrant DOC with the water flux, the same way as we modelled mineral N leaching. We did not explicitly model DOC- and DON pools in the top soil as separate state variables, but assumed that a certain fraction of the active pool ('max_fdoc') exists as biologically recalcitrant DOC. The proportion of the DOC which is not leached due to adsorption to soils was assumed to increase with decreasing sand content, as in the original CENTURY model. The leaching rates of DOC from different soil layers are written as:

$$fra\_doc = max\_fdoc \cdot (omleach1 + omleach2 \cdot T_S)$$

$$J_{DOC,1} = C_5 \cdot fra\_doc \cdot \frac{F_1}{S_1}$$

$$J_{DOC,2} = DOC_2 \frac{F_2}{S_2}$$

$$J_{DOC,3} = DOC_3 \frac{F_3}{S_3}$$

where *fra_doc* is the fraction of active C pool which is prone to leaching (fraction, between 0 and 1), *max_fdox* is the fraction of active C pool which is dissolved and prone to leaching (fraction between 0 and 1), *omleach1*and *omleach2* are the intercept and slope for effect of sand on adsorption of DOC, $J_{DOC',i}$ is the leaching rate of DOC from soil layer *i* (g C/m²/d), C_5 is the amount of active C pool (g/m²), S_i is the water content in soil layer *i* (cm), and F_i is the water flux from soil layer *i* (cm/d), and DOC_i is the amount of DOC in soil layer *i* (g C/m²). The values of *omleach1*and *omleach2* were taken from CENTURY ver. 4 (*omleach1*=0.01, *omleach2*=0.04). We consider the leaching rate of the third soil layer as leaching rate of the system.

3.3 Soil moisture and temperature

Soil water fluxes and soil temperature were simulated using the hydrological model Hydrus 1D (Šimůnek *et al.*, 2008). The soil vertical layers were divided into 0 - 20 cm, 20 - 50 cm, and 50 – 120 cm. Soil hydraulic properties of each layer were approximated based on sand box measurements of soil cores sampled from three depths (Appendix I). We used a free drainage boundary condition at 3 m below the surface to simulate groundwater independent conditions. For temperature simulation, Chung & Horton formula (Chung & Horton, 1987) was used to calculate thermal conductivity. Air temperature was used as the upper boundary condition. Zero gradient was chosen as the lower boundary condition. The model has been validated with evapotranspiration data of the same site (Voortman, B., unpublished).

In addition, we simulated water conditions and soil temperature within free-drainage lysimeter of 50 cm depth (as in the three lysimeters which we collected leachates from), as they can be slightly different from those in ambient conditions. For this, we used two soil layers (0 – 20 cm, 20 – 50 cm), and the lower boundary at 50 cm depth was set to be pressure head of 0 cm.

Meteorological input data needed for the Hydrus simulation were: daily precipitation, daily Makkinks evapotranspiration, average daily temperature, and daily amplitude of temperature (i.e. half of the difference between minimum and maximum daily temperature).

The following output of Hydrus simulation was used as input for CENTURY: volumetric water content of middle points of each layer, water fluxes between soil layers, and soil temperature at 10 cm depth, all on daily intervals. Downward water flux from 50 cm depth was considered as drainage from the lysimeter.

3.4 pH effect on decomposition

Pre-simulations with the Century model indicated that the modelled N mineralization rate (using the default parameter values of decomposition rates and pool divisions, as well as actual climate conditions of Veluwe) was much higher compared to measured mineralization rates of heathland soils in Oldebroek (Beier *et al.*, 2009). Our model simulated that ca. 4 % of total soil N was mineralized in a year, whereas the measurements in Oldebroek showed that less than 1 % of total soil N was mineralized in a year. This may be due to inhibition effects of low pH in heathlands on decomposition rates. To reflect that, we introduced a pH effect on organic matter decomposition, using the equations of Walse *et al.* (1998):

$$f_{pH} = \frac{1}{1 + K_{pH} \cdot \left(10^{-pH}\right)^{m}}$$

where f_{pH} is the reduction term on decomposition rate (fraction, between 0 and 1), K_{pH} is the response coefficient, and m is the exponent. K_{pH} value can vary depending on the type of substrate to be decomposed. We set the K_{pH} to be 4640, an intermediate value of K_{pH} for holocellulose (20500) and lignin (1050) (i.e. the geometric mean of the two values). m value was set to be 1, as was fitted for most cases (Walse *et al.*, 1998). With these parameter values, the modeled N mineralization rate was tuned to be ca. 2 % of total N per year. Note that the choice of this parameter has a direct and strong influence on the modelled inorganic N concentrations in the top soil.

3.5 Model parameterization and input data

We defined three soil layers in the CENTURY model. Soil organic matter (SOM) accumulation occurs only in the top layer, whereas flows of dissolved N are simulated in all three layers. Depth of the top layer, second layer, and third layer was set to be 0 – 20 cm, 20 – 50 cm, and 50 – 120 cm depth, respectively. All plant roots were assumed to be distributed in the top soil layer.

Initial values of soil and plant pools as well as of important model parameters values are summarized in Table 6 in Appendix V. When possible, the values were estimated based on the field measurement of this study.

N deposition levels of 2016 and 2017 were estimated from our own measurements of rain water (see § 2.4). We took the lower estimates of the deposition level (i.e. the average values calculated without possibly contaminated months).

Meteorological data of the last decades was downloaded from the KNMI website (http://www.knmi.nl/nederland-nu/klimatologie/daggegevens). We used the data of the nearest weather station, Deelen. Makkinks potential evapotranspiration, precipitation, maximum and minimum temperature on a daily basis were retrieved. Note that a complete set of these variables are available only after April 1987.

To validate the model with the field data of 2017, the model was run for the period of 2016 – 2017. The newly introduced parameter for N leaching, *max_fdoc* (i.e. the fraction of active C pool which is dissolved and subject to leaching) was tuned so that the predicted DON leaching resembled the field measurement in 2017.

3.6 Scenario study

3.6.1 Future climate scenarios

KNMI published future climate change in the Netherlands as 'KNMI' 14 climate scenarios' (KNMI, 2014). To assess the impact of the most extreme change and a moderate change, we used the W_H and G_L scenario. The weather data for the reference period 1981 -2010 of station Deelen (for potential evapotranspiration, temperature) or Schaarsbergen (for precipitation, since data of Deelen was not available of this period) were transformed for the time horizon of 2050 (i.e. 2036 – 2065) with the transformation tool of KNMI (http://www.klimaatscenarios.nl/toekomstig_weer/transformatie).

The heath ecosystem was simulated with the CENTURY model for 22 years, with the transformed weather data for the W_H and G_L scenario (projected for the years 2044 - 2065), as well as with the reference weather data (1989 – 2010). We used the measurement data of 2017 to estimate the input values of C and N pools in soil and plant. Atmospheric N deposition level of 2016 was used for all simulated years.

3.6.2 Atmospheric N deposition scenarios

To set different levels of atmospheric N deposition, we referred to the annual national average values of atmospheric N deposition (as NHx and NOy, both dry and wet, mol/ha/year) in the Netherlands. Historical average values from 1900 to 1980 were taken from Noordijk (2007), and the average values from 1981 to 2016 were retrieved from CBS *et al.* (2017) (Figure 7). As N deposition scenarios, we assumed three levels of N deposition: high N deposition level (i.e. the level of 1980 in the Netherlands; 4.46 g N/m^2/year), intermediate N deposition level (i.e. the level of 2016 in the Netherlands; 2.27 g N/m^2/year), and low N deposition level (i.e. the level of 1900 in the Netherlands; 0.48 g N/m^2/year) (Figure 7). The model was run for the period of 1989 – 2010 with actual climate data from weather station Deelen. We used the measurement data of 2017 to estimate the input values of C and N pools in soil and plant.

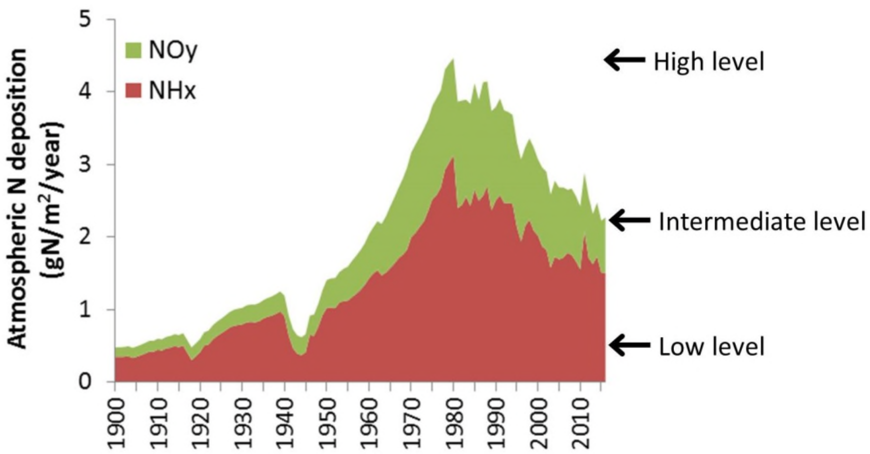


Figure 7. National average of atmospheric N deposition in the Netherlands, between 1900 and 2016. The three levels of deposition used for the scenario study are indicated with arrows.

4 Results and discussion

4.1 Field measurements of N balance and N fluxes

4.1.1 N concentrations in rainwater, soil water, and leachates

Average nitrogen concentrations in three types of water samples (rain water, soil pore water, lysimeter leachates) are shown in Figure 8.

In rain water, the range of N-NH_4 and N-NOx is in line with other studies. Our estimates were 0.92 – 1.43 mg/L for N-NH_4 and 0.47 mg N/L for N-NOx. National averages of concentrations of N- NH_4 and N-NOx in the Netherlands are similar; 1.5 mg N-NH_4 /L and 0.76 mg N-NO_3 /L in 2016 (CBS *et al.*, 2017); 0.5-1.1 mg N-NH_4 /L and 0.6-1.0 mg N-NO_3 /L in Amsterdamse Waterleidingduinen in 1981-1983 (Stuyfzand, 1991). In the monitoring station Speuld of RIVM, which is located ca. 30 km north-west of our study site, the average concentrations in rain water (collected in wet-only rainwater collectors) were 0.81 mg N/L for N-NH_4 and 0.35 mg N/L for N-NO_3 (see Appendix III for seasonal variation of wet N deposition in Speuld).

Concentrations of DON in rain water were on average 0.43 mg/L, accounting for ca. 19 % of total dissolved N in rain water. If we excluded the possibly contaminated samples of September and October (i.e. ‘low estimate’ in Figure 8), the DON concentration was 0.09 mg/L, accounting for ca. 6 % of total N. This is slightly higher than another measurement at 11 stations in the Netherlands in 2008, where only negligible amounts of organic N (0 - 0.14 mg N/L, accounting for at most 3 - 4 % of total N) were found in rain water (Buijsman, 2010). In these samples, the DON was detected in the form of urea but not amine or amino acid. At the Belgian coast, ca. 7 – 20 % of total N in rain water was DON (Bencs *et al.*, 2009). In other regions of the world, DON sometimes contributes to a substantial part of total N in rain water; ca. 19% in New England, the USA (Campbell *et al.*, 2000); up to 17 % on a coastal site of Turkey (Mace *et al.*, 2003b); and ca. 19 % in Tasmania, Australia (Mace *et al.*, 2003a).

Concentrations of DIN (N-NH_4 and N-NOx) in leachate were constantly low (Figure 8, Figure 27). DIN in soil pore water was also mostly low, but nitrate was occasionally relatively high (>0.1 mg N/L) at 50 cm depth (Figure 28). There was no clear seasonal dynamics in the DIN concentrations (Figure 27, Figure 28).

DON concentrations in soil water and leachates were rather high (0.34 – 0.57 mg N/L; Figure 8), but they still fall within the observed range in agricultural soils or in forest soils. The review of Van Kessel *et al.* (2009) showed that DON concentrations in leachates from agricultural systems, collected from the lowest depth being reported, range between 0.2 and 3.5 mg N/L. In the Netherlands, the range of leached DON from agricultural soils in 2015 was, on average per soil type, between 0.52 and 3.2 mg N/L (‘Landelijk meetnet effecten mestbeleid’, RIVM). DON concentrations in discharge water from forests are usually much lower: <0.2 mg N/L in both regions under high and low atmospheric N deposition in America (Perakis & Hedin, 2002); up to 0.5 mg N/L in New England, the USA (Campbell *et al.*, 2000).

It is striking that DON concentrations in our soil water and leachates were high compared to very low concentrations of NH_4 and NOx. The proportion of DON in total dissolved N concentration in leachates and soil water accounted for 89 - 99% in our study. This is much higher than the observed range at the similar heath ecosystem in Oldebroek, where DON makes up ca. 27% of total leached N, averaged over 7 years of 9 replicates (pers. comm. Albert Tietema (UvA)). However, it is not uncommon in forest ecosystems that DON makes up a high percentage of total N leaching. In stream water discharged from unpolluted forests in South America (i.e. with low atmospheric deposition), very low concentrations of NH_4 and NO_3 and high concentrations of DON (8-135 µg/L, which accounts for 61-97 % of total leached N) were observed (Perakis & Hedin, 2002). Furthermore, Neff *et al.* (2003) postulated that DON leaching may represent a significant 'leak' of N from terrestrial ecosystems, even in times of high N demand, because most (but not all) forms of DON are not readily available for plants and microbes. High leaks of DON in N-deficient ecosystems are suggested by a number of empirical studies. For example, in forested ecosystems, precipitation inputs of DON were approximately equal to output of DON in stream, whereas DIN outputs had clear seasonal patterns (i.e. DIN outputs were much lower than DIN inputs especially in summer when biological activity was high) (Campbell *et al.*, 2000). Similarly, DIN leaching loss was strongly reduced by increase in species richness of plants, whereas DON leaching loss was less reduced by changes in plant communities (De Deyn *et al.*, 2009).

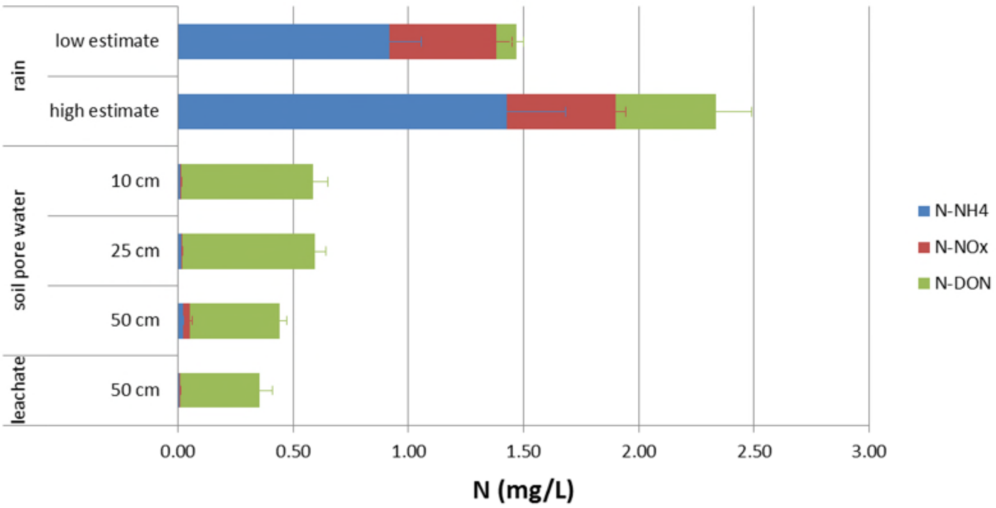


Figure 8. N concentrations in rain water, soil pore water at three different depths, and in lysimeter leachates. Average + SE over the entire measurement period and three locations are shown. For rain water, there were 2 sample periods when the collected water samples were possibly contaminated (september and October). Here the N concentrations were calculated as high estimate (i.e. including all samples) and low estimate (excluding the possibly contaminated samples). See Figure 26, Figure 27, and Figure 28 in Appendix II for raw data of each month at each location.

4.1.2 P concentrations in rainwater, soil water, and leachates

P concentrations in soil pore water and leachates were negligible, almost always under detection limit of 0.02 mg P/L. Phosphate concentrations in rain water was on average 0.02 mg P/L (with a standard error of 0.01).

4.1.3 Soil bulk properties

Extractable concentrations of inorganic N (NH_4 and NOx) were low, not exceeding 2 mg N/kg soil at any depth (Figure 9). This is comparable to the KCl-extractable DIN concentrations in a heath stand in Veluwe (1.2 mg N-NH_4 /kg and 0.3 mg N-NO_3 /kg (Fujita *et al.*, 2013b)), but lower than those in other heath soils in the Netherlands (6.4 mg N-NH_4 /kg and 0.12 mg N-NO_3 /kg on average of five locations (Ordoñez *et al.*, 2010)). C:N ratio of our soils was high, ranging between 24 – 38, which was much higher than the typical range of soil C:N ratio in global studies (ca. 14.5 for forest soils (Cleveland & Liptzin, 2007); ca. 14 for evergreen shrubs (Ordonez *et al.*, 2009)). Similarly, C:P ratio of our soil was rather high, ranging between 138 (deep soil) and 304 (top soil), compared to the typical level of forest soils (ca.

212) (Cleveland & Liptzin, 2007). These high ratios of C:N and C:P imply that the soil in our study site is nutrient-deficient, supporting very low concentrations of dissolved inorganic N and P in the soils.

Extractable amount of NH4 was higher than that of NOx, indicating somewhat inhibited nitrification process due to low pH (pH 3.6 at top soil – pH 4.4 at 45 cm depth (Table 1). Some, but not much, amount of NH4 could be extracted by KCl, but not by CaCl2, indicating that this fraction of NH4 was adsorbed to soils. These fractions are most probably adsorbed to SOM and metal oxides, but not to clay, as the clay content in our soil was low (on average 0.7 % as measured in different locations in the Hoge Veluwe (Fujita et al., 2013b)). The oxalate-extractable Fe and Al were rather high in our soils (Table 1).

Concentrations of DON were much higher than concentrations of DIN, and decreases with depth. DON is produced by many processes, such as microbial turnover, extracellular enzymes excretion by microbes and plant roots, physical dissolution or desorption of organic matter from litter or soil, and association of dissolved organic matter with nitrate (Neff *et al.*, 2003). Some forms of DON (e.g. amino acid) disappear rapidly because they can be directly taken up by plants or rapidly mineralized. Other parts of DON remain in soils and are prone to leaching even when plant and microbes have high demand for N.

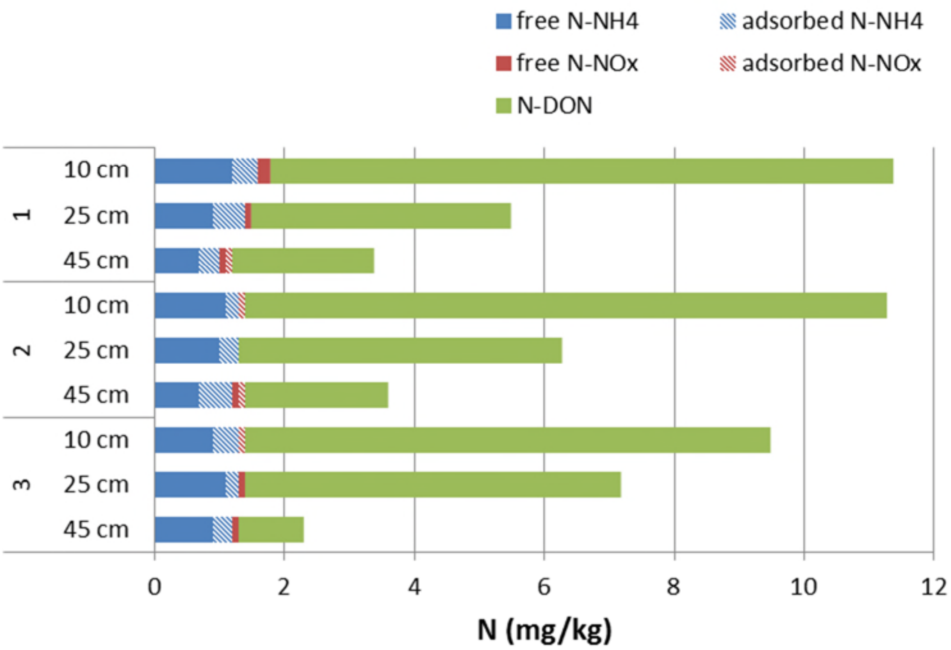


Figure 9. Concentrations of available N in soil at three depths of three locations. Free N-NH_4 and N-NOx are the $CaCl_2$-extractable N-NH_4 and N-NOx; adsorbed N-NH_4 and N-NOx are the differences between KCl extractable and $CaCl_2$ extractable N-NH_4 and N-NOx; DON is total dissolved inorganic N in $CaCl_2$ extracts minus $CaCl_2$-extractable N-NH_4 and N- NO_3 .

Table 1. Soil pH_CaCl$_2$, total carbon, total N, total P, oxalate-extractable Al, Fe, and P at three depths. Average and standard errors of 3 locations are shown.

depth	**pH_CaCl$_2$**		**Total C (g C/kg)**		**Total N (g N/kg)**		**Total P (mg P/kg)**		**Alox (mg Al/kg)**		**Feox (mg Fe/kg)**		**Pox (mg P/kg)**	
	ave	SE	ave	SE	ave	SE	ave	SE	ave	SE	ave	SE	ave	SE
10 cm	3.64	0.11	29.40	3.41	0.99	0.17	96.7	14.4	998.3	229.3	1468.3	119.9	49.7	4.3
25 cm	4.01	0.09	21.63	0.96	0.69	0.01	87.7	5.2	1857.3	106.4	1904.3	424.6	52.0	2.1
45 cm	4.42	0.02	10.32	1.94	0.31	0.02	75.0	7.2	2224.3	96.2	958.7	67.4	35.0	1.0

4.1.4 N and P content in plant biomass

The concentrations of C, N, and P in different parts of plant biomass are summarized in Table 2. The plant N:P ratio of the whole above-ground biomass (i.e. ‘canopy’ + ‘branches’) was 35.5, indicating a strong P limitation on the plant growth.

Table 2. Element concentrations of Carbon (C), N, and P and N:P ratio in plant biomas of heath, samples in summer months in 2017. ‘Canopy’ is the upper part of biomass which include green leaves and thin branches. ‘Branches’ is the lower part of biomass which does not include green leaves but only branches.

	C (g/kg)		**N (g/kg)**		**P (g/kg)**		**N:P ratio**	
	ave	SE	ave	SE	ave	SE	ave	SE
living leaf	518.3	1.2	14.33	0.53	0.55	0.03	23.14	0.57
dead leaf	534.7	3.2	13.70	0.21	0.25	0.01	48.56	2.41
canopy	523.7	1.8	10.90	0.49	0.35	0.01	27.97	0.96
branches	513.0	0.6	7.92	0.43	0.18	0.01	39.92	1.54
roots	352.3	31.2	9.78	0.41	0.32	0.02	25.33	1.45

4.1.5 N leaching from lysimeters

Amounts of leached N from lysimeters were calculated based on the monthly measurements of the lysimeter leachates (Figure 10). Although the amount of drainage was similar between the three lysimeters, the estimated amount of N leaching differed strongly between the lysimeters, ranging from 78 mg N/m^2 to 129 mg N/m^2 for the measurement period of ca. 1 year. This indicates a strong spatial heterogeneity of the N transformation processes. In all lysimeters, the amount of DIN was minor, whereas DON accounts for the majority of N leaching.

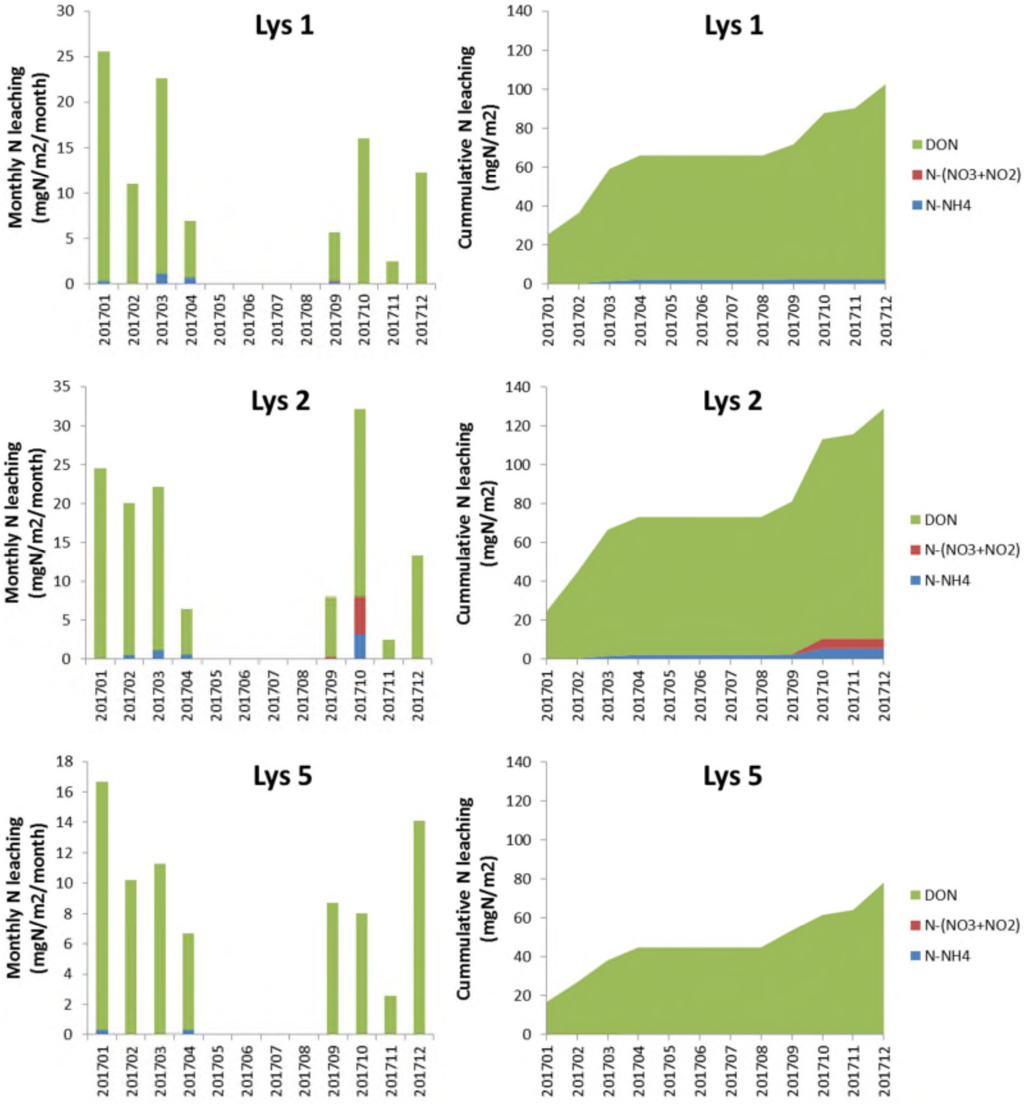


Figure 10. Monthly N leaching (left) and cumulative amount of N leaching (right) from three free-drainage lysimeters (Lysimeter 1, 2, and 5). Leached N is split into reduced N (NHx), nitrogen oxides (NOy) and dissolved organic nitrogen (DON). Note that monthly leaching was calculated not exactly on a monthly interval; the measurement interval sometimes deviate from 30 days (with a range of 24 – 38 days).

4.1.6 N pools and fluxes of heath ecosystems

N pools in the heath ecosystem were calculated for different parts of the ecosystem (Figure 11). Most of N (ca. 93 %) in the heath ecosystem is stored in the soil. See Table 3 in Appendix IV for pool sizes of other elements (i.e. C and P).

Measured N fluxes are shown in Figure 12. N outflux is much lower than N influx, even when the lower estimate of N deposition is used. From this, it is anticipated that ca. 1 - 2 g N/m^2 of N is accumulated in the ecosystem this year, either in plants or in soil.

The N balance of our study site was compared with those of other shrublands in Europe measured by Beier *et al.* (2009), especially with the heathland in Oldebroek (Figure 13). The heathland of Oldebroek is located ca. 50 km away from our study site and has similar climatic and edaphic conditions. Our heath site has comparable top soil N pools as Oldebroek, yet plant N pools were much larger in our site. The soil N pool in Oldenbroek was characterised by a very organic-rich (65% SOM) shallow top soil of 4 cm depth and an organic-poor (3.3% SOM) subsoil of 4-16 cm depth, whereas the soil N pool in our study site had a less sharp gradient with depth, with moderately organic-rich soil (SOM 4.3 %) even at 25 cm depth. Furthermore, N deposition rate is lower in our study site than in Oldebroek, as the measurements in Oldebroek took place in years when N deposition level in the Netherlands was higher (ca. 1999-2004). These two reasons explain why we observed much lower N leaching rates than at Oldebroek (i.e. higher plant N uptake and lower N input via deposition).

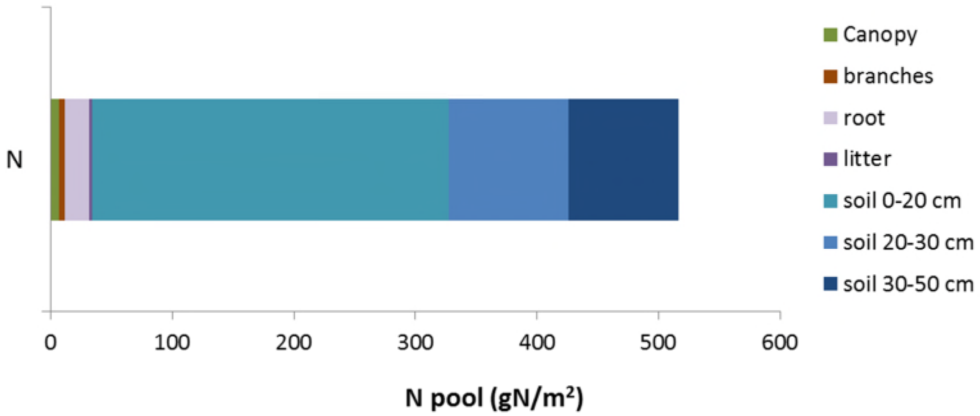


Figure 11. N pool in different parts of heath vegetation, litter, and soils.

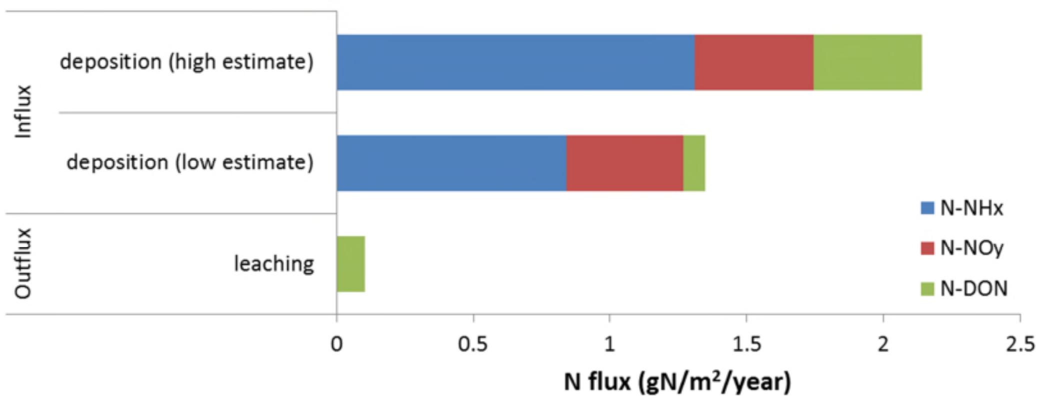


Figure 12. Annual Influx and outflux of N in 2017. The flux is divided into reduced nitrogen (NHx), nitrogen oxides (NOy) and dissolved organic nitrogen (DON). N influx via atmospheric deposition was calculated as high estimate (using the average concentrations of N in all measurements) and as low estimate (using the average concentrations of N except the samples in september and october, which was possibly contaminated). N leaching is the average value of three free-drainage lysimters, measured for the period of 10 dec 2016 - 17 dec 2017.

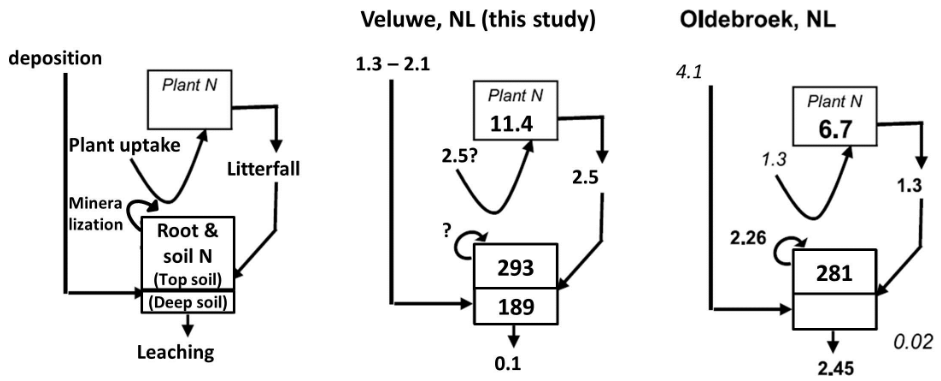


Figure 13. N balance of the heath ecosystem in Veluwe (top), compared with the heath system of Oldebroek (Beier et al., *2009). N pools (g N/m²) are shown in boxes, and fluxes (g N/m²/year) are shown with arrows. Note that the N pool in soil in Oldebroek is of 0 – 16 cm depth. N pool size of this study is divided into 0-20 cm and 20 - 50 cm depth. Figures adapted from Beier et al. (2009).*

4.2 Modelling hydrology of heath ecosystems

The Hydrus 1D model simulated drainage from the free-drainage lysimeter well. The drainage in the period of 10 December 2016 to 17 December 2017 was modelled as 456 mm, and measured in the field as 478 mm (average over three lysimeters). The seasonal dynamics of drainage was also well captured by the model (Figure 14), yet there was a small mismatch between modelled and measured values in spring (i.e. the model underestimate drainage during the wet period in February-March). The higher drainage in the measurements might reflect preferential flow along the wall of the lysimeter.

The modelled drainage in ambient conditions was slightly lower (411 mm) than the modelled drainage from the free-drainage lysimeters (456 mm). This is probably because in ambient conditions upward capillary flow of water from deeper layers can occur during periods of drought.

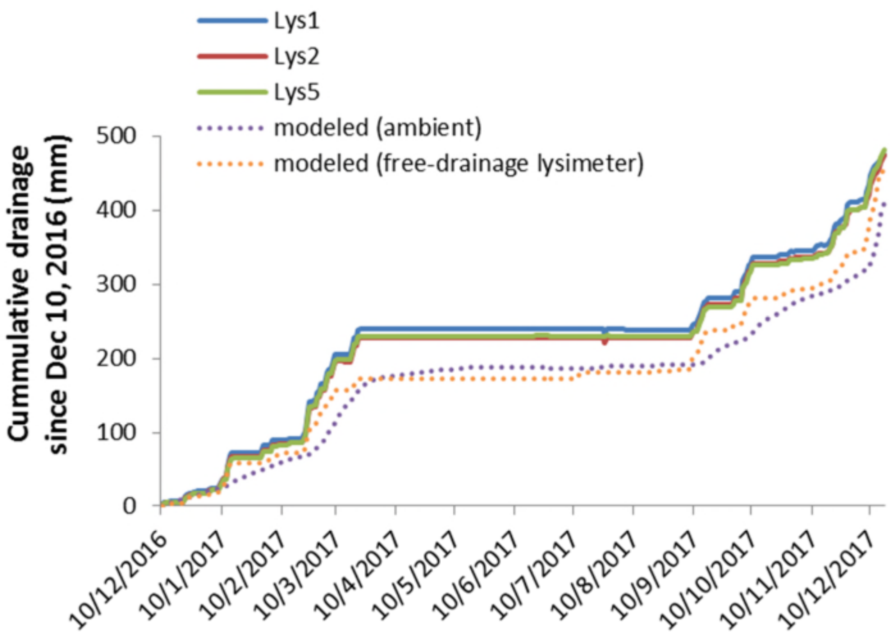


Figure 14. Measured water fluxes from bottom of three free-drainage lysimeters (at 50 cm depth), for the period of 10 Dec 2016 – 17 Dec 2017. Water fluxes at 50 cm depth were simulated with Hydrus 1D for ambient conditions (with deep groundwater level of 3 m) (dotted purple line), as well as for the condition of free-drainage lysimeter (i.e. pressure head of 0 cm at 50 cm depth) (dotted orange line).

4.3 Modelling N leaching and N balance of heath ecosystems

The CENTURY model predicted that the leaching of 2017 (or more accurately, for the period of 10 Dec 2016 – 17 Dec 2017) was 0.14 g N/m^2, to which DIN and DON contributed 0.03 and 0.11 g N/m^2, respectively (Figure 15). The predicted DON leaching was in the same range as the measured values, whereas the model predicted a slightly higher level of DIN leaching.

The model input for N inflow was 1.27 g N/m^2/year. The denitrification level was predicted to be neglectable (0.0004 g N/m^2/year). Thus, the model predicted that the heath system accumulated 1.13 g N/m^2 during that period. This is in the same range as our estimates of N accumulation based on our field measurement (i.e. 1-2 g N/m^2/year) (§ 4.1.6).

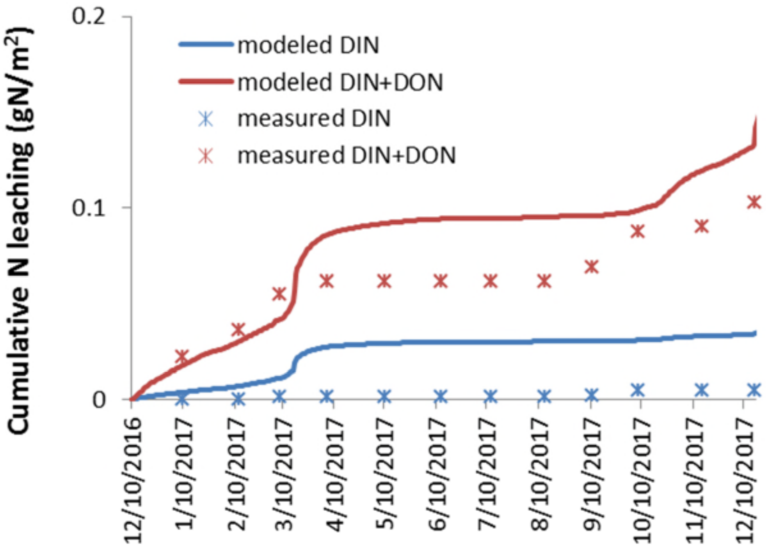


Figure 15. Modeled N leaching for the period of 10 Dec 2016 to 17 dec 2017. The monthly measured values of N leaching during the same period are also shown. DIN include N-NH_4 and N-NOx.

4.4 Scenario study with future climate

The W_H scenario of KNMI predicted that in future temperature increases, precipitation increases in winter, and precipitation decreases in summer (Figure 16). The G_L scenario has similar but less prominent trends (Figure 16).

The Hydrus 1D model predicted that, with the W_H scenario, soil temperature increases throughout the year. Soil moisture decreases in summer time, increasing transpiration deficiency in that period. Furthermore, drainage from 50 cm depth increases in winter periods (Figure 17).

The CENTURY simulation showed that, despite the increased drainage in the W_H scenario, cumulative N leaching was not higher in the W_H scenario than under the reference climate (Figure 18 top). DON leaching was indifferent between the three climate scenarios, whereas DIN leaching was similar during the first 10 years and then lowest in the W_H scenario. Several processes caused this pattern, and some of these processes were counteracting. First, due to the elevated soil temperature, plant production becomes higher in the W_H scenario. Although increased transpiration stress in summer supressed the plant production in the W_H scenario, this negative effect on biomass was not large enough to counteract the positive effect of temperature, leading to the net increasing effect of the climate on the plant biomass (Figure 19, left). This led to higher uptake of DIN by plants, which lowers the DIN concentration in the soil. Furthermore, higher soil temperature speeds up decomposition of soil organic matter. This increases mineralization of N from the soil, but at the same time slows down accumulation of the SOM pool compared to the reference climate (Figure 19, left). All together, the weather conditions of W_H led to a decreased concentration of DIN in the top soil and therefore slightly decreased the N leaching. Note that whether the net effect of the climate on DIN availability is positive or negative depends on the subtle balance among the different processes. This means that the net effect of the W_H scenario on N leaching can be positive for a year but negative for another year (Figure 18 bottom).

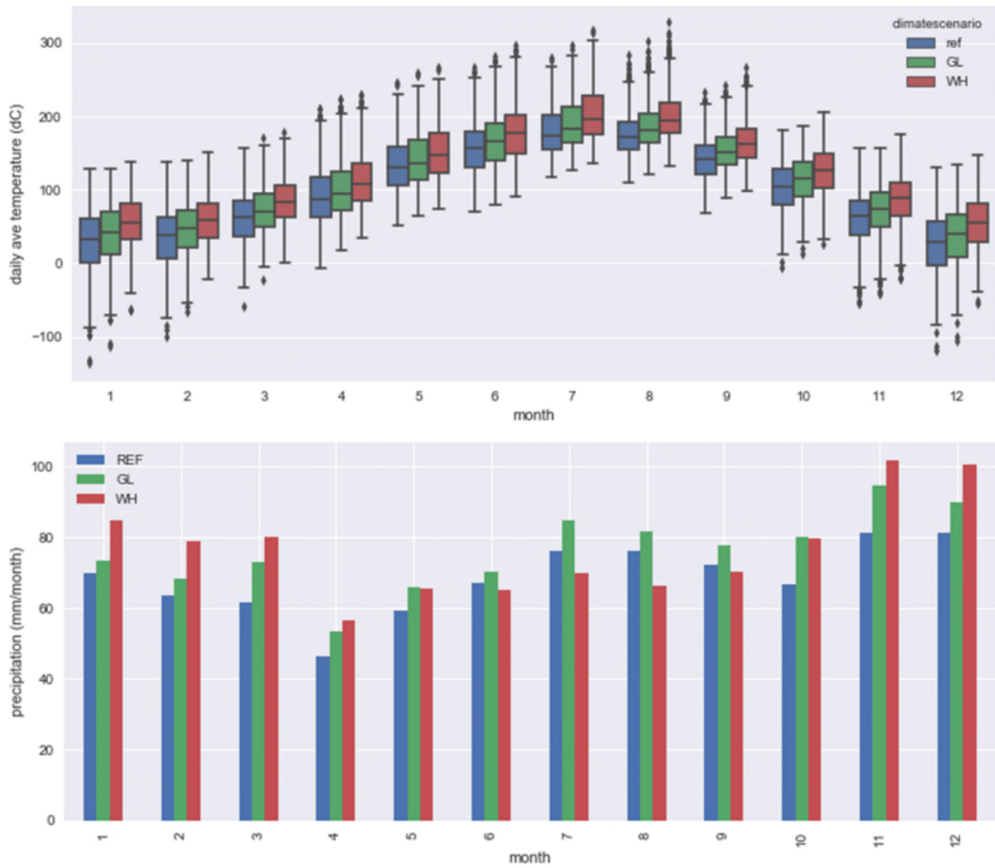


Figure 16. Difference in monthly average of daily temperature (top) and monthly cumulative precipitation (bottom) under different climate scenarios (reference, W_H and G_L), averaged over the period of 1989 - 2010.

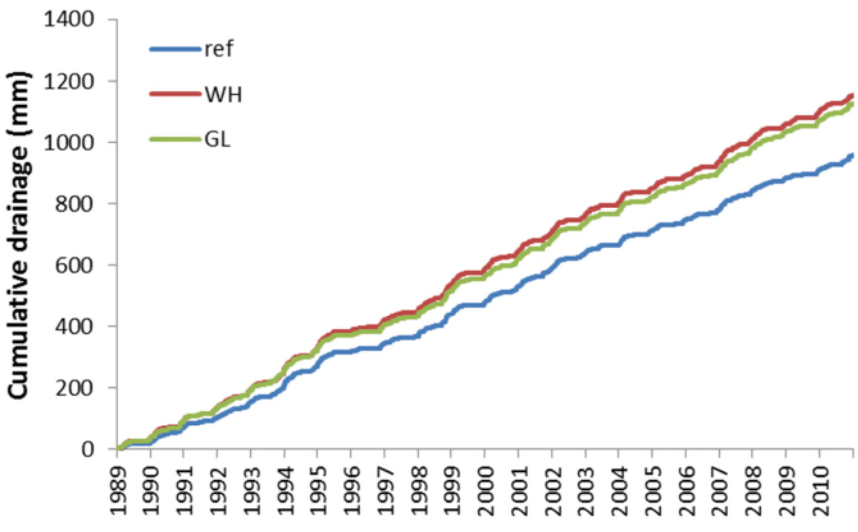


Figure 17. Simulated cumulative drainage for 22 years under reference climate of 1989 – 2010, and under W_H and G_L scenarios projected for year 2044 – 2065.

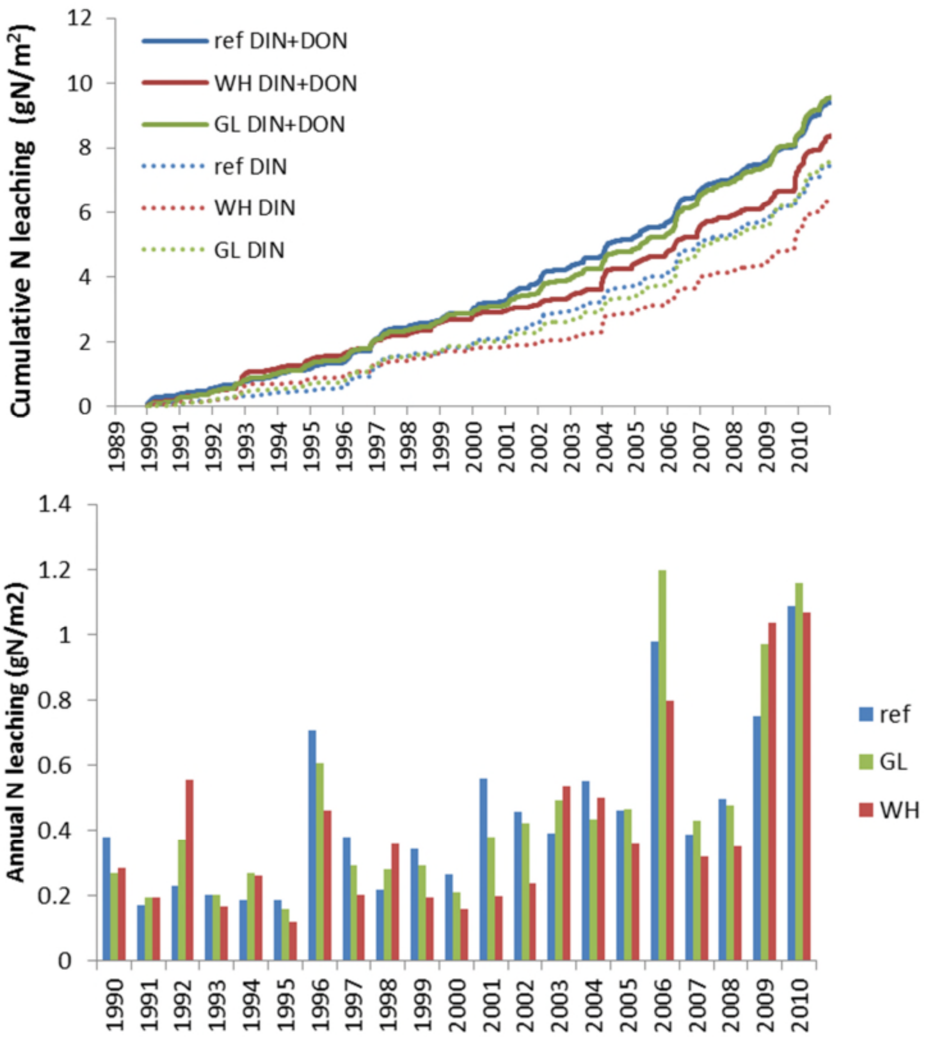


Figure 18. Simulated cumulative N leaching (top) and annual N leaching (bottom) under reference climate and future climate (WH and GL scenarios). N leaching was split into DIN (i.e. N-NH_4 plus N-NOx) and DON. The simulation was made for 22 years; 1989 – 2010 for the reference climate and 2044 – 2065 for WH and GL scenarios. The cumulative amount was calculated excluding the first year (i.e. since 1-January-1990) as the first-year leaching was strongly influenced by the input values.

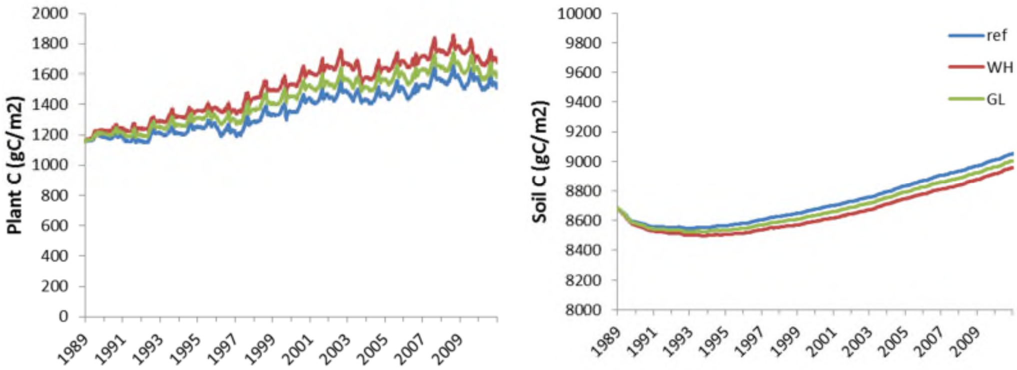


Figure 19. SImulated plant C pool and soil C pool for 22 years under reference climate of 1989 – 2010, and for W_H and G_L scenarios projected for year 2044 – 2065. Plant C includes both above-ground and below-ground biomass. SOil C includes surface microbe, active, slow, and passive pools.

4.5 Scenario study with elevated atmospheric N deposition

The model predicted that a higher level of atmospheric N deposition led to a higher DIN leaching rate (Figure 20). The predicted DON leaching rates hardly differed among different scenarios of N deposition.

High N input from atmosphere increased plant production and SOM pool (Figure 21). Although the former effect (i.e. increased plant production) increases N uptake by plant and therefore decreases DIN in top soil, the latter effect (i.e. increased SOM pool, which lead to higher mineralization) increases DIN in top soil. Furthermore, DIN in top soil increases directly due to elevated atmospheric N deposition. Altogether, DIN in top soil increases, leading to a higher leaching rate of DIN.

It should to be noted that N leaching under high N deposition was much higher than that under intermediate N deposition; leaching increased more than proportionally with the increase in deposition levels (i.e. more than 4.46/2.26). This is because the heath ecosystem becomes saturated with N when N deposition exceeds a certain level, and therefore the increased N deposition is directly leached out from the system without being taken up by plants or soil microbes.

The critical load of atmospheric N deposition is 1.5 gN/m^2/year for the dry heath habitat (H4030) of the Netherlands (van Dobben *et al.*, 2012). Similarly, the empirical critical loads for Europe-wide dry heath are 1 – 2 gN/m^2/year (Hettelingh & Bobbink, 2011). These levels of critical loads coincides roughly with the simulated tipping point of the N input level where the ecosystem becomes saturated and therefore N leaching rates increases more rapidly with increasing N input (Figure 22). In this light, the maintenance of low N deposition level is also important in order to keep the ability of ecosystem to retain nutrients in a sustainable manner.

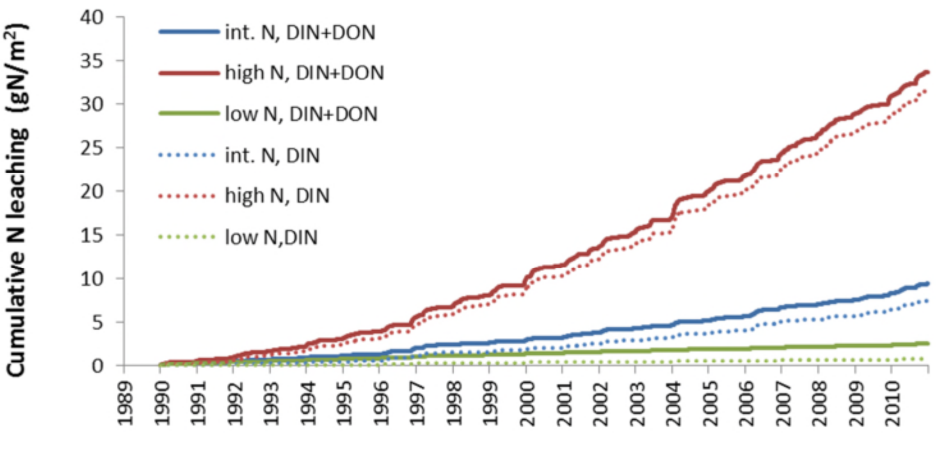


Figure 20. Simulated cumulative N leaching under high (4.46 g N/m^2/year), intermediate (2.26 g N/m^2/year), and low (0.48 g N/m^2/year) atmospheric N deposition levels. N leaching was split into DIN (i.e. N-NH_4 plus N-NOx) and DON. The simulation was made for 22 years, from 1989 to 2010. The cumulative amount was calculated excluding the first year (i.e. since 1-January-1990) as the first-year leaching was strongly influenced by the input values.

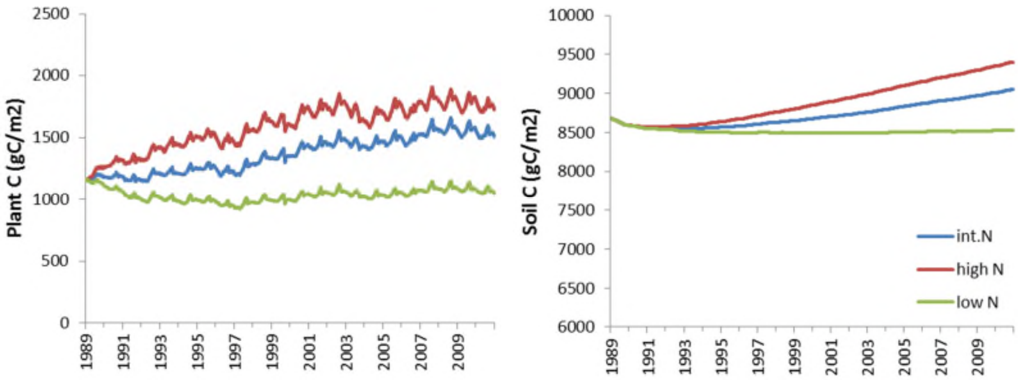


Figure 21. SImulated plant C pool and soil C pool for 22 years under high, intermediate, and low atmospheric N deposition levels. Plant C includes both above-ground and below-ground biomass. Soil C includes surface microbe , active, slow, passive pools.

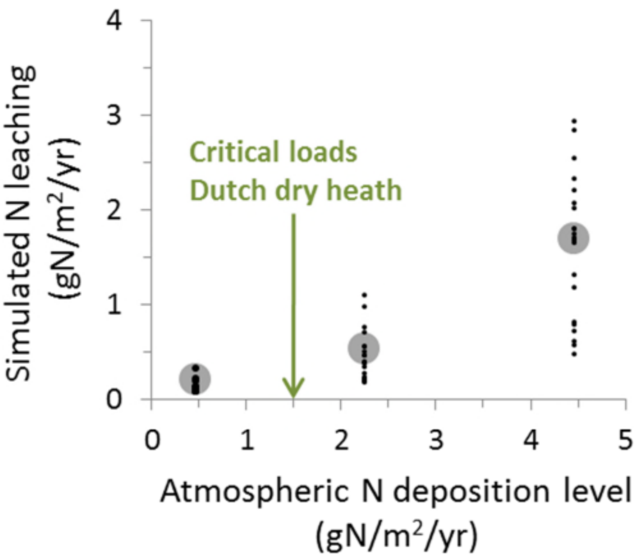


Figure 22. Simulated annual rates of N leaching (DIN + DON) from heathlands from 1990 to 2010, under three atmospheric N deposition scenarios. Large circles represent the mean annual N leaching rates, averaged over the 21 years. Arrow dipicts the critical load of N deposition for the habitat type dry heathland (H4030) of the Netherlands.

5 Synthesis

5.1 Levels and patterns of N leaching from heath ecosystem in Veluwe

Our field measurement revealed that the level of N leaching from the heath site in Veluwe was very low. Approximately 0.1 g N/m^2 of N was leached out in a year of 2017, which was less than 10 % of the N input that the heath received from the atmospheric N deposition that year. The observed low N leaching rate is in line with the finding of the Europe-wide study that dry heath strongly retains N in the system, associated with hardly any leaching losses (Hettelingh & Bobbink, 2011).

The N leaching rate was estimated by sampling the lysimeter leachate. Among the different methods to estimate N leaching rate, the use of lysimeter is one of the most direct and reliable methods (Wang *et al.*, 2012). There is a disadvantage in the use of the lysimeter leachate that the soil moisture conditions in the free-drainage lysimeters are slightly different from those in the ambient conditions. However, by using the hydrological model Hydrus 1D, we have checked that the difference in drainage levels between ambient conditions and free-drainage lysimeters was rather small. Furthermore, we have also measured N concentrations in soil water under ambient conditions using porous cup samplers, and confirmed that N concentrations at 50 cm depth were roughly the same inside and outside the lysimeters. Based on these findings, we can conclude that our estimates of N leaching levels are fairy trustworthy.

DON was the major form of leached N in our study. Since the DON concentration in soil water was higher in top soils than in deeper soils, and was low in rain water, it is expected that the DON was produced in the top layer (e.g. from SOM, from plant roots). Uptake of DON by plants and microbes seems to be insignificant.

What is striking is that there was almost no leaching of DIN, even in the winter period when biological activity is largely ceased, whereas DIN input from atmospheric deposition was constantly high throughout the year. This contrasts to observations in other studies that N leaching is usually higher in winter (Bhatti *et al.*, 2013). Based on the data of soil extraction, it was indicated that some amount of DIN (especially N-NH_4) was adsorbed to the soils until a quite deep depth (up to at least 45 cm) in our soils. Since the soil contains low concentrations of clay, the adsorption probably occurred at the surface of organic matter and/or metal (hydro)oxides. In addition, it was reported that a substantial amount of microbial immobilization can take place during winter in arctic soils (Schmidt *et al.*, 1999). Thus, besides adsorption, microbial immobilization in winter may be another important mechanism to reduce N leaching.

Our model and measurements indicated that the heath site at the Veluwe is currently not in an equilibrium state in terms of N balance, but is accumulating N. Our longer term (~30 years) model simulation indicated that, if N deposition and climate remain at current levels, the vegetation biomass will reach a new equilibrium state and level off in a few decades, yet soil N continues to increase and therefore soil N leaching rates will also gradually increase. The increase in N accumulation in the long run can be potentially hampered either by reduction in N deposition levels or by climate change (as in W_H scenario), yet only the former will likely reduce N leaching rates.

5.2 Plausibility of model simulation results

Our model of soil water using the hydrological model Hydrus 1D predicted the drainage of lysimeters very well. The dynamics of soil and heath vegetation predicted by the CENTURY model were associated with much more uncertainties. Predicted N leaching rates were much higher than measured N leaching rates. The mismatch exists especially in the leaching rates of DIN during winter, which was negligible in the measurements but substantial in the model. Even after trials of changing several key parameter values, the low levels of DIN leaching in winter could not be reconstructed by the model (results not shown). It seems that the model lacks a mechanism which enables to retain DIN during winter period in non-clay soils, such as adsorption to SOM and (hydro)oxides or microbial immobilization in winter periods.

Nevertheless, our model predicted N leaching and N accumulation in the same order of magnitude as the measurements. Therefore, the model can be used as a starting point for scenario studies of future conditions.

5.3 Factors influencing soil N retention and N leaching

The amount of N leaching is obviously affected by the amount of N input to the system. Elevated N deposition leads to higher N leaching, as we demonstrated in the scenario studies with different levels of atmospheric N deposition. However, the existing field studies indicated that the ratio between N input and N leaching differ largely between sites (Figure 23). It seems that open vegetation structure (bare soil/moss) and grasslands have poorer soil N retention than woody vegetation (heath/shrubland/forest), but the pattern was not always consistent. Even under very high levels (> 4 g N/m^2/year) of N input, some ecosystems manage to retain a large part of N input in the system.

Manipulation experiments with soil columns help to explore the factors which differentiate N retention among sites. A number of experimental studies indicated that soil N retention is related to the type of plant community and/or microbial properties (De Deyn *et al.*, 2009, Vauramo & Setälä, 2010, De Vries *et al.*, 2011, Grigulis *et al.*, 2013). It is shown that fungi-dominated grasslands retain N better than bacteria-dominated grasslands (Vauramo & Setälä, 2010, De Vries *et al.*, 2011). However, the bacteria are probably not the direct cause of high soil N retention but rather the results of low N availability (De Vries *et al.*, 2011). Furthermore, species-rich plant communities had lower N leaching rates, which was due to presence of specific functional groups such as forbs and N fixers (De Deyn *et al.*, 2009).

Härdtle *et al.* (2007) reported that the effects of nature management on N leashing were much larger than that of atmospheric N deposition for dry heathlands. This is because the dry heathlands have high N retention and therefore their N leaching rates are highly influenced by its internal turnover processes rather than input. N leaching rates increased after the application of management measures, particularly after choppering and sod-cutting.

Further research is needed to examine factors which have causal relationships with soil N retention, and to quantify their impacts on N leaching. Such knowledge is indispensable to improve model equations and parameter values to properly predict N leaching from natural ecosystems.

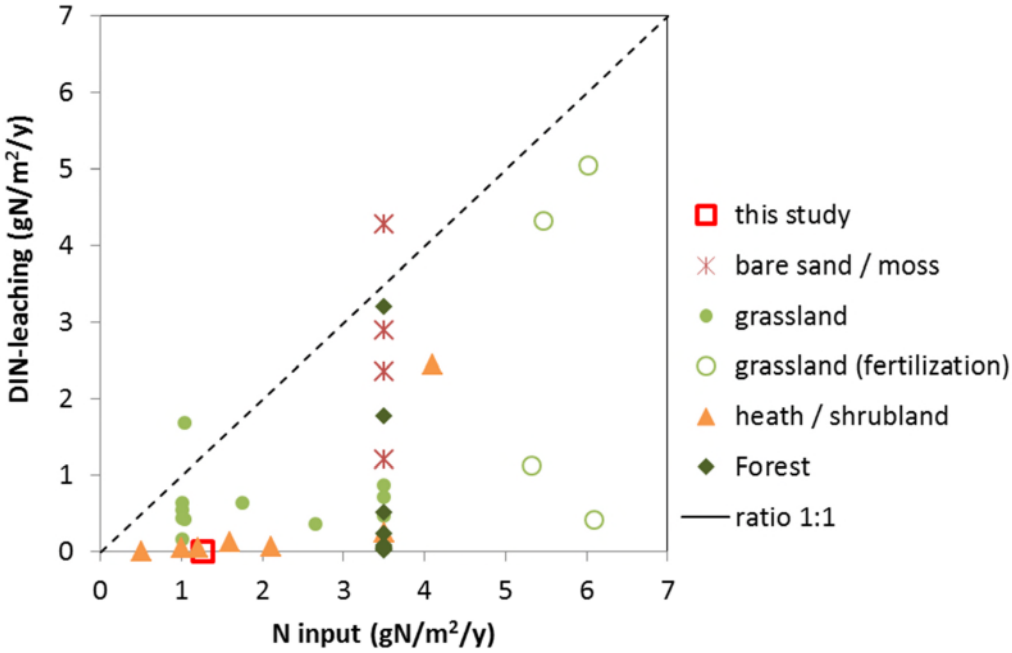


Figure 23. Leaching levels of inorganic N versus N input levels, taken from different field studies in Europe. The symbols depict vegetation types. N input consists of atmospheric N deposition and N fertilziation (only for the category 'grassland (fertilization)'). Source: (Stuyfzand, 1991, ten Harkel et al.*, 1998, Jones* et al.*, 2005, Beier* et al.*, 2009)*

5.4 Uncertainty in predicting effects of climate change on N leaching

Our model scenario studies showed that future climate will increase groundwater recharge from the heath ecosystems, mainly due to increased recharge in winter. Whether the increase in recharge leads to an increased N leaching was not very certain. Our model predicted a slightly decreased N leaching under future climate, yet the difference in N leaching rate between the future climate and the reference climate was highly sensitive to several key parameter values (e.g. temperature reduction terms on SOM decomposition and plant production, moisture reduction terms on SOM decomposition and plant production, fraction of soil N which is prone to leaching as DON, C:N ratio of different soil pools, maximum production rate of plants). This is because the amount of N leaching is an 'end product' of many ecosystem processes, which are influenced by N input via atmospheric deposition, SOM accumulation and turnover, uptake of N by plants, and water fluxes. Furthermore, many of the effects of future climate conditions are counteracting in terms of their effect on the 'end product'. See diagram in Figure 24 for the schematic representation of causal chains of different processes on N leaching, and how these processes are influenced by future climate. This indicates that subtle changes in plant properties, soil microbial properties, and geochemical variables can change the direction of future climate effects on N leaching rates.

Effects of changing climate on N leaching can be directly measured by field-scale manipulation experiments. In the old heath ecosystems of Oldebroek, a climate manipulation system was installed for a long term since 1999 (Kopittke *et al.*, 2012, Kröel-Dulay *et al.*, 2015). The 'drought' experiment reduces summer rainfall (April - July) using rain shields which close with rain sensor. On annual average, the drought plots receive 19 % less rain compared to the control plots. The 'warm' plots receive passive warming at night by automatically-covering curtains that reflected outgoing radiation. The warmer plots are on annual average 0.3˚C warmer than the control plots. N leaching rates were comparable between control and drought plots, with slight decrease in DON leaching by the drought treatment (Figure 25; Tietema, unpublished). The warming treatment significantly increased N leaching. It almost doubled NO_3 leaching, whereas the effect on NH_4 and DON leaching was limited (Figure 25; Tietema, unpublished). Since KNMI's W_H scenario expects less rain in summer and higher temperatures, one could extrapolate the results of Oldebroek to a prediction that N leaching will increase under future climate. However, since during the measurement period the heath site of Oldebroek was much more saturated with N due to high atmospheric deposition, having ca. 17 times more N leaching than our study site under ambient climate condition, we cannot simply project the pattern of Oldebroek to our site. Furthermore, as shown in Figure 24, both higher temperature and less rain in summer can theoretically lead to either higher or lower N leaching. A promising way forward is to use empirical data of climate manipulation experiment such as Oldebroek to validate the model, especially for the key processes for N leaching such as N mineralization rates, plant N uptake, concentrations of DIN and DON in soils. In this way, the model will be able to make more reliable predictions of climate effects on N leaching.

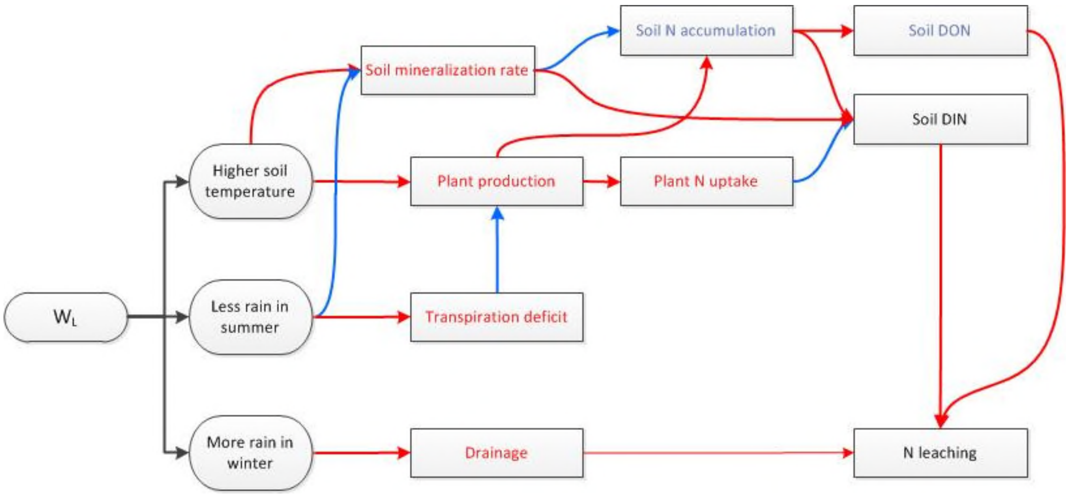


Figure 24. Causal chains of how future climate scenario 'W_L' influences N leaching. Red arrows depict positive effects, whereas blue arrows depict negative effects. Red (or blue) letters in the boxes means that the simulated value under the W_H scenario was higher (or lower) than that under reference climate on our scenario study. Black letters in the boxes means that the simulated values were not largely different between W_H scenario and reference.

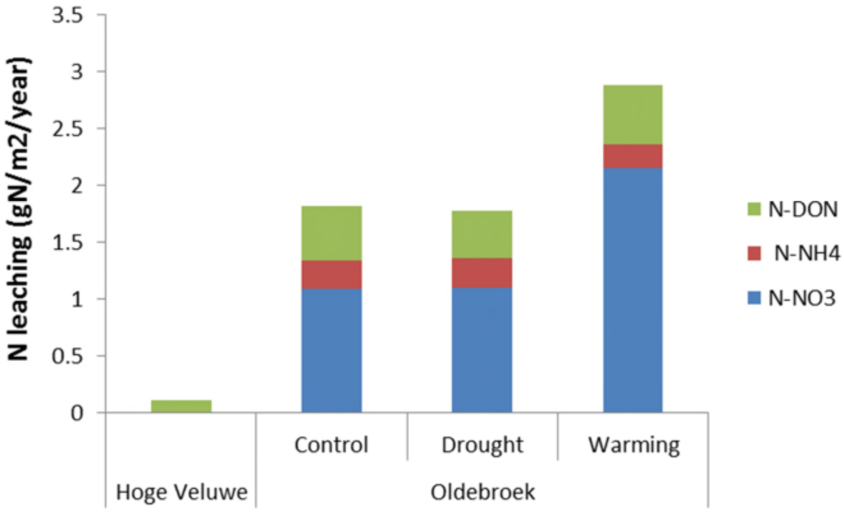


Figure 25. Difference in N leaching rates in control, drought, and warming plots in old heath ecosystems of Oldebroek, the Netherlands (Tietema, unpublished). The average values of 7 years data, samples on 122 occations between 1999 and 2011. Maximum 7 replicas per treatment. The drought plots receives less rain in summer.

5.5 Implications for nature management policies

Our study showed that increased atmospheric N deposition will increase N leaching. The heath vegetation acts as a N sink to some extent, by capturing a part of the N input by atmospheric deposition in vegetation and soil before it leaches to the groundwater. However, the more atmospheric deposition increases (above ca. 2-3 g N/m^2/year; Figure 22, Figure 23) and thus saturating the ecosystem with N, the lower the ecosystems importance becomes as an N sink. Furthermore, it should be noted that increased N atmospheric deposition will, even if it does not immediately lead to an increase in N leaching, accumulate N in the ecosystem. This will have many consequences in a long term, such as accelerated N turnover and increased N availability, possibly followed by encroachment of undesired plant species. Thus, in order to reduce N leaching and also to sustainably manage the ecosystem, it is important to continue the effort of reducing the level of atmospheric N deposition.

Under predicted future climate conditions, groundwater recharge will increase under heath ecosystems, mainly due to increased recharge during winter periods. The effects of the future climate on N leaching may be limited, and could be either positive or negative, depending on the balance between multiple (and possibly counteracting) impacts on plant growth, soil accumulation, and soil nutrient transformation. The climate effects on N leaching will also highly depend on plant properties, soil microbial and geochemical properties, as well as the N balance of the ecosystem under ambient climate conditions. Thus, it is not possible to deduce a generic strategy for climate adaptation to reduce N leaching in heathlands in the future. Same holds true for other types of sandy groundwater-dependent systems, To enable site-specific predictions of N leaching under future climate, it is necessary to improve process-based models by validating it with extensive datasets of climate manipulation experiments, preferably on different types of ecosystems.

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Appendix I Soil physical characteristics

Two soil cores of 100 cm^3 were sampled from three depths in the heathland ecosystem of the Veluwe. Soil water retention characteristics were measured in a sand box manipulation experiment, ranging from pF 0 to 2.0 (i.e. drying curve). Parameter values of Van Genuchten equation were obtained with non-linear fitting procedures.

	θres (cm^3/cm^3)	**θsat (cm^3/cm^3)**	**α (1/cm)**	***n*** **(-)**
1st layer (~10 cm depth)	0.0213502	0.446497	0.0305175	1.4826
2nd layer (~25 cm depth)	0.0235844	0.479875	0.0295075	1.70453
3rd layer (~50 cm depth)	0.0166217	0.461688	0.050665	2.28071

Appendix II Seasonal dynamics of N concentrations in water samples

SEASONAL DYNAMICS OF NITROGEN CONCENTRATIONS IN RAIN WATER

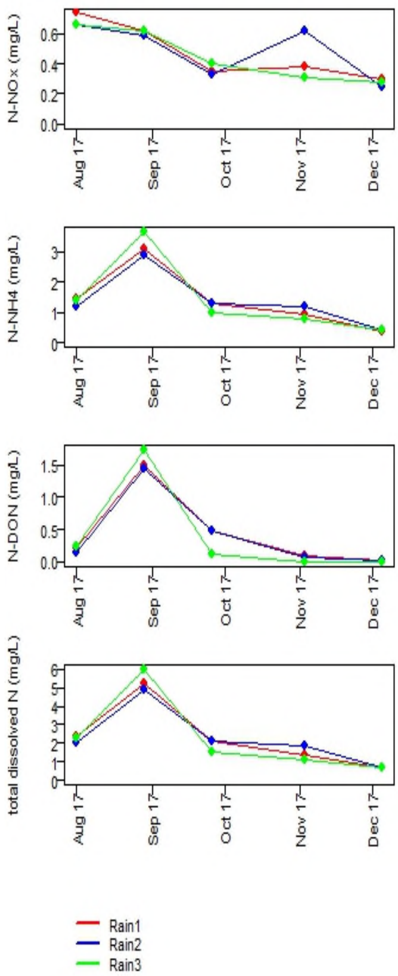

Figure 26. Seasonal fluctuation of nitrogen concentrations in rain water. Note that the filters were attached with algae in September and October, causing possible contamination of the sampled water in these months

SEASONAL FLUCTUATION OF N CONCENTRATIONS IN LYSIMETER LEACHATES

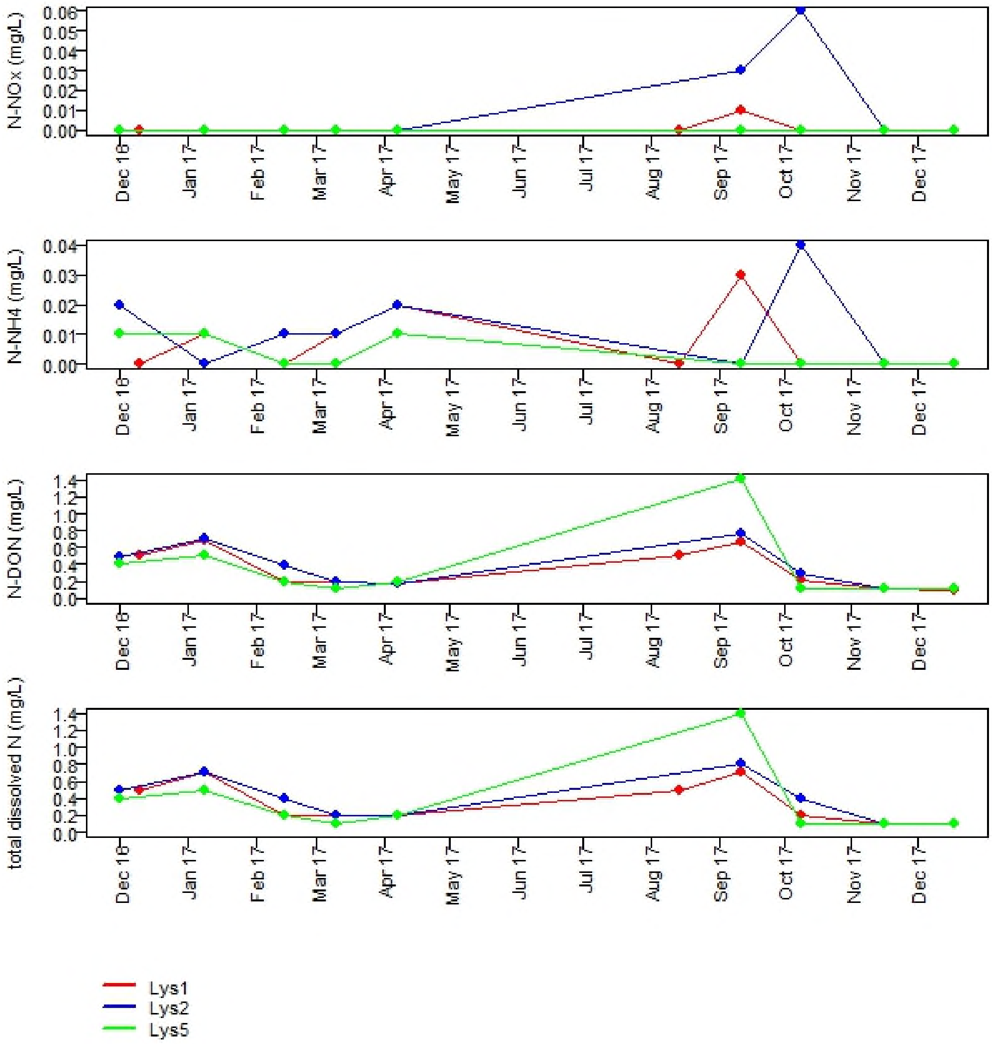

Figure 27. Seasonal fluctuation of N concentrations in lysimeter leachates. Note that the concentrations of N-NOx and N-NH_4 were very low all through the year, mostly falling under the detection limits (i.e. the detection limits for the analysis was: 0.04 mg/L for N-NH_4 , 0.03 mg/L for N-NOx and 0.3 mg/L for total dissolved N).

SEASONAL FLUCTUATION OF N CONCENTRATIONS IN SOIL WATER

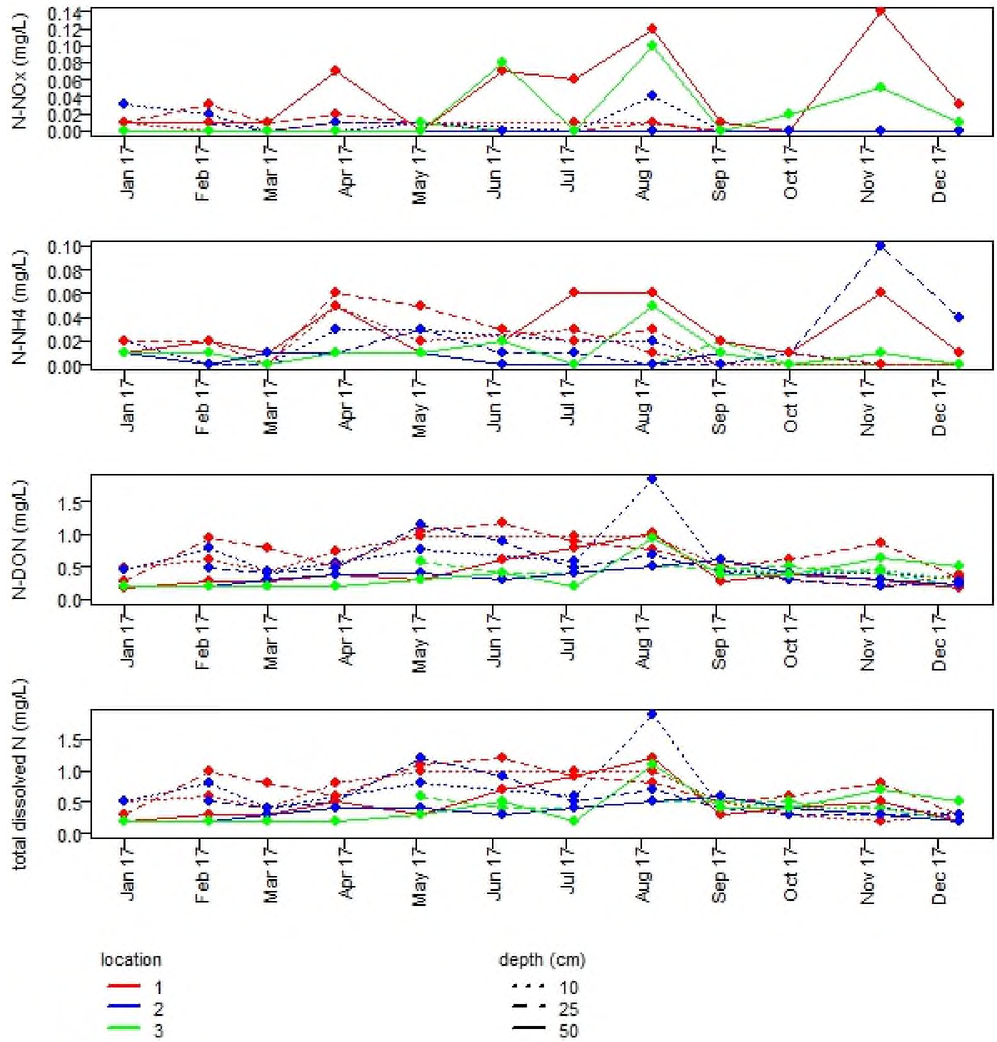

Figure 28. Seasonal fluctuation of N concentrations in soil pore water.

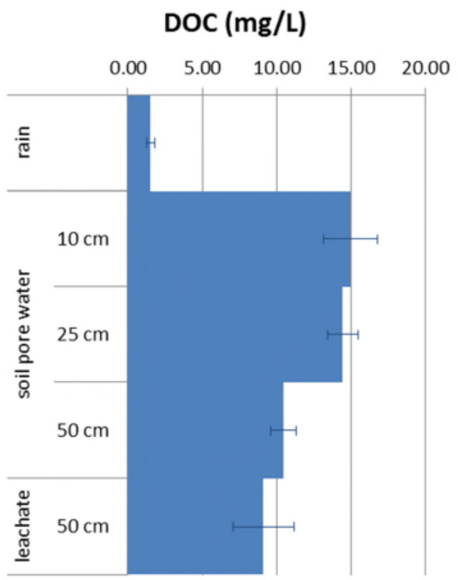

Figure 29. Concentration of dissolved organic carbon in rain water, soil pore water at three different depths, and in lysimeter leachates. Average ± SE over the entire measurement period and three locations are shown.

Appendix III Atmospheric N deposition from external data sources

Since our measurement of atmospheric N deposition was limited to a few months, we also referred to the data of atmospheric N deposition collected by RIVM in nearby locations.

WET DEPOSITION IN SPEULD

Wet deposition of NH_4 and NO_3 was measured in the monitoring station Speuld, located ca. 30 km north-west of our study site (Figure 30). It has been validated for the dataset until year 2016. The average concentrations of the year 2016 were 0.81 mg N/L for NH_4 and 0.35 mg N/L for NO_3.

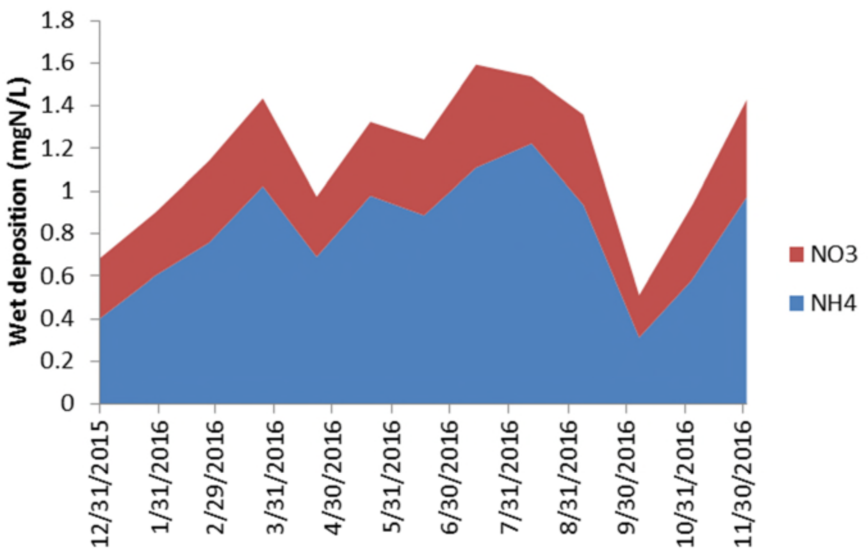

Figure 30. Wet deposition of nitrate and ammonium in 2016, measured in the monitoring station of RIVM in Speuld. The samples were collected using wet-only rain collectors (Eigenbrodt NSA 181 KHT).

Appendix IV Raw data of pools and fluxes

Table 3. Soil N, P, and C pools in different compartments of heath ecosystem in Hoge Veluwe. Dry mass per square meter and concentrations of each element are also shown. For soil compartments, bulk density is shown. See section 2.4 for the calculation method.

	g/cm3	g/m^2	g/kg	g/kg	g/kg	g/m^2	g/m^2	g/m^2
	bulk density	mass	[N]	[P]	[C]	N pool	P pool	C pool
canopy		628.2	10.9	0.35	523.7	6.8	0.2	328.9
branches		575.3	7.9	0.18	513.0	4.6	0.1	295.1
root		2092.5	9.8	0.32	352.3	20.5	0.7	737.3
litter		184.9	13.7	0.25	534.7	2.5	0.0	98.8
soil 0-20 cm	1.47	294820.0	1.0	0.10	29.4	292.9	28.5	8667.7
soil 20-30 cm	1.43	142752.5	0.7	0.09	21.6	98.5	12.5	3088.2
soil 30-50 cm	1.47	294685.0	0.3	0.08	10.3	90.4	22.1	3040.2

Table 4. Concentrations of dissolved N at different depth. Average values of three locations are shown. (See Figure 9 for values for each location.) Free N-NH_4 and N-NOx are the $CaCl_2$-extractable N-NH_4 and N-NOx; adsorbed N-NH_4 and N-NOx are the differences between KCl extractable and $CaCl_2$ extractable N-NH_4 and N-NOx; DON is total dissolved inorganic N in $CaCl_2$ extracts minus $CaCl_2$-extractable N-NH_4 and N-NO_3 .

depth	[mg N/kg] free N-NH_4	[mg N/kg] free N-NOx	[mg N/kg] adsorbed N-NH_4	[mg N/kg] adsorbed N-Nox	[mg N/kg] DON	[mg N/kg] total dissolved N
10 cm	1.07	0.07	0.33	0.07	9.20	10.73
25 cm	1.00	0.07	0.33	0.00	4.93	6.33
45 cm	0.77	0.10	0.37	0.07	1.80	3.10

Table 5. In- and outflux of N. High estimate of deposition.N influx via atmospheric deposition was calculated as high estimate (using the average concentrations of N in all measurements) and as low estimate (using the average concentrations of N except the samples in september and october, which was possibly contaminated). N leaching is the average value of 3 free-drainage lysimters, measured for the period of 10 dec 2016 - 17 dec 2017. See section 2.4 for the calculation method.

		$g N/m^2/year$	$g N/m^2/year$	$g N/m^2/year$	$g N/m^2/year$
		N-NHx	N-NOy	N-DON	Total
Influx	deposition (high estimate)	1.31	0.43	0.40	2.14
	deposition (low estimate)	0.84	0.43	0.08	1.35
Outflux	leaching	0.00	0.00	0.10	0.10

Appendix V Forest module specifications

EQUATIONS FOR TREE DYNAMICS IN CENTURY MODEL

Tree growth

Living biomass of trees is divided into 5 components: leaves, fine roots (<2mm diameter), fine branches, large woods (>10cm diameter), and coarse roots. Furthermore, there are three additional woody debris components: Dead fine branches, dead large woods, and dead coarse roots.

Potential production of trees ($Ptree_P$, g C/m^2/day) is calculated as potential gross production of trees minus maintenance respiration:

$$Ptree_P = \min\left(\max\left(0, \frac{GP_P}{ratbioC} - sumrsp\right), fcmax\right)$$

where GP_P is the potential gross production of trees (g biomass/m^2/day), *ratbioC* is the ratio of biomass (g dried mass) over biomass C (g C), *sumrsp* is the sum of maintenance respiration from all tree parts (g C/m^2/day), and *fcmax* is the maximum net production of trees (g C/m^2/day). To prevent negative net primary production, we restricted that the respiration does not exceed gross production.

The potential gross production of trees ($GP_{P,}$ g biomass/m^2/day) is computed as follows:

$$GP_P = prdx2 \cdot T_P \cdot M_P \cdot laprod$$

where *prdx2* is the maximum gross forest production rate (g biomass/m^2/day), T_P is the temperature reduction term (fraction between 0 and 1; explained in the section for grassland growth model), M_P is the moisture reduction term (fraction between 0 and 1; explained in the section for grassland growth model), *laprod* is the shading effect of LAI on tree production (fraction between 0 and 1). *labrod* is defined as:

$$laprod = 1 - \exp\left(laitop \cdot \max\left(2, LAI\right)\right)$$

where LAI is the leaf area index (i.e. leaf area per unit ground surface area, m^2/m^2), *laitop* is the parameter determining the relationship between LAI and forest production.

LAI is approximated based on large wood C pool as:

$$LAI = maxlai \cdot \frac{TreeC_3}{klai + TreeC_3}$$

where *maxlai* is the theoretical maximum leaf area index achieved in a mature forest (m^2/m^2), $TreeC_3$ is the C content of large wood pool (g C/m^2), and *klai* is the size of $TreeC_3$ at which half of theoretical maximum leaf area (*maxlai*) is achieved. Since heath trees don't form large wood (>10cm diameter), we used the C content of fine blanch (C_2) to calculate LAI.

The maintenance respiration of trees (*sumrsp,* g C/m^2/day) is calculated according to Ryan (1991) as:

$$sumrsp = \sum_{i=0}^{4}\left(\frac{0.0106}{4}\cdot\frac{12}{14}\cdot 24\cdot\exp\left(\frac{\ln(2)}{10}\right)\cdot tave_i\cdot frlive_i\cdot TreeN_i\cdot 1.1\right)$$

where i is the ith pool of the tree, tve_i is the temperature relevant for the pool i (˚C), $frlive_i$ is the fraction of the pool i which is counted for respiration, and $TreeN_i$ is the N content of tree pool i (g N/m^2). Air temperature was used for $tave_i$ of pool 0 (leaf), 2 (fine branch), and 3 (large wood), while soil temperature was used for $tave_i$ of pool 1 (fine roots) and 4 (coarse roots). With the assumption that only sapwood part of the tree respire C, $frlive_i$ was calculated for fine branches (i=2), large wood (i = 3) and coarce roots (i = 4) as follows:

$$frlive_i = \frac{spak}{spak + TreeC_i}$$

where *spak* is the parameter which controls the ratio of sapwood to total stem wood, expressed as g C/m^2. spak was set to be 1500, the values for 'BFGH' in CENTURY ver.4. For other pools (i.e. i = 0, 1), $frlive_i$ was set to be 1.

The maximum net production of trees (*fcmax*, g/m^2/day) is calculated as:

$$fcmax = prdx3\cdot T_P\cdot M_P\cdot laprod$$

where *prdx3* is the maximum net forest production rate (g C/m^2/day).

The allocation of newly-assimilated biomass to each tree compartment i is given as forest-specific parameters $fcfrac_i$, and can be separately given for young and old forests.

Actual growth of trees is calculated based on total available N (soil mineral N + symbiotic N fixation), in an identical manner as for the herbaceous plants.

Tree death

Death rates of plant pools are noted as $dtree_{i,m}$ (fraction/month), where i is the pool (i=1and m is the month (m=1, 2, ...12). The death rates are month-specific only for leaves in CENTURY.

We estimated death rates of heathland ecosystems based on literature. The death rate of the leaves ($dtree_{0,m}$) were estimated based on observed leaf turnover rate of *Erica tetralix* in the Netherlands (Aerts & Berendse, 1989) as: 0.01 from October to February, 0.03 from April to August, and 0.2 for September.

The death rate of fine roots were estimated from the measured annual root turnover rate of *Erica tetralix* using soil core sampling and minirhizotron (Aerts *et al.*, 1989), divided by 12 (i.e. $dtree_{1,m}$ = 0.11/month).

Since we have no data of turnover rate of fine branch and large wood, it was approximated by the annual production divided by biomass (as suggested in the 'parameterization workbook' of CENTURY ver. 4). The observed values of annual production and biomass of *Erica tetralix* (Aerts, 1989) was used to compute $dtree_{2,m} = dtree_{3,m} = 0.027$.

There was no empirical data available for coarse root. As suggested in the 'parameterization workbook' of CENTURY ver.4, we used the same value as that of large wood (i.e. $dtree_{4,m}$ = 0.027)

Decomposition of woody litter

Carbon flows between different pools are formulated according to CENTURY ver. 4 as:

$$\frac{dC_i}{dt} = -k_i \cdot C_i + \sum_j f_{ji} \cdot k_j \cdot C_j$$

where Ci is amount of C in pool i (g C/m^2), ki is the decomposition rate of the pool i (fraction/day), and fji is the flow rate of C from pool j to i (fraction). ki is a multiple of maximum decomposition rate (*kmax*i), reduction factor of soil moisture (*Md*), and reduction factor of temperature (*Td*). Maximum decomposition rates of woody litters are: $kmax_9$=decw1 = 4.4/365 (day^{-1}), $kmax_{10}$=decw2 = 4.4/365 (day^{-1}), $kmax_{11}$=decw3 = 9.2/365 (day^{-1}).

For the forest specific C pools (C_9, C_{10}, C_{11}), the flow rates are defined as:

$$f_{9,4} = (1 - L_{FB}) \cdot 0.4$$
$$f_{9,6} = L_{FB} \cdot 0.7$$
$$f_{9,8} = (1 - L_{FB}) \cdot 0.6 + L_{FB} \cdot 0.3$$
$$f_{10,4} = (1 - L_{LW}) \cdot 0.4$$
$$f_{10,6} = L_{LW} \cdot 0.7$$
$$f_{10,8} = (1 - L_{LW}) \cdot 0.6 + L_{LW} \cdot 0.3$$
$$f_{11,5} = (1 - L_{CR}) \cdot 0.45$$
$$f_{11,6} = L_{CR} \cdot 0.7$$
$$f_{11,8} = (1 - L_{CR}) \cdot 0.55 + L_{CR} \cdot 0.3$$

where L_{FB}, L_{LW}, and L_{CR} are lignin content (fraction in relation to C content) in fine branches, large woods, and coarse roots, respectively. Following Kirschbaum and Paul (2002), the coefficient of lignin effects on decomposition rate of structural pools were changed from -3 to -5.

INPUT AND PARAMETER VALUES FOR HEATH ECOSYSTEM SIMULATION WITH CENTURY

Table 6.

	value	Reference	Note
Initial values			
Soil total C (g C/m^2)	8668	This study	Values of 0-20 cm depth were used. Total C was further divided into different pools with the following ratios: 0.030261 (act), 0.598231(slow), 0.371508 (passive) (as in 'mixed forest' of CENTURY ver.4)
Soil N:C (ratio)	0.0338	This study	Values of 0-20 cm depth were used
Clay (fraction)	0.00665	Fujita *et al.* (2013a)	average of 4 sites in Veluwe
Silt + clay (fraction)	0.022575	Fujita *et al.* (2013a)	average of 4 sites in Veluwe
θres (cm^3/cm^3) (1st, 2nd, and 3rd soil layer)	0.0213502, 0.0235844, 0.0166217	This study	
θsat (cm^3/cm^3) (1st, 2nd, and 3rd soil layer)	0.446497, 0.479875, 0.461688	This study	
α (1/cm) (1st, 2nd, and 3rd soil layer)	0.0305175, 0.0295075, 0.050665	This study	
n (-)(1st, 2nd, and 3rd soil layer)	1.4826, 1.70453, 2.28071	This study	
Plant C(g C/m^2)	leaves 80, fine roots 737, fine branches 337	This study	Biomass content of April, multiplied with C concentrations of each organ in summer. With an assumption that 19 % of above-ground biomass was leaves (based on moisture content of each organs and whole biomass)
Plant N (g N/m^2)	leaves 2.2, fine roots 16.9, fine branches 5.2	This study	Biomass content of April, multiplied with N concentration of each organ in summer.
herbaceous litter C (g C/m^2)	24	CENTURY parameterization workbook	20% of corresponding living pools (i.e. leaves)
Dead tree C (g C/m^2)	100	CENTURY parameterization workbook	20 % of corresponding living pools (i.e. branches)
nNH$_4$ (g N/m^2) of top soil	0.314	This study	calculated from $CaCl_2$-extractable N-NH_4 at 10cm depth (mg N/kg soil) and bulk density

nNO_3 (g N/m²) of top soil	0.020	This study	calculated from $CaCl_2$-extractable N-NOx at 10cm depth (mg N/kg soil) and bulk density
Mineral N (g N/m²) of second layer	0.414	This study	calculated from sum of $CaCl_2$-extractable $N\text{-}NH_4$ and N-NOx (average over 25 cm and 45 cm depth) and bulk density
Mineral N (g N/m²) of third layer	0	Arbitrary chosen	
Dissolved organic N (g N/m²) of second layer	1.442	This study	calculated from $CaCl_2$-extractable DON (average over 25 cm and 45 cm depth) and bulk density
Dissolved organic N (g N/m²) of third layer	0	Arbitrary chosen	
Parameter values			
plant CN ratio ('cerfor')	leaf 36, fine roots 36, branches 65	This study	
lignin fraction ('wdlig')	leaf 0.2, fine roots 0.2, fine branch 0.4, large wood 0.4, coarse roots 0.4	values of 'BFGN' in CENTURY ver.4	
death rate leaf ('leafdr') (fraction/day)	for each month: [0.01, 0.01, 0.03, 0.03, 0.03, 0.03, 0.03, 0.03, 0.2, 0.01, 0.01, 0.01] * 1/30.	(Aerts & Berendse, 1989)	Based on leaf turnover of Erica
death rate fine root ('wooddr(1)', fraction/day)	0.026	(Beier *et al.*, 2009)	Based on root turnover of Calluna-dominated heath
death rate fine branches ('wooddr(2)', fraction/day)	0.027	(Aerts & Berendse, 1989)	Based on stem annual production and biomass of Erica
allocation of newly-assimilated biomass to each tree compartment ('$fcfrac_1$' –' $fcfrac_5$')	leaves 0.4312, fine roots 0.23, fine branches 0.3388, large woods 0, coarse roots 0	(Aerts, 1990) (Aerts & Berendse, 1989)	based on the ratio of above- and below-ground annual production of Calluna and the ratio of annual production of leaves and supporting tissues
maximum gross forest production rate (*prdx2)(* g biomass/m²/day)	1200 /30	parametrization workbook of CENTURY ver.4	Common values are 1200 – 1500 g/m²/month
maximum net forest production rate (*prdx3*) (g C/m²/day)	300 /30	parametrization workbook of CENTURY ver.4	Common values are 300-400 g C/m²/month
theoretical maximum leaf area index achieved in a	3.7	(Asner *et al.*, 2003)	mean + 1xSD of the metaanalysis data of

mature forest (*maxlai*) (m^2/m^2)			shrubland
size of branches at which half of theoretical maximum leaf area,*maxlai*, is achieved (*klai*) (g C/m^2)	600		the same value as prdx3
parameter determining the relationship between LAI and forest production (*laitop*)	-0.5	Default value in CENTURY ver. 4	
C/N ratio of SOM pools	10 -20 (surface microbe), 8-16 (active), 12-40 (slow), 8-20 (passive)	parametrization workbook of CENTURY ver.4	for high-C/N forest soils
Coefficients for temperature response for decomposition (*tempp*)	[0., 0.08, 0.095]	CENTURY ver.4	
Coefficients for temperature response for decomposition (*ppdf*)	([22., 42., 1., 3]),	CENTURY ver.4	Values for Northern hardwood, except for the fourth value (which was lowered from 3.5, in order to weaken temperature effect at very low temperature)
Coefficients for pH effect on decomposition (KpH. m)	([4640, 1])	(Walse *et al.*, 1998)	Geometric means of values for Holocellulose and lignin
Proportion of species associated with symbiotic N fixers (‘*nfxrat*’)	0		No symbiotic N fixation was assumed
fraction of active C pool which is dissolved and prone to leaching ‘*max_fdoc*’	0.001	CENTURY ver.4	Calculated to be similar to the original DON leaching rate of century ver. 4: decomposition rate of active pool (0.02/day) * leaching rate from decomposed active pool (0.05)