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Estimation of nitrogen budgets for contrasting catchments at the landscape scale

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Abstract

A comprehensive assessment of nitrogen (N) flows at the landscape scale is fundamental to understand spatial interactions in the N cascade and to inform the development of locally optimised N management strategies. To explore this interactions, complete N budgets were estimated for two contrasting hydrological catchments (dominated by agricultural grassland vs. semi-natural peat-dominated moorland), forming part of an intensively studied landscape in southern Scotland. Local scale atmospheric dispersion modelling and detailed farm and field inventories provided high resolution estimations of input fluxes. Agricultural inputs (i.e. grazing excreta, organic and synthetic fertiliser) accounted for most of the catchment N inputs with 80 % in the grassland and 57 % in the moorland catchment, while atmospheric deposition made a significant contribution, particularly in the moorland catchment with 38 % of the N inputs. The estimated catchment N budgets highlighted areas of key uncertainty, particularly N_2 emissions from denitrification and stream N export. The resulting N balances suggest that the study catchments have a limited capacity to store N within soils, vegetation and groundwater. The "catchment N retention", i.e. the amount of N which is either stored within the catchment or lost through atmospheric emissions, was estimated to be 3 % of the net anthropogenic input in the moorland and 55 % in the grassland catchment. These values contrast with regional scale estimates: catchment retentions of net anthropogenic input estimated within Europe at the regional scale range from 50 % to 90 % with an average of 82 % (Billen et al., 2011). This study emphasises the need for detailed budget analyses to identify the N status of European landscapes.

1 Introduction

Human activities dominate the global nitrogen (N) budget by adding reactive forms of nitrogen (N_r) to the environment (Galloway et al., 2004). The main forms of anthropogenic N_r are reduced (e.g. NH_3, NH_4^+), oxidised (e.g. NO_2, N_2O, NO_3^-) and organic

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forms of N (e.g. urea). Between 1995 and 2005 alone, the anthropogenic production of N_r increased by 20 % which is largely due to agricultural activities (Galloway et al., 2008). The environmental consequences of N_r input can be significant, such as a loss of biodiversity in terrestrial and aquatic ecosystems through eutrophication and acidification (Vitousek et al., 1997). Nitrogen balances as indicators of environmental pressure have recently been developed and applied at various scales (e.g. de Vries et al., 2011), ranging from the farm and field level (e.g. Ammann et al., 2009; Schröder et al., 2003) to the regional catchment (e.g. Billen et al., 2009; Howarth et al., 1996) and global scale (e.g. Bouwman et al., 2005; Seitzinger et al., 2005).

The assessment of budgets at the landscape scale is a critical part of quantifying the impact of disturbance on nutrient cycling (McDowell and Asbury, 1994). A landscape is defined as a spatially heterogeneous area that includes interacting ecosystems and extends from hectares to many square kilometres (Turner and Gardner, 1994). Nitrogen is transported between those ecosystems by atmospheric, hydrological and human transfers (Cellier et al., 2011). Fluxes of N_r at the landscape scale are particularly relevant as both management decisions (e.g. through farm activities) and the environmental impacts occur at this scale, particularly in European rural landscapes (Cellier et al., 2011; Sutton et al., 2007). This makes determining landscape N_r fluxes important for environmental protection and policy makers, since a good understanding of the quantities and dynamics of N fluxes at the landscape scale is essential for designing effective regulations aimed at reducing environmental impacts. However, accurate estimation of N fluxes at high spatial resolution poses a significant challenge (de Vries et al., 2011), e.g. the estimation of spatially variable N dry deposition represents one of the key uncertainties in quantifying nitrogen inputs to terrestrial ecosystems (Tang et al., 2009).

In this study, we estimated N budgets for two adjacent catchments at the landscape scale. The catchments contrast in their land use: one is dominated by semi-natural moorland, the other by grazed grassland. To the authors' knowledge, this is the first such study which includes high resolution atmospheric modelling combined

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with a detailed spatial landscape inventory of field specific agricultural activities. The study shows how landscape N budget analysis provides an insight into the main N flux terms, key uncertainties associated with these terms and the overall implications for the environmental status of the landscape.

2 Methods

2.1 Study landscape

As part of the NitroEurope Integrated Project (Sutton et al., 2007), a landscape study area of 6 km × 6 km was established in southeast Scotland, an area with a temperate oceanic climate, for detailed inventory of agricultural activities, N_r concentration and flux measurements (see Vogt et al., 2012a, b, for further details). The study landscape was located to include the two contrasting catchments. The moorland peat-dominated catchment covered 621 ha, while the grassland dominated catchment covered 895 ha. Together these two catchments represent 42 % of the study landscape (Fig. 1).

A detailed local survey of all farms and fields in the study landscape was conducted throughout 2008. This provided land cover and farm activity data, which were collated into a relational database and spatially represented in a geographical information system (ArcGIS, ESRI). Land cover and soil types within the landscape together with the boundaries of the two studied catchments are shown in Fig. 1. Moorland and rough grass, including peat cutting and areas of both deciduous and coniferous afforestation dominate the northwestern part of the landscape and the Black Burn catchment, whereas the southeast and the Lead Burn catchment is dominated by agricultural land (henceforth referred to as the Moorland and the Grassland catchments, respectively, Table 1). Agricultural activities in the landscape range from extensive beef cattle and sheep farming to intensive poultry farming, with 24 poultry houses in the study area containing nearly 1.5 million laying hens.

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2.2 Catchment N budgets

Two recent studies have compared different budgetary approaches to quantify N balances in agricultural systems. Oenema et al. (2003) presented farm gate, soil surface and soil system budget methodologies, and de Vries et al. (2011) presented regional farm, land and soil N budgets, both studies delineated inputs and outputs for each of these approaches. In our study, the Moorland and Grassland catchment annual N budgets were assessed for 2008 using a soil budgeting approach which most closely matches the soil N budget approach of de Vries et al. (2011) (except the N pool changes), i.e. all N that enters and leaves the soil was accounted for. This type of approach was chosen as the inputs and outputs are directly associated with the catchment soils and linked to the downstream flux. The balance of the N input and output terms indicate the change in N storage within the catchment over time. There were significant N fluxes occurring in connection with the poultry housing, i.e. housing emissions and farming operations such as feed import, manure export or livestock export within one of the catchments, but for the purpose of a soil budget approach, housing emissions and farming operations not affecting the catchment land surface were considered decoupled from the soil. Thus they were excluded from this approach, except via the N deposition fluxes resulting from housing emissions.

The soil N budget was derived as follows:

$$\Delta N/\Delta t = N_{NH_3\ \mathrm{dry\ dep}} + N_{NH_x\ \mathrm{wet\ dep}} + N_{NO_y\ \mathrm{dep}} + N_{\mathrm{syn\ fert}} + N_{\mathrm{org\ fert}} + N_{\mathrm{excreta}} + N_{\mathrm{bio\ fix}} - N_{NH_3} - N_{N_2O} - N_{NO} - N_{N_2} - N_{\mathrm{harvest}} - N_{\mathrm{grass}} - N_{\mathrm{stream}} \quad (1)$$

where $\Delta N/\Delta t$ is the change in N balance (ΔN) over time (Δt); $N_{NH_3\ \mathrm{dry\ dep}}$ is the atmospheric dry deposition of ammonia (NH_3); $N_{NH_x\ \mathrm{wet\ dep}}$ is the atmospheric wet deposition of reduced nitrogen (NH_x); $N_{NO_y\ \mathrm{dep}}$ is the atmospheric dry and wet deposition of oxidised nitrogen (NO_y); $N_{\mathrm{syn\ fert}}$ is the N content in applied synthetic fertiliser; $N_{\mathrm{org\ fert}}$ is the N content in applied organic fertiliser; N_{excreta} is the amount of N excreted by grazing livestock; $N_{\mathrm{bio\ fix}}$ is the biological N_2 fixation; N_{NH_3}, N_{N_2O}, N_{NO} and N_{N_2} are emissions

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of NH_3, nitrous oxide (N_2O), nitric oxide (NO) and N_2 to the atmosphere; $N_{harvest}$ is the N offtake through harvested vegetation for silage and hay production; N_{grass} is the N offtake through harvested grass by grazing livestock; N_{stream} is the downstream export flux of total dissolved nitrogen (TDN).

The uncertainties of individual budget terms are given by estimated positive and negative errors (Sect. 3.7). The overall uncertainty of the N balance ($E_{\Delta N/\Delta t}$) was calculated as the square root of the sum of the error (E) squares, hereby accounting for the depending variables N_{grass} and $N_{excreta}$:

$$\begin{aligned} E_{\Delta N/\Delta t} = \mathrm{sqrt}[& (E_{NH_3\ dry\ dep})^2 + (E_{NH_x\ wet\ dep})^2 + (E_{NO_y\ dep})^2 + (E_{syn\ fert})^2 + (E_{org\ fert})^2 \\ & + (E_{grass} - E_{excreta})^2 + (E_{bio\ fix})^2 + (E_{NH_3})^2 + (E_{N_2O})^2 + (E_{NO})^2 + (E_{N_2})^2 \\ & + (E_{harvest})^2 + (E_{stream})^2] \end{aligned} \quad (2)$$

In the following sections the method of quantifying individual budget terms and their uncertainties is described.

2.3 Catchment N inputs

2.3.1 Atmospheric deposition

The spatial and temporal variability of atmospheric NH_3 across the landscape, in which the two catchments are contained, was described in detail by Vogt et al. (2012b). Monthly mean NH_3 concentrations at 31 sites were measured through 2008 with ALPHA passive diffusion samplers (Tang et al., 2001). Sites were distributed across the study landscape with an emphasis on capturing high and low emission areas as well as the variability around sources. Ammonia emissions were calculated for each individual field, manure store and livestock house, based on the field and farm activities recorded on a monthly basis combined with emission rates for each activity (manure housing, storage and spreading, grazing and fertiliser application, Vogt et al., 2012b). The emission estimates were used in the Local Area Dispersion and Deposition model

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(LADD) (Hill, 1998; Loubet et al., 2009) at a resolution of 25 m x 25 m to model spatial concentrations and dry deposition of NH_3 within the study landscape. Measured annual mean concentrations of the 31 sampling sites were used for verification of the LADD model. As NH_3 has a high dry deposition rate (Cellier et al., 2011) and is thus expected to be driven by local sources, NH_3 dry deposition inputs to the studied catchments ($N_{NH_3\ dry\ dep}$) were calculated from fluxes modelled by LADD within the study landscape only (accounting for atmospheric NH_3 import to the landscape using national modelling). This N budget term is considered to carry a relatively low uncertainty of ±20 % in this instance due to the detailed local study, involving an intensive measurement programme and local atmospheric dispersion modelling.

Catchment atmospheric inputs due to NH_x wet deposition ($N_{NH_x\ wet\ dep}$) and dry and wet deposition of NO_y ($N_{NO_y\ dep}$) which are expected to be largely driven by non-local sources (e.g. Hertel et al., 2011; Sutton et al., 1998) were simulated by the UK national model FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange) (Dore et al., 2012, 2007; Hallsworth et al., 2010) at a resolution of 1 km × 1 km. The contribution of particulate ammonium (NH_4^+) to NH_x dry deposition is considered minor compared to NH_3 (e.g. Asman et al., 1998; Duyzer, 1994). FRAME simulations were combined with land cover data of 25 m × 25 m resolution in order to apply land cover specific deposition rates to different land cover types, as described in detail by Vogt et al. (2012b). For the atmospheric inputs of NH_x wet deposition and dry and wet deposition of NO_y, national modelling at a relatively fine scale resolution, applied to local land cover data, is considered to deliver adequate deposition estimates for this purpose with a relatively low uncertainty in the range of ±20 %.

2.3.2 Agricultural land surface input

Agricultural inputs to the land surface through applications of synthetic fertiliser ($N_{syn\ fert}$), organic fertiliser ($N_{org\ fert}$) and excreta of grazing livestock ($N_{excreta}$) were derived from farm activity data (Vogt et al., 2012a). A typical N content was used for the different manure types (Defra, 2010). The N input from grazing livestock was estimated using grazing records and daily N excretion data as used in the UK NH_3 inventory (Misselbrook et al., 2009). Nitrogen inputs from applications of synthetic fertiliser are considered accurate as this value is known by individual farmers (estimated uncertainty ±10 %). A higher uncertainty of ±30 % is associated with the N input through applications of organic fertiliser, as a typical N content was applied to different manure types as specified by the farmer. The uncertainty associated with the N input through grazing livestock excreta is estimated to be ±50 % as the N content of the grazed grass is not known.

2.3.3 Biological N_2 fixation

Experimentally derived data on biological N_2 fixation are rare in the literature. DeLuca et al. (2008) measured fixation rates to mainly range between 1 and 2 $kg\,N\,ha^{-1}\,yr^{-1}$ in a Swedish boreal forest; Limmer and Drake (1996) cite a mean fixation rate of 1 $kg\,N\,ha^{-1}\,yr^{-1}$ from studies conducted in European and North American forests and Waughman and Bellamy (1980) measured a fixation rate of 0.7 $kg\,N\,ha^{-1}\,yr^{-1}$ in German bogs. The catchment N input through biological N_2 fixation (N_{biofix}) was thus estimated to be 1 $kg\,N\,ha^{-1}\,yr^{-1}$ for both catchments as there was little or no clover in most of the grassland. The N input through biological N_2 fixation is highly uncertain (−70/+300 %) as this term is estimated from only a few experimentally derived literature values.

2.4 Catchment N outputs

2.4.1 Gaseous emissions from land surfaces

Ammonia emissions were calculated by applying UK average emission factors (EFs) of the UK emission inventory to the land surface inputs from synthetic and organic fertiliser and grazing excreta (Misselbrook et al., 2009). The housing emissions and manure storage emissions were excluded from the calculation of catchment budgets

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as discussed in Sect. 2.2. As calculations of NH_3 emissions are based on the local farm inventory and national emission factors, the uncertainty is estimated to be relatively low (±20 %).

Direct N_2O emissions are associated with soil N input ($N_{NH_3\ dry\ dep} + N_{NH_x\ wet\ dep} + N_{NO_y\ dep} + N_{syn\ fert} + N_{org\ fert} + N_{excreta}$) and were calculated using the method of Lesschen et al. (2011), which uses specific EFs depending on the source of N input, soil type and annual precipitation. The clay soil EF parameterisation in Lesschen et al. (2011) was selected linked to the modification of the catchment surface soils by agricultural activity. The local 2008 annual precipitation of 1208 mm was used to derive a precipitation adjustment factor (f_p) in the method of Lesschen et al. (2011) of 2.16. Peat cutting areas and other peat bog areas without agricultural activities are assumed to have insignificant N_2O emissions due to soil C/N ratios exceeding 25 (Klemedtsson et al., 2005). Also, measurements within the Moorland catchment showed negligible N_2O emissions (Drewer et al., 2010). Indirect N_2O emissions, i.e. degassing of N_2O from waters resulting from soil leaching, were estimated using the 2009 IPCC Guidelines (De Klein et al., 2009).

Emissions of NO were derived by applying a Tier 1 EF of 2.6 % to synthetic fertiliser N applied as recommended in the EEA/EMEP guidelines (McGlade and Vidic, 2009). As there is no specific EF recommended for applications of organic fertiliser and grazing livestock excreta a literature value of 0.5 % was applied (Bouwman et al., 2002).

The uncertainty of N_2O and NO emissions is estimated at ±50 % as they are based on data from the farm inventory and also literature emission factors. Emissions are known to vary substantially depending on soil conditions.

Emission factors of N_2 are highly uncertain. Recently, Ammann et al. (2009) applied a literature-derived EF of 12.5 % to N inputs from fertilisation and biological N_2 fixation for a Swiss grassland with an error of ±100 %. For a grazed grassland in southeast Scotland (<10 km from this study landscape), N_2 emissions were modelled and an EF of 10 % of applied N through grazing excreta and synthetic and organic fertilisation calculated (Skiba, personal communication, 2011). This N_2 EF was applied to all fields

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with agricultural activities in our study catchments. It is noted that there is a large uncertainty (−50/+200 %) associated with this budget term (Sect. 3.7).

2.4.2 Harvested vegetation

Nitrogen output also occurs via removal of vegetation by harvesting ($N_{harvest}$) and by grazing livestock (N_{grass}). The amount of harvested crop and grass removed by farmers for silage and hay production was derived from the farm survey activity data with a specific N content applied to each main crop type (Møller et al., 2005). The uncertainty of $N_{harvest}$ is thus estimated at ±20 %. The amount of N removed through grass consumption by grazing livestock (N_{grass}) was estimated as follows:

$$N_{grass} = N_{excreta} + N_{animal} - N_{feed}$$

where $N_{excreta}$ is the amount of N excreted by grazing livestock (Sect. 2.3.2), N_{animal} is the N content in the exported wool and meat, calculated using N content values in Roche (1995) and Flindt (2003) and N_{feed} is the N content of the supplementary animal feed, derived by farm activity data and a specific N content of different feed types (Møller et al., 2005). Both N_{animal} and N_{feed} are estimated to have an uncertainty of ±20 %, however considering the ±50 % uncertainty associated with $N_{excreta}$, the uncertainty of N_{grass} is estimated at ±50 %.

2.4.3 Fluvial export

Annual downstream fluxes (N_{stream}) of total dissolved nitrogen (TDN), which is the sum of ammonium (NH_4^+-N), nitrate (NO_3^--N) and dissolved organic nitrogen (DON), were established by Vogt et al. (2012a) by sampling at gauged outlets of the two catchments at both fortnightly and hourly intervals during selected high flow events through 2008. As N_{stream} is based on local measurements conducted throughout the study year, it is considered to carry a relatively low uncertainty, conservatively estimated at ±20 %.

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Additional information on sources of streamwater N concentrations within the catchments was derived by spatial sampling at stable low flow conditions, conducted in July, September and December 2008.

3 Results and discussion

The outcomes are explored here using spatially differentiated results of the agricultural land surface N input, the associated land surface N emissions and atmospheric N deposition and fluvial N export. In addition, the catchment N inputs and output terms are summarised and the overall catchment N budgets are given with a discussion of uncertainty.

3.1 Agricultural land surface N input

Agricultural N inputs to the land surface were dominated by grazing excreta in both catchments: in the Moorland catchment, grazing excreta contributed 73 %, organic fertiliser 17 % and synthetic fertiliser 10 % to the land surface input; in the Grassland catchment, grazing excreta contributed 51 %, organic fertiliser 31 % and synthetic fertiliser 18 %. Most of the N in grazing excreta originated from sheep with contributions of 89 % in the Moorland and 69 % in the Grassland catchment. Fields within the Grassland catchment received more than four times the land surface N input (51.9 kg N ha^{-1} yr^{-1}) than fields in the Moorland catchment (12.1 kg N ha^{-1} yr^{-1}). The range of land surface inputs between fields was large, varying from 0 to 261 kg N ha^{-1} yr^{-1} in the Moorland and up to 346 kg N ha^{-1} yr^{-1} in the Grassland catchment.

No fields of the study landscape are located within a Nitrate Vulnerable Zone (NVZ), thus agricultural practice is not restricted by the Nitrate Directive (Defra, 2012), under which a maximum of 170 kg N ha^{-1} yr^{-1} of organic manures is set. In the present study, only 1 % of the Moorland and 4.5 % of Grassland catchment received manure, through organic fertiliser applications or grazing excreta, exceeding 170 kg N ha^{-1} yr^{-1}, although

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it is noted that there are significant uncertainties associated with the calculation of these N inputs.

3.2 Atmospheric N emissions

Gaseous NH_3 emissions from the catchment land surface (excluding housing and manure store emissions) are shown in Fig. 2a. In the Moorland catchment, field based emissions ranged from 0 to 48 $kg\,N\,ha^{-1}\,yr^{-1}$ (mean: 0.9 $kg\,N\,ha^{-1}\,yr^{-1}$) with 58 % originating from applications of organic fertiliser, 40 % from grazing excreta and 2 % from synthetic fertiliser. In the Grassland catchment, NH_3 emissions ranged from 0 to 53 $kg\,N\,ha^{-1}\,yr^{-1}$ between individual fields (mean: 4.5 $kg\,N\,ha^{-1}\,yr^{-1}$) with 66 % arising from organic fertiliser, 30 % from grazing excreta and 4 % from synthetic fertiliser. Despite most of the agricultural land surface input originating from grazing excreta (Sect. 3.1), the dominant source of NH_3 emissions were applications of organic fertiliser in both catchments, due to high NH_3 volatilisation losses. In contrast, almost all N in grazing excreta (~95 %) can be expected to enter the catchment soils and thus contribute to soil emissions of N_2O and N_2 or can be leached. Overall, 7 % of the agricultural land surface input of N to the Moorland catchment was estimated to be emitted as NH_3 compared with 9 % from the Grassland catchment.

Direct N_2O emissions from the Moorland catchment averaged to 0.8 $kg\,N\,ha^{-1}\,yr^{-1}$ with field emissions ranging from 0 to 7.0 $kg\,N\,ha^{-1}\,yr^{-1}$ (Fig. 2b). The Grassland catchment emitted 2.4 $kg\,N\,ha^{-1}\,yr^{-1}$ as N_2O with emissions ranging from 0.4 to 12.5 $kg\,N\,ha^{-1}\,yr^{-1}$ between fields. Most of the direct N_2O emissions were from grazing excreta (79 % in the Moorland and 75 % in the Grassland catchment). Around 7 % of the grazing excreta were estimated to be lost as N_2O in both catchments. Figure 2c shows field emissions of N_2 within the catchments. In the Moorland catchment, N_2 emissions (1.2 $kg\,N\,ha^{-1}\,yr^{-1}$) are estimated to be similar to N_2O emissions, whereas in the Grassland catchment, N_2 emissions (5.3 $kg\,N\,ha^{-1}\,yr^{-1}$) are about 2.5 times higher than N_2O emissions. Emissions per field ranged from 0 to 26.3 $kg\,N\,ha^{-1}\,yr^{-1}$ in the Moorland

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and from 0 to 36.2 $kg\,N\,ha^{-1}\,yr^{-1}$ in the Grassland catchment. However, the uncertainties within those field based emission estimates were relatively large (Table 4) as there is substantial within field variation of N_2O and N_2 emissions due to the heterogeneity of soil processes (e.g. Hofstra and Bouwman, 2005).

Soil NO emissions were estimated to be insignificant for both catchments with emissions of 0.1 $kg\,N\,ha^{-1}\,yr^{-1}$ in the Moorland and of 0.3 $kg\,N\,ha^{-1}\,yr^{-1}$ in the Grassland catchment. The field with the highest NO emission was common to both catchments, thus the field specific emission range of 0 to 1.8 $kg\,N\,ha^{-1}\,yr^{-1}$ was the same for both catchments.

3.3 Atmospheric N deposition

The total atmospheric N deposition to the two studied catchments was estimated to be 8.2 $kg\,N\,ha^{-1}\,yr^{-1}$ in the Moorland and 12.3 $kg\,N\,ha^{-1}\,yr^{-1}$ in the Grassland catchment (Fig. 3). The dry deposition of NH_3 to the study catchments ($N_{NH_3\ dry\ dep}$) was estimated by modelling emissions of all agricultural NH_3 sources within the study landscape, including housing and manure storage emissions (Sect. 2.3.1). Dry deposition of NH_3 showed a high spatial variability at 25 m × 25 m grid resolution within the catchments, ranging from 0.1 to 23 $kg\,N\,ha^{-1}\,yr^{-1}$ in the Moorland (mean: 2.4 $kg\,N\,ha^{-1}\,yr^{-1}$) and from 0.2 to >100 $kg\,N\,ha^{-1}\,yr^{-1}$ in the Grassland catchment (mean: 6.4 kg N $ha^{-1}\,yr^{-1}$). The larger input to the Grassland catchment was due to the catchment containing six intensive poultry farming houses with a total NH_3 emission of 28 t N yr^{-1}.

Catchment inputs from NH_x wet deposition were similar for both catchments (2.5 and 2.6 $kg\,N\,ha^{-1}\,yr^{-1}$, respectively), as were inputs from NO_y deposition (both 3.3 $kg\,N\,ha^{-1}\,yr^{-1}$). Atmospheric deposition to the Moorland catchment was estimated to be driven by non-local sources with $N_{NH_x\ wet\ dep}$ and $N_{NO_y\ dep}$ contributing 71 % to the total N deposition, while 52 % of deposition to the Grassland catchment was estimated to originate from local sources ($N_{NH_3\ dry\ dep}$) and 48 % from non-local sources ($N_{NH_x\ wet\ dep} + N_{NO_y\ dep}$).

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3.4 Fluvial N export

Both catchments were characterised by highly variable stream flow with high discharge events making an important contribution to annual downstream fluxes (Vogt et al., 2012a). For example, in 2008, the highest 10 % of the discharge data contributed 53 % to the total discharge in the Moorland and 40 % in the Grassland catchment. The annual downstream flux (N_{stream}) of total dissolved nitrogen (TDN) was 8.7 kg N ha^{-1} yr^{-1} in the Moorland and 14.4 kg N ha^{-1} yr^{-1} in the Grassland catchment. The difference in the TDN flux was mainly due to the significantly larger nitrate (NO_3^-) flux in the Grassland catchment. Dissolved organic nitrogen (DON) contributed 81 % to the TDN flux in the Moorland and 49 % in the Grassland catchment. However, the absolute annual DON flux of 7.0 kg N ha^{-1} yr^{-1} was very similar in both catchments.

Maps of annual mean concentrations of NO_3^-, NH_4^+ and DON measured during the three spatial sampling campaigns are shown in Fig. 4, together with the underlying land cover. The streamwater NO_3^- concentrations of both catchments have been shown to be significantly positively related to N input through agricultural land surface and atmospheric deposition (Vogt et al., 2012a). Ammonium concentrations were significantly negatively related to N input and could be related to the coverage of wet peaty soils (Vogt et al., 2012a). However, local point source contributions, such as suspected sewage discharge observed while collecting samples, may also contribute to the large spatial variability of NH_4^+ concentrations within the Grassland catchment. The sources of DON can vary widely and differed between the catchments (Vogt et al., 2012a). In both catchments, flushing of organic-rich soil water contributed to streamwater DON concentrations, however in the Grassland catchment, there were additional major sources, such as agricultural runoff.

To analyse the potential contribution of the peat cutting area to the DON as well as to the linked dissolved organic carbon (DOC) export flux of the Moorland catchment, the catchment was divided into eight subcatchments based on the drainage pattern. A regression analysis between the % area of peat soil in these subcatchments and DON and DOC concentrations at the subcatchmet outlets mostly showed a positive relationship between DOC and DON concentrations and the % area of peat soil (Fig. 5a, b). This relationship was more pronounced for DOC than DON, however, in both cases there was substantial scatter in the relationship. Other studies (e.g. Aitkenhead et al., 1999) have shown that the area of peat soil in a catchment is directly related to streamwater DOC concentration. Clark et al. (2004) found DON concentrations to be positively related to peat cover in the summer only. In this study, the relationship between DON concentrations and % area of peat soil was also strongest in July. The same regression analysis with % peat cutting area also showed a similar positive relationship to DOC and DON concentrations (Fig. 5c, d) with a slightly stronger relationship observed between concentrations and % peat cutting area (compared to % peat area). This is likely to be a reflection of peat cutting taking place in the areas of deepest peat in the catchment leading to the enhanced effect shown in Fig. 5c, d. The areas affected by peat cutting are mostly in the upper parts of the catchment, with the effect decreasing significantly downstream. Also, a previous study in the Moorland catchment noted that DOC concentrations were not significantly different in a large tributary originating from an area of peat cutting compared to concentrations in the main stream (Dinsmore et al., 2010). Thus, peat rich areas (whether cut or not) are considered to be the main source of streamwater DOC and DON concentrations. However, peat cutting and associated drainage will change hydrological flow paths which may enhance the "peat effect" on DOC and DON concentrations and contribute to higher annual fluxes because of greater runoff due to drainage. The longer term effect of peat cutting on the catchment fluvial N flux remain a question for further study.

3.5 N inputs to land in the study catchments

The various components which contribute N inputs to the two study catchments are summarised in Fig. 6 (input estimates expressed per hectare) and Table 1 (total input per catchment area). Overall, the inputs to the Grassland catchment (65.2 kg N ha^{-1} yr^{-1}) were about three times higher than those to the Moorland

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catchment (21.3 kg N ha^{-1} yr^{-1}). Inputs were largely driven by agricultural land surface inputs. In the Grassland catchment, 80 % of all N inputs originated from agricultural land surface inputs, 18 % from atmospheric N deposition and 2 % estimated from biological fixation of N_2. Atmospheric deposition accounted for a larger contribution in the Moorland catchment with 38 % of all N inputs. However, the majority (57 %) originated from agricultural land surface inputs and 5 % from estimated biological N_2 fixation. Grazing livestock excreta represented the largest single input source, contributing 41 % to the inputs in the Moorland and 40 % in the Grassland catchment. The fraction of the grazing excreta subject to gaseous emissions to the atmosphere (Sect. 3.2) was estimated to be around 21 %, thus the majority of the catchment input through grazing excreta stayed either within the system, i.e. in soil or vegetation, or was leached into surface or groundwaters.

3.6 N outputs from land in the study catchments

Catchment outputs are shown as per hectare values in Fig. 7 and as per catchment values in Table 3. The gaseous land surface emissions of N_r ($N_{NH_3} + N_{N_2O} + N_{NO}$) led to losses of 1.7 kg N ha^{-1} yr^{-1} from the Moorland and 7.3 kg N ha^{-1} yr^{-1} from the Grassland catchment. Whereas emissions of N_2O are similar to those of NH_3 in the Moorland catchment, emissions from the Grassland catchment were dominated by NH_3 emissions (62 %). Emissions of NO were relatively insignificant in both catchments: 0.1 kg N ha^{-1} yr^{-1} in the Moorland and 0.3 kg N ha^{-1} yr^{-1} in the Grassland. The estimated N_2 emissions were large compared with the N_r fluxes of the catchments, contributing 42 % to the overall N emission flux from both catchments. However, the uncertainty within the N_2 emission estimations is large (see Table 4 in Sect. 3.7).

Grazed grass (N_{grass}) constituted a large output term in both catchments, contributing 45 % to the overall catchment output in the Moorland and 46 % in the Grassland catchment. However, these losses were mostly recycled back to the soil by grazing livestock excreta ($N_{excreta}$) with $N_{excreta}$ representing 83 % of N_{grass} in the Moorland and

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96 % of N_{grass} in the Grassland catchment. Thus, the main importance of this "grazing livestock N cycle" are increased rates of soil N cycling associated with the grazing excreta which lead to gaseous and streamwater losses. When considering the grazed grass as a recycling budget term, the largest output fluxes of both catchments were the stream exports.

3.7 Total N budgets for the study catchments

The overall nitrogen budgets for two catchments are compared in Table 4 and Fig. 8. The Moorland catchment showed a negative N balance of −1.6 + 3.8/−3.4 (error) $kg\,N\,ha^{-1}\,yr^{-1}$, potentially indicating a small loss of N from catchment storage to the stream, however within the uncertainty estimates the catchment N budget could also be in balance. Reynolds and Edwards (1995) stated that N accumulation is to be expected in moorland catchments. However, that study did not take stream exports of DON into account due to lack of data. The present study thus shows the importance of DON as a component of stream export: DON accounted for 81 % of TDN export. The N loss calculated for the Moorland catchment is in agreement with an overall N loss of −2.4 $kg\,N\,ha^{-1}\,yr^{-1}$, derived for a field site within the Moorland catchment (Drewer et al., 2010). Drewer et al. (2010) compiled budget terms from different years, accounting for inputs through inorganic N deposition, as well as losses through N_2O emissions and stream export of measured inorganic and estimated organic N.

Nitrogen saturation has been defined for "an ecosystem where N losses approximate or exceed the inputs of N" (Ågren and Bosatta, 1988; Butterbach-Bahl et al., 2011). Thus, according to our catchment soil budget approach, the Moorland catchment showed signs of N saturation. If the Moorland catchment is losing N, it is of interest to know whether carbon (C) loss is also occuring. Recently, Dinsmore et al. (2010) showed the DOC downstream flux to be a significant loss within the C budget of the Moorland catchment, although the moorland was still found to act as a strong C sink, mainly due to a large C uptake from the atmosphere. However, in the past, the same moorland has also been found to be either C neutral or a small C source (Billett et al., 2004). The differing C balances reflect large inter-annual variability in flux terms, particularly C uptake from the atmosphere which in turn is influenced by the annual fluctuations in weather. Thus, the studied Moorland catchment may shift at an annual level from acting as a net C sink to a source, while at the same time releasing a significant amount of C from the catchment via downstream DOC export. The effects of future climate change on catchment scale C and N budgets remain highly uncertain.

The Grassland catchment had a positive N balance of 5.9 + 7.4/−12.3 (error) $kg\,N\,ha^{-1}\,yr^{-1}$, indicating that the catchment stored N inputs in soil, vegetation and groundwater for this study year. However, as with the Moorland catchment, the error bars overlap the balance point. The stream export of the Grassland catchment represented a relatively large budget term compared with the other terms. By comparison with other European regional catchment budgets reported by Billen et al. (2011), the retention of N was low (Sect. 4).

3.8 Uncertainties in the catchment nitrogen budgets

For both catchments, the budget terms with the largest error bars were the outputs through grazed grass (N_{grass}) and the input through grazing excreta ($N_{excreta}$), as noted above. However, as those terms are interdependent and it is the difference between them that contributes to the overall uncertainty of the N balance calculation, the net error is smaller than the individual errors. In the Moorland catchment, the budget terms contributing the most to the uncertainty of the N balance were biological N_2 fixation, stream export and N_2 emissions. In the Grassland catchment, the most important terms contributing to uncertainty were N_2 emissions, followed by applied organic fertiliser and stream export. The overall uncertainty of the N balances were large, the Moorland catchment balance being −1.6 $kg\,N\,ha^{-1}\,yr^{-1}$ with estimated upper and lower balance values of +2.2 and −5.0 $kg\,N\,ha^{-1}\,yr^{-1}$, accounting for uncertainties. Similarly, the upper and lower estimates of the Grassland catchment of +5.9 $kg\,N\,ha^{-1}\,yr^{-1}$ range between +13.3 and −6.4 $kg\,N\,ha^{-1}\,yr^{-1}$. Hence, although we present a detailed budget analysis, the uncertainties remain inherently large.

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There are several terms still missing from the N budget calculation, which may add further uncertainty to the current balance estimate. In particular, atmospheric deposition of gaseous and particulate organic N compounds were not quantified nor estimated due to lack of information, although organic deposition may be an important input (Cape et al., 2004; Neff et al., 2002). Moreover, fluvial N export through particulate organic N (PON) was not measured, although the PON flux is likely to be insignificant compared to the DON flux as was the POC flux to the DOC flux measured in the Moorland catchment by Dinsmore et al. (2010).

Although our study was detailed, it was carried out over a relatively short time period (one year), which may affect some of the conclusions drawn from the data. In particular, stream export fluxes are known to vary year-on-year due to climatic fluctuations (Gascuel-Odoux et al., 2010). Further study on the N budgets of these catchments is needed to clarify the role of annual variation. Another source of uncertainty is the assumption that land use and N input remain approximately constant with time allowing the balancing of N exported through the aqueous system with the N exchange at the surface.

3.9 Comparison with a regional catchment N budget approach

Regional scale catchment N budgets have been estimated for many European catchments (Billen et al., 2011). The approach combines a calculation of the net anthropogenic input of reactive nitrogen (NANI, Howarth et al., 1996) to the catchment with data on atmospheric NO_y deposition, crop N fixation, fertiliser use and import of food and feed. This is a simple approach which can be applied to large regions, but does not account for processes like NH_3 volatilisation or soil denitrification. In European regional catchments, NANI ranges between 0 and 84 $kg\,N\,ha^{-1}\,yr^{-1}$ (mean: 37 $kg\,N\,ha^{-1}\,yr^{-1}$) (Billen et al., 2011). The relative difference of NANI to the stream export of total N (TN = DIN + DON + PON) is then associated with catchment N retention. Catchment retention refers to the amount of N which is either stored in soils and groundwater or lost through emissions to the atmosphere. In regional European catchments, catchment N

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retention varies between 50 % and 90 % of NANI (mean: 82 %) (Billen et al., 2011). There is some evidence that the fraction of NANI exported by the stream is larger in northern European catchments with high discharges.

These regional budget calculations differ substantially to the one presented here, (e.g. coarser scale data, no NH_x deposition, no land emissions, no organic fertiliser applications); however, the catchment retention calculated as the percentage of the net anthropogenic input which is stored or emitted using our budget terms for the landscape scale may emphasise the differences of regional and landscape scale N budgets. Thus, a landscape NANI was calculated (see Sect. 2.2 for budget term definitions):

$$\text{landscape NANI} = N_{NH_3\ \text{dry dep}} + N_{NH_x\ \text{wet dep}} + N_{NO_y\ \text{dep}} + N_{\text{syn fert}} + N_{\text{org fert}} + N_{\text{excreta}} - N_{\text{harvest}} - N_{\text{grass}} \tag{3}$$

The landscape NANI differs to the budget calculation of Eq. (1) in that biological N_2 fixation, the land emissions and stream export are not taken into account. Atmospheric emissions were excluded in order to calculate what hydrologists term "catchment retention", i.e. the fraction that is not exported in streamwater (which includes N losses to the atmosphere). Landscape NANI was estimated here at 9.0 kg N ha^{-1} yr^{-1} for the Moorland and 31.8 kg N ha^{-1} yr^{-1} for the Grassland catchment. These values are relatively small compared with the NANI calculated for European regional catchments with an average of 37 kg N ha^{-1} yr^{-1} (Billen et al., 2011). The stream N export (not including PON) represented, therefore, 97 % of landscape NANI in the Moorland, compared with 45 % in the Grassland catchment. This implies a catchment retention of 3 % of landscape NANI in the Moorland and 55 % in the Grassland catchment. These values are low, particularly the retention of the Moorland catchment, compared to the catchment retention calculated at regional scale in Europe with an average of 82 % (Billen et al., 2011). Reasons for the difference between these two budget approaches are likely to be the finer scale resolution of our landscape scale study allowing, firstly, for more accurate quantification of the N budget terms and secondly, for the calculation of more budget terms to account for the net anthropogenic input related to catchment

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soils. Expressed in terms of our comprehensive landscape N budgets, the actual "net nitrogen retention" ([all N_{input} – all N_{output}]/all $N_{input} \cdot 100$) would be +9 % and −7.6 % for the Grassland and Moorland catchments, respectively.

4 Conclusions

Nitrogen budgets for two adjacent catchments with contrasting land use within a single landscape unit were calculated taking into account all agricultural activity and each of the important gaseous and aqueous inputs and outputs. This allowed a detailed analysis of catchment inputs and outputs at a much higher spatial resolution than before. Within the errors associated with components of the N budget, the two catchments are in an approximate net N balance, although the best estimates suggested a tendency for the Grassland catchment to gain nitrogen (+6 [−6, +13] kg N ha^{-1} yr^{-1}) and for the Moorland catchment to lose nitrogen (−2 [−5, +2] kg N ha^{-1} yr^{-1}). The key uncertainties of our N budget approach were N_2 emissions and stream N export. This emphasises, firstly the need for more studies addressing the quantification of N_2 emissions and, secondly the importance of estimating downstream fluxes accurately.

The N budgets of the two study catchments indicate that both catchments have a limited capacity to store nitrogen within soils, vegetation and groundwater. This important finding contrasts with regional scale estimates. The "catchment retention" of N, calculated as the percentage of net anthropogenic N input which is not lost in streamwater (i.e. stored within the catchment or emitted to the atmosphere), amounted to 3 % in the Moorland and 55 % in the Grassland catchment. These values are relatively small compared with estimated catchment retentions in European catchments at the regional scale, ranging from 50 % to 90 % (Billen et al., 2011). Whereas larger, regional scale approaches to estimating catchment input/output may be important for a global overview, these approaches tend to hide the landscape scale N dynamics and thus the local scale environmental impact of human activities.

This work on compiling landscape scale nitrogen budgets represents the beginning of a better understanding of the anthropogenic impact via agricultural activities on European landscapes. Within the NitroEurope Integrated Project (Sutton et al., 2007), the outcomes of this study are being further analysed in the context of nitrogen fluxes and budgets quantified in different landscapes across Europe, with differing agricultural land use and climate. This will provide a quantitative comparison of the key N fluxes and their spatial dynamics across European landscapes, providing a basis to tune locally optimised management strategies.

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Table 1. Characteristics of the Moorland and the Grassland catchment.

	Moorland catchment	Grassland catchment
Area (km^2)	6.2	8.9
Average altitude	270	280
% main land cover types:		
Grassland	11	59
Rough grass	10	10
Moorland	63	5
Peat cutting	12	2
Woodland	2	14
% main soil types:		
Brown forest soils	16	48
Peat	67	21
Peaty gleys	10	2
Noncalcareous gleys	5	22

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Table 2. Catchment totals of N inputs ($kg\,N\,yr^{-1}$).

	Moorland catchment	Grassland catchment
$N_{dry\ NH_3\ dep}$	1480	5700
$N_{wet\ NH_x\ dep}$	1560	2310
$N_{NO_y\ dep}$	2030	2980
$N_{syn\ fert}$	760	8290
$N_{org\ fert}$	1310	14 590
$N_{excreta}$	5460	23 570
N_{fix}	620	890
Total input	13 220	58 340

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Table 3. Catchment totals of N outputs ($kg\,N\,yr^{-1}$).

	Moorland catchment	Grassland catchment
N_{NH_3}	540	4050
N_{N_2O}	470	2160
N_{NO}	43	300
N_{N_2}	770	4730
$N_{harvest}$	450	4460
N_{grass}	6580	24 520
N_{stream}	5370	12 860
Total output	14 230	53 080

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Table 4. Soil N budgets for the Moorland and the Grassland catchment with fluxes and errors shown in $kg\,N\,ha^{-1}\,yr^{-1}$ (see subsections under Sects. 2.3 and 2.4.1 for details of individual error estimations).

		Moorland catchment Fluxes	Moorland catchment Error	Grassland catchment Fluxes	Grassland catchment Error
Catchment N inputs:					
NH_3 dry deposition	$N_{NH_3\ dry\ dep}$	2.4	±0.5	6.4	±1.3
NH_x wet deposition	$N_{NH_x\ wet\ dep}$	2.5	±0.5	2.6	±0.5
NO_y deposition	N_{NO_y}	3.3	±0.7	3.3	±0.7
Synthetic fertiliser applications	$N_{syn\ fert}$	1.2	±0.1	9.3	±0.9
Organic fertiliser applications	$N_{org\ fert}$	2.1	±0.6	16.3	±4.9
Grazing livestock excreta	$N_{excreta}$	8.8	±4.4	26.3	±13.2
Biological N_2 fixation	N_{fix}	1.0	+3.0/−0.7	1.0	+3.0/−0.7
Total N input		21.3		65.2	
Catchment N outputs:					
NH_3 emission	N_{NH_3}	0.9	±0.2	4.5	±0.9
N_2O emission	N_{N_2O}	0.8	±0.4	2.4	±1.2
NO emission	N_{NO}	0.1	±0.0	0.3	±0.2
N_2 emission	N_{N_2}	1.2	+2.5/−0.6	5.3	+10.6/−2.6
Harvested silage and hay	$N_{harvest}$	0.7	±0.1	5.0	±1.0
Harvested grass by grazing livestock	N_{grass}*	10.6	±5.3	27.4	±13.7
Stream export	N_{stream}	8.7	±1.7	14.4	±2.9
Total N output		22.9		59.3	
N balance		−1.6	+3.8/−3.4	+5.9	+7.4/−12.3

* $N_{grass} = N_{excreta} + N_{animal} - N_{feed}$.
N_{animal} is N exported via wool and meat production.
N_{feed} is N imported via supplementary animal feed.
Moorland catchment: $N_{animal} = 2.0\,kg\,N\,ha^{-1}\,yr^{-1}$, $N_{feed} = 0.2\,kg\,N\,ha^{-1}\,yr^{-1}$.
Grassland catchment: $N_{animal} = 5.4\,kg\,N\,ha^{-1}\,yr^{-1}$, $N_{feed} = 4.3\,kg\,N\,ha^{-1}\,yr^{-1}$.

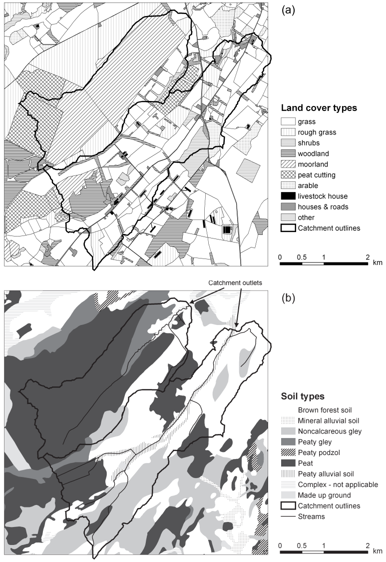

Fig. 1. Maps of land cover **(a)** and soil types[1] **(b)** within the study landscape with outlines of the two studied catchments[2].

[1] © The James Hutton Institute 2011 (license MI/2008/296). Soil types are based on the Scottish Soil Survey, the equivalent FAO names are: brown forest soil = cambisol, mineral alluvial soil = fluvisol, noncalcareous gley = gleysol, peaty gley = humic gleysol, peaty podzol = humic podzol, peat = histosol, peaty alluvial soil = humic fluvisol.

[2] Some features of these maps are based on data licensed from Intermap Technologies Inc. © 2010 Intermap Technologies Inc. All Rights Reserved.

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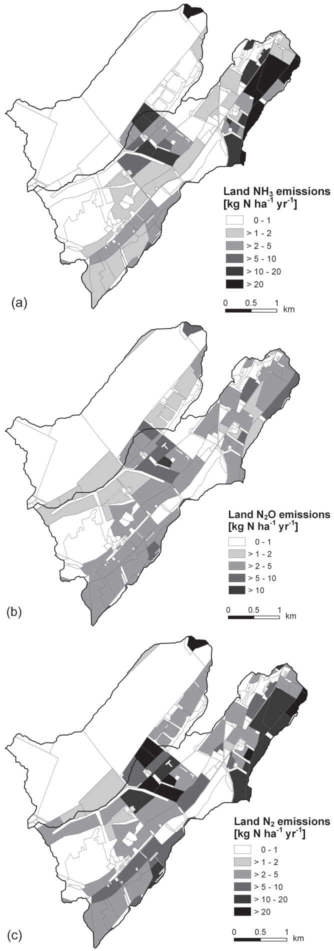

Fig. 2. Field specific land surface emission maps of **(a)** NH_3 emissions, **(b)** direct N_2O emissions, and **(c)** direct N_2 emissions.

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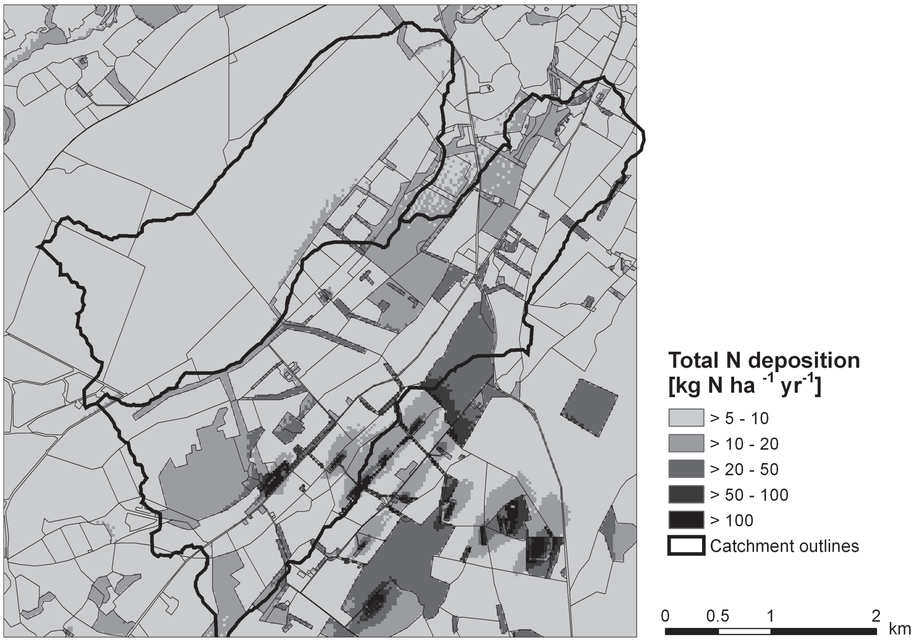

Fig. 3. Map of estimated total N deposition within the study landscape. Source: Vogt et al. (2012b).

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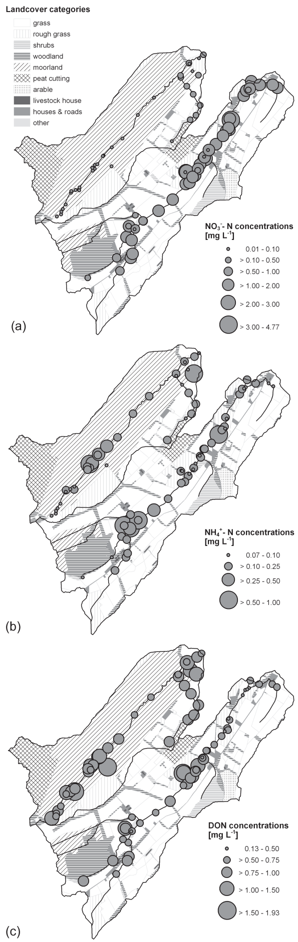

Fig. 4. Maps of annual mean concentrations derived from spatial samplings in July, September and December 2008: **(a)** NO_3^-, **(b)** NH_4^+, and **(c)** DON. Source: Vogt et al. (2012a).

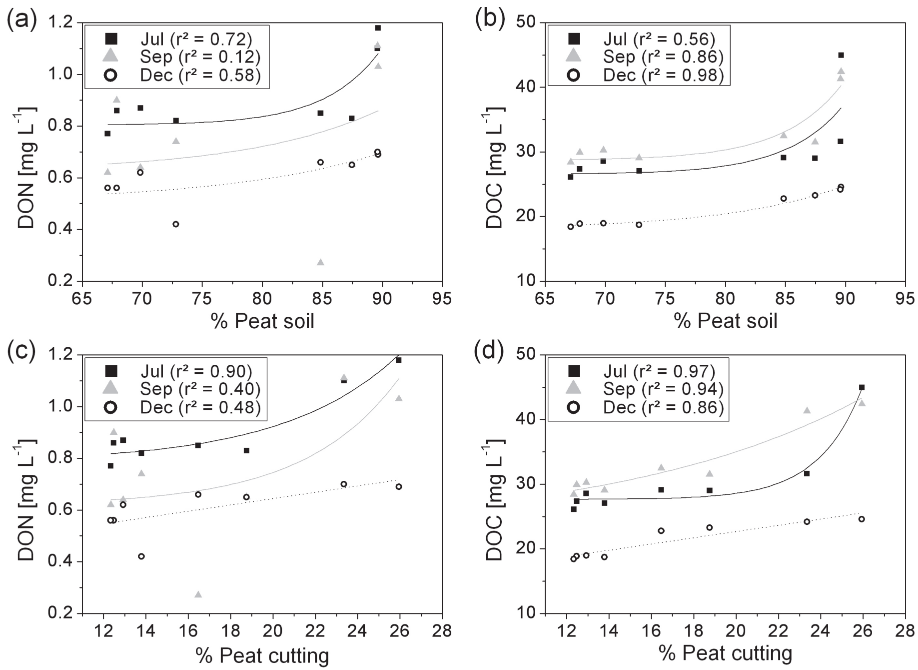

Fig. 5. Relationships between % area of peat soil **(a, b)** and peat cutting **(c, d)** in eight subcatchments of the Moorland catchment and spatial concentrations of DON **(a, c)** and DOC **(b, d)** at subcatchment outlets. The results are shown for July (black squares and line), September (grey triangles and line) and December (black circles and dotted line) and fitted as $y = A \cdot \exp(x/t) + y0$). Coefficients of determinations (r^2) are given for each campaign.

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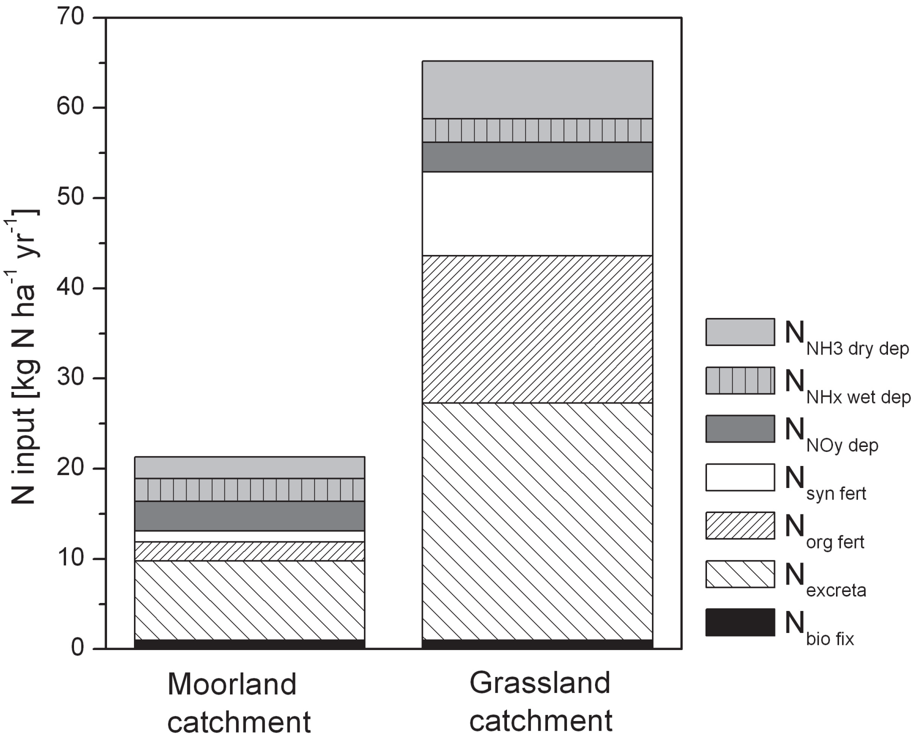

Fig. 6. N inputs ($kg\,N\,ha^{-1}\,yr^{-1}$) to the Moorland (left) and the Grassland catchment (right).

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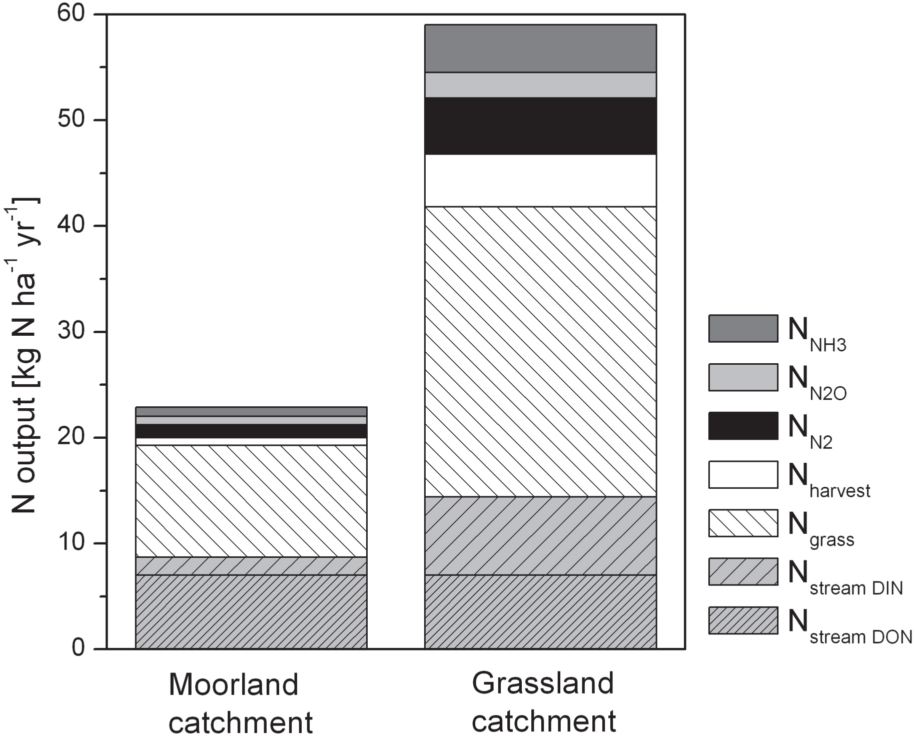

Fig. 7. N outputs (kg N ha^{-1} yr^{-1}) to the Moorland (left) and the Grassland catchment (right). Stream TDN export fluxes (N_{stream}) are split into the dissolved inorganic flux ($N_{stream\ DIN}$ = fluxes of NH_4^+ and NO_3^-) and the dissolved organic flux ($N_{stream\ DON}$).

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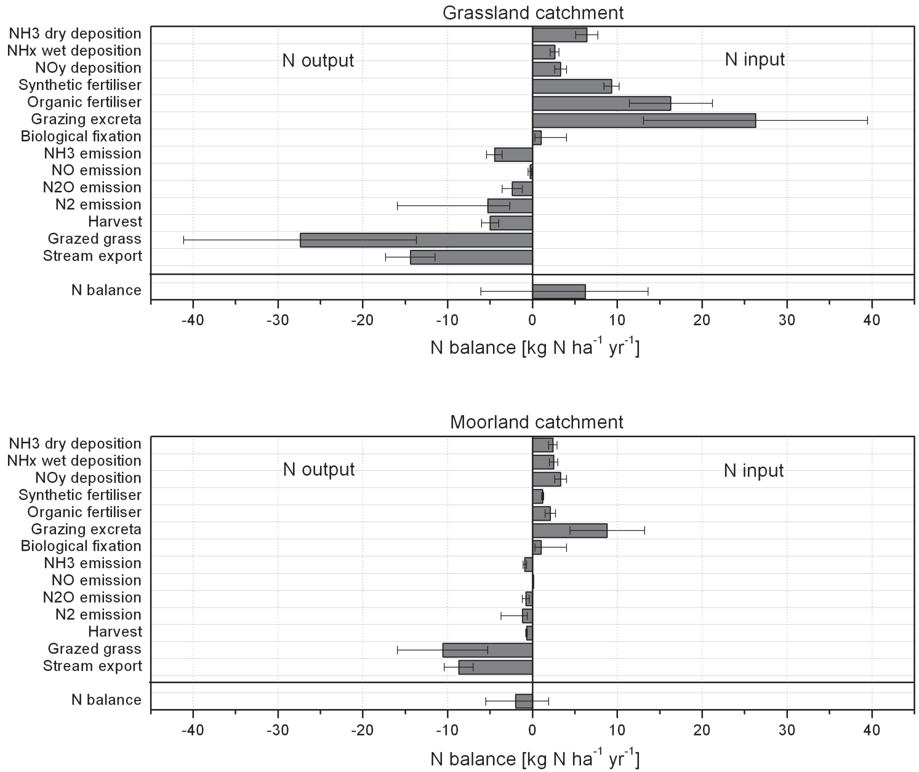

Fig. 8. Catchment soil N budgets for the Grassland catchment (top) and the Moorland catchment (bottom). Inputs and outputs are shown as positive and negative N exchanges ($kg\,N\,ha^{-1}\,yr^{-1}$) with the overall N balance shown at the bottom. Error bars represent the estimated uncertainty for the individual budget terms (see subsections under Sects. 2.3 and 2.4) with the N balance error calculated accordingly (see Sect. 2.2).

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