Biogeosciences Discuss., 7, 8345–8379, 2010
www.biogeosciences-discuss.net/7/8345/2010/
doi:10.5194/bgd-7-8345-2010

Biogeosciences
Discussions

Direct contribution of nitrogen deposition to nitrous oxide emissions in a temperate beech and spruce forest – a ^{15}N tracer study

N. Eickenscheidt, R. Brumme, and E. Veldkamp

Soil Science of Tropical and Subtropical Ecosystems, Buesgen Institute,
University of Goettingen, Buesgenweg 2, 37077 Goettingen, Germany

Received: 29 September 2010 – Accepted: 3 November 2010
– Published: 12 November 2010

Correspondence to: N. Eickenscheidt (neicken@gwdg.de)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

The impact of atmospheric nitrogen (N) deposition on nitrous oxide (N_2O) emissions in forest ecosystems is still unclear. The objective of our study was to investigate the direct contribution of N deposition to N_2O emissions in temperate forests exposed to chronic high N deposition using a ^{15}N labelling technique. In a Norway spruce stand (*Picea abies*) and in a beech stand (*Fagus sylvatica*) in the Solling, Germany, we added a low concentrated ^{15}N-labelled ammoniumnitrate solution to simulate N deposition. Nitrous oxide fluxes and ^{15}N isotope abundances in N_2O were measured using the closed chamber method combined with ^{15}N isotope analyses. Emissions of N_2O were higher in the beech stand ($2.6 \pm 0.6\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$) than in the spruce stand ($0.3 \pm 0.1\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$). We observed a direct effect of N input on $^{15}N_2O$ emissions, which lasted less than three weeks and was mainly caused by denitrification. No progressive increase in ^{15}N enrichment of N_2O occurred over a one-year experiment, which we explained by immobilisation of deposited N. The annual emission factor for N_2O from deposited N was 0.1% for the spruce stand and 0.6% for the beech stand. Standard methods used in the literature applied to the same stands grossly overestimated emission factors with values of up to 25%. Only 6–13% of the total N_2O emissions were derived from direct N deposition. Whether the remaining emissions resulted from accumulated anthropogenic N deposition or native N, can not be distinguish with the applied methods. The ^{15}N tracer technique represents a precise tool, which may improve estimates of the current contribution of N deposition on N_2O emissions.

1 Introduction

In Europe and in many other parts of the world, emissions of reactive nitrogen (N) have rapidly increased in the last decades mainly due to agricultural and industrial activities (e.g. Galloway et al., 2008; Vitousek et al., 1997). As a consequence, European forests have been exposed to high deposition rates of acidity and reactive N compounds (Berge et al., 1999). Pre-industrial European forests were supposed to be N-limited, however widespread N deposition has caused a shift in the forest's N status which under chronic N deposition may even lead to N-saturated conditions (Aber et al., 1998). Using N input-output balances, Brumme and Khanna (2008, 2009b) classified German forests into four groups with different N status that cover the complete N enrichment continuum. In their theory "steady state type forests" with mull type humus (STFa: forests where deposited N is only accumulating in trees and not in the soil) were probably widespread in Europe in pre-industrial times. They assume that soil acidification caused these forests to loose organic matter and mineral N from the soil and turned into "degradation type forests" (DTF: forests where the mineral soil has become a source of N and C). With progressing acid and N deposition, forests could turn into to "accumulation type forests" (ATF: forests where deposited N and carbon is accumulating in the upper organic soil), and finally at the end of the N enrichment continuum, they could end in another "steady state type forests" with moder type humus (STFb: forests where deposited N is only accumulating in trees and not in soil). Fluxes of nitrous oxide (N_2O), which is an important greenhouse gas (IPCC, 2007) and contributes to the chemical destruction of the stratospheric ozone layer (Crutzen, 1979), may increase if the soil becomes a source of N (transition from STFa → DTF) and deposited N exceeds the storage capacity of the ecosystem (transition from ATF → STFb). Consequently, European forests which experience chronic acid and N deposition frequently act as considerable source for N_2O (e.g. Brumme and Beese, 1992; Butterbach-Bahl et al., 1998; Papen and Butterbach-Bahl, 1999; Skiba et al., 1999).

Temperate forest soils are estimated to add between 0.1 and 2.0 Tg N_2O-N yr^{-1} to the atmosphere, which is between 0.6 and 11% of the total global N_2O emissions (Brumme et al., 2005; IPCC, 2001; Kroeze et al., 1999) and illustrates the high degree of uncertainty. The unclear role of atmospheric N deposition on N_2O emissions is one of the reasons for this uncertainty (Pilegaard et al., 2006). The IPCC (2006) uses a default emission factor for N_2O of 0.01, which means that 1% of the N deposited in temperate forests contributes to N_2O emissions. The uncertainty of this emission factor is huge ranging from 0.002 to 0.05 where the higher fraction comes from deciduous forests and the lower fraction from coniferous forests (Brumme et al., 1999; Denier van der Gon and Bleeker, 2005). Tree species induced differences in litter quality, litter structure, and soil moisture may play a role in the variation of emission factors, but much uncertainty remains (Brumme et al., 1999; Pilegaard, 2006).

Three different approaches have been used to examine the impact of N deposition on N_2O fluxes: (1) regression analysis between N deposition and N_2O fluxes (e.g. Butterbach-Bahl et al., 1998), (2) comparison of similar forest ecosystems receiving different N loads (e.g. Skiba et al., 1999), and (3) N fertilisation experiments (e.g. Brumme and Beese, 1992). These approaches have the following disadvantages: (1) regression analysis between N deposition and N_2O fluxes requires a strong correlation, which can only occur if direct N_2O emission of deposited N is considerable. Moreover, N deposition is usually correlated with factors (e.g. precipitation) that also control N_2O fluxes (Butterbach-Bahl et al., 1998; Kitzler et al., 2006a). (2) Comparing sites receiving different N loads has the disadvantage that sites are usually not completely comparable in climatic and soil properties. Finally, (3) studies have shown that in general the application of mineral N to simulate N deposition does not reflect the mode of application during chronic N deposition (Sitaula et al., 1995; Skiba and Smith, 2000).

The use of ^{15}N isotopes as a tracer is potentially a powerful tool to investigate the impact of N deposition on N_2O fluxes in forests. Tracing of ^{15}N can be applied even if low direct N_2O emissions occur; it is independent of precipitation amount and does not cause artificially high mineral N concentrations in the soil. At present we are not aware of studies where the emission factor for N_2O from forest soils was estimated using ^{15}N tracer techniques. In forest ecosystems ^{15}N tracing has only been used to distinguish between sources of N_2O production (e.g. Ambus et al., 2006; Wolf and Brumme, 2002).

Our objectives were (i) to examine the direct contribution of ammonium (NH_4^+) and nitrate (NO_3^-) deposition to N_2O emissions and (ii) to determine the one-year effect including the remineralisation of deposited and immobilised N. We hypothesised that (1) N deposition contributes considerably to direct N_2O emissions which is caused by the availability of mineral N independent of mineralisation and nitrification rates and (2) N deposition contributes considerable to one-year N_2O emissions as a result of remineralisation. To test our hypotheses, we conducted (i) a short-term as well as (ii) a one-year lasting in situ ^{15}N tracer experiment comparing a coniferous and a deciduous stand which have been exposed to high atmospheric N deposition for decades.

2 Materials and methods

2.1 Study area

The study was carried out on the Solling plateau in Central Germany (51°46′ N, 9°34′ E; 500 m a.s.l.; mean annual temperature: 6.9 °C; annual precipitation: 1193 mm). The soil type is an acidic Dystric Cambisol (silt loam), which has developed in a loess solifluction layer overlying Triassic sandstone bedrock. The humus type is a typical moder. A detailed site description is given in Bredemeier et al. (1995, 1998) and in Brumme and Khanna (2009a). Experiments took place in a 74-yr-old Norway spruce stand (*Picea abies* (L.) Karst.) and in an adjacent 160-yr-old beech stand (*Fagus sylvatica*). Soil characteristics for both sites are presented in Table 1.

Both stands have been exposed to high N deposition for decades. In 2007 and 2008, stand deposition amounted to 33 kg N ha^{-1} yr^{-1} in the spruce stand with a NH_4^+-N:NO_3^--N:DON ratio of 49:44:7 (N. Lamersdorf, personal communication, 2009) and to 20 kg N ha^{-1} yr^{-1} in the beech stand with a NH_4^+-N:NO_3^--N:DON ratio of 47:38:16 (H. Meesenburg, personal communication, 2009). Stand N deposition was usually measured in monthly intervals in both stands and represents the sum of throughfall deposition and stemflow. Stemflow was negligible in the spruce stand. In the beech stand, data collection and chemical analyses were conducted by the Northwest German Forest Research Station (Meesenburg et al., 2009).

2.2 Experimental design

We conducted the short-term experiment in August and September 2009 in order to examine the direct contribution of NH_4^+ and NO_3^- deposition to N_2O emissions. Ten chamber bases were randomly installed in each stand, four of which were labelled once with $^{15}NH_4NO_3$ solution whereas the other four were labelled once with $NH_4^{15}NO_3$ solution (both 98 atom% ^{15}N). Two chambers served as controls. We added 0.7 L labelling solution per chamber base and irrigation. The N concentration of the solution was 1.65 mg L^{-1}, which was within the range of the N concentration measured in the throughfall. The label was distributed down to 10 cm mineral soil depth within few hours after the application. In addition to the fluxes of N_2O and $^{15}N_2O$, we determined air and soil temperature, soil moisture, and extractable mineral N ($N_{min} = NH_4^+$-N and NO_3^--N) content. Soil samples were taken at randomly chosen locations between the chamber bases. Measurements were conducted 3.5 h (spruce) and 5 h (beech), 24 h, 48 h, 7 days, and 21 days after the label application.

The one-year experiment was performed to examine the impact of NH_4^+ and NO_3^- deposition on N_2O emissions including the remineralisation of the deposited N within one year. At both stands we randomly installed 17 chamber bases for gas flux measurements and soil sampling. Seven of the chamber bases received a $^{15}NH_4NO_3$ solution (98 atom% ^{15}N) and another seven received a $^{15}NH_4^{15}NO_3$ solution (95 atom% ^{15}N). Of the seven chamber bases, five were used for gas flux measurements whereas soil samples were taken in two chamber bases. The remaining three chamber bases served as controls for N_2O and $^{15}N_2O$ fluxes. Apart from the N_2O and $^{15}N_2O$ fluxes we did the same ancillary measurements as described in the short-term experiment. Measurements were done bi-weekly in the summer months and monthly in the winter months from May 2007 to June 2008. The ^{15}N solution was irrigated biweekly in the summer months and monthly during the winter months (18 applications over the year). During one irrigation event, amounts of added N and water were equal to the amounts added with one irrigation event in the one-year experiment. To each N-manipulated chamber we added the equivalent of 0.78 kg N ha^{-1} yr^{-1}, which was 2.4% of the yearly deposition in the spruce stand and 3.8% of the yearly deposition in the beech stand. Mineral nitrogen was added with the equivalent of 47 mm rain, which was approx. 4.8% of the annual precipitation. Application of ^{15}N was conducted one week before we measured gas fluxes.

2.3 N_2O and $^{15}N_2O$ measurements

Fluxes of N_2O were measured using the closed chamber method (static, vented chambers made of PVC; area: 0.2665 m^2; vol.: 18.9–29.3 L (beech), 27.5–34.7 L (spruce)). Gas samples of 100 mL were removed at three (May 2007 to January 2008) or four (February to June 2008 and 2009) regular time intervals following chamber closure (42 min at most) using a portable gas sampler equipped with a pressure sensor (Loftfield et al., 1997). Analysis of N_2O was carried out with a gas chromatograph equipped with an EC detector (GC 14A, Shimadzu, Duisburg, Germany). We calculated N_2O fluxes from the linear change of N_2O concentrations in the chamber versus time, and we adjusted fluxes for air temperature and atmospheric pressure. The mean annual flux was calculated using the trapezoid rule. Samples for $^{15}N_2O$ analysis were collected in glass bottles (100 mL) closed with a butyl-hallow stopper at time point zero and after 24 to 28 min. Analysis of ^{15}N in N_2O was done using an isotope ratio mass spectrometer coupled with a preconcentration unit (PreCon-GC-IRMS, Thermo Finnigan MAT, Bremen, Germany).

2.4 Chemical analyses

Mineral N was extracted after shaking 25 to 30 g fresh mineral soil with 100 mL 0.5 M K_2SO_4 solution or 15 g of the organic layer with 50 mL K_2SO_4 solution, using pre-washed filter papers. Extracts of K_2SO_4 were frozen until analysis. Mineral N analysis was carried out using continuous flow injection colorimetry (Cenco/Skalar Instruments, Breda, The Netherlands). The water content was determined gravimetrically.

Soil bulk density was determined using undisturbed soil samples that were oven-dried at 105 °C and sieved to 2 mm to remove stones as well as living roots. The mass of the organic layer was detected using a metal ring (594 cm^2). Organic layer samples were oven-dried at 60 °C and living roots were removed. Total carbon and nitrogen measurements were done using a CNS Elemental Analyzer (Heraeus Elementar Vario EL, Hanau, Germany) and ^{15}N concentrations using a Delta C plus isotope ratio mass spectrometer (Finnigan MAT, Bremen, Germany). The pH was determined in distilled water (1:2.5 (*v*/*v*)) using 10 mL sieved soil or shredded organic material.

2.5 Calculation of ^{15}N abundance

Isotope abundances were expressed in absolute proportion [atom%]:

$$^{15}\mathrm{N}\ [\mathrm{atom\%}] = \frac{^{15}\mathrm{N}}{15\mathrm{N} + {}^{14}\mathrm{N}} \cdot 100 \tag{1}$$

The $^{15}N_2O$ abundance of the N_2O emitted from the soil was calculated using the Keeling plot approach (Pataki et al., 2003; Tilsner et al., 2003):

$$^{15}\mathrm{N}_{\mathrm{emitted}} = \frac{^{15}\mathrm{N}_{\mathrm{chamber\ air}} \cdot c(\mathrm{N_2O})_{\mathrm{chamber\ air}} - {}^{15}\mathrm{N}_{\mathrm{ambient\ air}} \cdot c(\mathrm{N_2O})_{\mathrm{ambient\ air}}}{c(\mathrm{N_2O})_{\mathrm{chamber\ air}} - c(\mathrm{N_2O})_{\mathrm{ambient\ air}}} \tag{2}$$

where $c(N_2O)$ is the concentration of N_2O and ^{15}N is the abundance of ^{15}N in N_2O calculated as described in Eq. (1). The $^{15}N_{emitted}$ excess value was calculated by subtracting the $^{15}N_{emitted}$ value of the control chambers. The $^{15}N_{emitted}$ excess value was used to obtain the $^{15}N_2O$ excess flux. In case of negative N_2O fluxes the $^{15}N_{emitted}$ calculation was not applicable since $^{15}N_2O$ emissions occurred from labelled soil but could not be quantified because we only measured net N_2O fluxes. Hence negative N_2O fluxes were omitted for $^{15}N_2O$ flux calculations.

2.6 Recovery of ^{15}N in N_2O and N_t

The recovery of ^{15}N from the tracers in emitted N_2O (emission factor) and in soil N_t was calculated using atom% values for ^{15}N mass calculations:

$$^{15}\mathrm{N\ recovery\ [\%]} = \frac{m^{15}\mathrm{N}_{\mathrm{N_2O/N_t}}}{m^{15}\mathrm{N}_{\mathrm{Tracer}}} \cdot 100 \tag{3}$$

where $m^{15}\mathrm{N}_{\mathrm{N_2O}}$ is mass of emitted $^{15}N_2O$ excess in mg $^{15}\mathrm{N\,m^{-2}}$ per time interval considered; $\mathrm{m}^{15}\mathrm{N}_{\mathrm{Nt}}$ is mass of $^{15}N_t$ excess in mg $^{15}\mathrm{N\,m^{-2}}$ after 19 tracer applications and $m^{15}\mathrm{N}_{\mathrm{Tracer}}$ is mass of ^{15}N excess in the tracer in mg $^{15}\mathrm{N\,m^{-2}}$ applied since the start of the corresponding experiment.

We used two approaches to estimate the mass of emitted $^{15}N_2O$ between two consecutive $^{15}N_2O$ flux measurements. In the first approach, we used the trapezoid method. As indicated by the short-term experiment, this approach does not account for the peak emissions during the first week leading to an underestimation of $^{15}N_2O$ fluxes whereas fluxes in the time interval between the measurement and the next tracer application were possibly overestimated. We therefore tested a second approach where we assumed that the $^{15}N_2O$ fluxes displayed the same temporal trend as observed in the short-term experiment. In this approach, the relative proportion of the tracer emission calculated for the period between two consecutive tracer applications was estimated using the short-term experiment data. We transformed the tracer emission of a measuring day from the one-year experiment into the tracer emission of the whole period between two sequent measurements by dividing it by its relative proportion. Finally, the annual cumulative excess $^{15}N_2O$ flux was calculated by adding up the emissions of all measurement intervals. Comparison of the two approaches did not yield differences in the emission factor for a particular treatment and stand, which indicates that the higher emissions within the first week are balanced by the lower emissions within the following week(s). Therefore, we only report results from the first approach.

Nitrous oxide derived from N deposition (N_2O-NDFD) was assessed:

$$N_2O\text{-NDFD}\ [\mathrm{kg\,ha^{-1}\,yr^{-1}}] = m\mathrm{N_{dep}} \cdot \frac{\mathrm{EF}}{100} \quad (4)$$

where $m\,\mathrm{N_{dep}}$ is the mass of inorganic N of the stand deposition in kg ha^{-1} yr^{-1}; EF is the emission factor (units in %).

2.7 Statistical analyses

Prior to analysis the assumptions of normality of residuals (Shapiro-Wilk or Cramer von Mises test) and homogeneity of variances (Levene test) were tested. If necessary, we transformed (log and arcsine) data prior to analyses. Two independent sample means were tested for significant differences using the independent Student's t-test, the Welch-test, or the non parametric Mann-Whitney U-test. We used analysis of variance (ANOVA) for comparison of more than two sample means. The weighted harmonic mean was utilised for proportion data with different bases. These proportions were weighted prior to statistical analyses. Temporal pseudoreplication occurred with time series data (N_2O, $^{15}N_2O$) because measurements were repeatedly done using the same chamber bases. Therefore, we applied linear mixed effects models (Crawley, 2007; Gueorguieva and Krystal, 2004). We set up a basic model including the forest type (beech and spruce) or the different ^{15}N labelling (NH_4^+-labelled, NO_3^--labelled, NH_4NO_3-labelled) as fixed effects and the spatial replication (individual chamber) nested in time as random effects. The model was extended by a variance function and by a first-order temporal autoregressive function if the extension increased the goodness of the fit of the model. The Akaike Information Criterion was used to assess the relative goodness of the fit. Simple and multiple regressions were performed using ordinary linear regression models or, if residuals were autocorrelated, generalized least squares extended by an autoregressive moving average function. Autocorrelation was checked with the Durbin-Watson test and by plotting the autocorrelation function. A non-linear least-squares model was used to estimate parameters of the non-linear multiple regression. Effects were considered significant if $P \leq 0.05$. In the text results are reported as means ±1 standard errors. Statistical analyses were carried out using R 2.10.1 (R Development Core Team, 2009).

3 Results

3.1 Annual N_2O fluxes

Fluxes of N_2O were lower in the spruce stand than in the beech stand ($P < 0.0001$) (Fig. 1a, b) with cumulative N_2O emissions of $0.3 \pm 0.1\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$ in the spruce stand and of $2.6 \pm 0.6\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$ in the beech stand. The beech stand displayed a distinct seasonality of N_2O fluxes with high emissions during the summer months and low emissions during the winter months. Labelling did not increase N_2O fluxes since no significant differences occurred in N_2O fluxes between the NH_4^+-labelled, NH_4NO_3-labelled, and the control chambers (data not shown).

3.2 Short-term ^{15}N tracer experiment

In the spruce stand, the application of the $NH_4^{15}NO_3$ tracer caused an immediate increase in $^{15}N_2O$ fluxes (Fig. 2a). Subsequently, $^{15}N_2O$ emissions decreased again and no differences in $^{15}N_2O$ fluxes were found three weeks after the $NH_4^{15}NO_3$ tracer application compared to the reference (pre-measurement at zero hours). Application of the $^{15}NH_4NO_3$ tracer also caused an increase in $^{15}N_2O$ fluxes 3.5 h after the addition, but the increase was smaller than in the NO_3^--labelled plots (Fig. 2a). After one week no differences in $^{15}N_2O$ fluxes occurred in the NH_4^+-labelled plots of the spruce stand compared to the reference.

In the beech stand, a similar temporal pattern as in the spruce stand was found after tracer application. In the NO_3^--labelled plots, we observed an immediate strong increase in $^{15}N_2O$ fluxes (Fig. 2b). After one week, $^{15}N_2O$ fluxes had decreased and no differences were measured compared to the reference measurement. The application of the $^{15}NH_4NO_3$ tracer only slightly increased $^{15}N_2O$ fluxes but one week after the tracer addition no differences existed compared to the reference (Fig. 2b). In both spruce and beech stands, the NO_3^--labelled plots displayed three times higher EFs during the first week compared to the NH_4^+-labelled plots (Table 2). However in the beech stand, the difference decreased when calculated for the first three weeks where EFs of the NH_4^+-labelled plots were 70% of the EFs of the NO_3^--labelled plots (Table 2).

Overall, we detected no differences in $^{15}N_2O$ emissions between NH_4^+-labelled and NO_3^--labelled chambers nor in the NO_3^--labelled chambers in the beech and spruce stands. However, $^{15}N_2O$ fluxes of the NH_4^+-labelled chambers were higher in the beech stand than in the spruce stand ($P = 0.029$).

Within one stand, fluxes of total N_2O were not different in time during the short-term experiment and were 10-times higher in the beech stand ($22.8 \pm 3.6\,\mu g\,N\,m^{-2}\,h^{-1}$) than in the spruce stand ($2.2 \pm 0.5\,\mu g\,N\,m^{-2}\,h^{-1}$; $P = 0.009$). Since no changes were observed in fluxes we assumed that total N_2O fluxes were not changed by the tracer application. The same was true for the contents of NH_4^+, NO_3^- and soil moisture, which remained unchanged over time of the short-term experiment.

3.3 One-year ^{15}N tracer experiment

In the spruce stand, fluxes of $^{15}N_2O$ were higher in the NH_4NO_3-labelled treatment compared to the NH_4^+-labelled treatment ($P = 0.0014$) whereas N_2O fluxes were not different (Fig. 1a, c, Table 3). Fluxes of $^{15}N_2O$ and N_2O were positively correlated for both labellings ($P \leq 0.05$), but emissions of $^{15}N_2O$ of the NH_4NO_3-labelled treatment displayed high variability especially during the winter months when low nitrate availability may have increased the proportion of deposited nitrate of the NH_4NO_3 treatment to the $^{15}N_2O$ fluxes in the spruce stand (Fig. 1c). We did not observe a progressive enrichment of ^{15}N in N_2O during the one-year experiment. The mean annual ratio of ^{15}N-N_2O/N_2O-N was 0.04% (0.00–0.22%) for the NH_4^+-labelled and 0.21% (0.01–0.71%)

for the NH_4NO_3-labelled treatment. In the beech stand, we observed no differences in $^{15}N_2O$ flux for both treatments and they followed the same seasonal trend as the total N_2O flux in both treatments (Fig. 1b, d, Table 3). Fluxes of $^{15}N_2O$ and N_2O displayed strong correlations for both treatments ($P \leq 0.0001$) and the ^{15}N-N_2O/N_2O-N ratio averages 0.06% (0.00–0.22%) for the NH_4^+-labelled treatment and 0.19% (0.04–0.49%) for the NH_4NO_3-labelled treatment. In both treatments $^{15}N_2O$ emissions were higher in the deciduous stand than in the coniferous stand ($P \leq 0.05$; Table 3).

Using the EFs approx. 0.12% (spruce) to 0.58% (beech) of the inorganic throughfall N deposition was lost as N_2O within one year, which corresponded to 12.5% (spruce) and 5.7% (beech) of the total annual N_2O emissions (N_2O-NDFD/N_2O-N, Table 3). In the spruce stand, the EFs of the first week and first three weeks were lower than the EF of one year ($P = 0.03$) but no differences among EFs were found for the beech stand (Tables 2, 3).

We recovered a large fraction of the applied NH_4^+ in N_t of the organic layer and the upper 10 cm of the mineral soil (Table 3). In contrast the recovery of NO_3^- tracer in the N_t of the NH_4NO_3-treatment (obtained by substracting the recovery in the NH_4^+ treatment) was negligible in the beech stand and less than 10% in the spruce stand.

3.4 Relation between N_2O flux rates and throughfall N deposition

In the beech stand, N_2O fluxes were positively correlated with total throughfall N deposition ($P = 0.001$; Fig. 3, Table 4) and with NH_4^+-N deposition ($P = 0.001$; Table 4). The fraction of throughfall N deposition emitted as N_2O calculated from the slope of the regression was 25%. However, both N_2O fluxes ($P < 0.0001$; Fig. 4) and throughfall N deposition (N deposition = temperature $\cdot$ 0.09 ($\pm$0.03) + 0.70 ($\pm$0.27), $r^2 = 0.37$, $P = 0.01$, $df = 14$) also were positively related with soil temperature. Inclusion of soil temperature in the regression reduced the calculated EF from 25% to 8.6% (Fig. 4). Throughfall N deposition was furthermore positively correlated with precipitation as well ($P = 0.004$; not shown). For the spruce stand, we did not observe correlations between N_2O fluxes with N deposition, however N_2O fluxes were positively correlated with precipitation (Table 4).

We also calculated EFs according to the fraction method, where EFs are calculated by relating total N_2O emissions to N deposition. The EF obtained from this approach was 0.9% in our spruce stand and 13% in our beech stand.

4 Discussion

4.1 Annual N_2O fluxes

The relatively low N_2O fluxes from our spruce stand and the higher N_2O fluxes from our beech stand were within the range of N_2O emissions reported for other temperate coniferous and deciduous forests (Butterbach-Bahl et al., 2001; Macdonald et al., 1997; Oura et al., 2001; Zechmeister-Boltenstern, 2002; see also Table 5). Our compilation of literature (Table 5) further supports that deciduous forests generally tend to be a higher source for N_2O (>20 µg N_2O-N m^{-2} h^{-1}) than coniferous forests (<10 µg N_2O-N m^{-2} h^{-1}). We attributed the differences in N_2O fluxes between the investigated forest types to the different structure and quality (e.g. pH, C/N ratio; see Table 1) of the litter. The laminar structure of the beech litter on the surface of the moder type humus reduced gas diffusivity whereas the needle litter built a well-aerated organic layer (Ball et al., 1997). As a result, the high N_2O emissions during the summer months were produced by denitrification (Wolf and Brumme, 2002) in anaerobic micro-sites in the soil due to high oxygen consumption by microorganisms and plants at high summer temperatures (Brumme et al., 1999).

4.2 Temporal dynamics of N_2O emissions derived from N deposition

The immediate increase in $^{15}N_2O$ fluxes that occurred within 3.5–5 h in the short-term experiment after $NH_4^{15}NO_3$ and $^{15}NH_4NO_3$ tracer application illustrates that deposited

N contributed promptly to N_2O production. However, the contribution of N deposition on N_2O fluxes decreased within the following three weeks. In the spruce stand release of N_2O derived from the ^{15}N label occurred over a longer period compared to the beech stand, in which loss of N input as N_2O almost completely occurred in the first three weeks following deposition. We related this short-lived increase to biotic and abiotic immobilisation and dilution processes of labelled N. In contrast to our hypotheses, repeated application of ^{15}N label did not lead to a progressive increase in ^{15}N enrichment of N_2O during the one-year experiment, which indicates that the main effect of N deposition on N_2O fluxes occurs when the mineral N enters the soil system ("short-lived effects"). We explained this lack of "medium-term" effect by N immobilisation (Corre et al., 2007), which prevented remobilisation of the deposited N in the soil over the course of one year. Output analysis indicates that both systems predominantly retained the deposited nitrogen despite N deposition in excess of plant increment (Brumme and Khanna, 2009b; Feng et al., 2008). Our recovery of NH_4NO_3 tracer in soil N_t also showed that a great proportion of N deposition was retained in the soil.

4.3 Processes of N_2O emissions derived from N deposition

The stronger increase in $^{15}N_2O$ fluxes after nitrate tracer application in contrast to ammonium suggests that denitrification was the dominant process for the input-derived emissions at both stands. Water addition probably created additional anaerobic microsites resulting in favourable conditions for denitrification, which mimics conditions when natural wet N deposition occurs. Denitrification was also found to be the dominant process at an adjacent beech stand at the Solling site (Wolf and Brumme, 2002). The weaker $^{15}N_2O$ flux increase that we observed after $^{15}NH_4NO_3$ tracer application was probably caused by low nitrifiers activity in these acid soils and the weak competition of nitrifiers for available NH_4^+ (Corre and Lamersdorf, 2004; Rennenberg et al., 1998). Fertilisation with $(NH_4)_2SO_4$ also triggered a delayed increase in N_2O emissions of 14 days (Brumme and Beese, 1992), which may have been the time needed for nitrification to process applied NH_4^+ before denitrification caused an increase in N_2O fluxes. In contrast, rapid increases in N_2O fluxes have been reported after fertilisation with NO_3^--containing fertilisers of coniferous forests (Klemedtsson et al., 1997; Macdonald et al., 1997; Sitaula et al., 1995).

The ^{15}N-N_2O/N_2O-N ratio of 0.05% for the NH_4^+-labelled treatment and of 0.20% for the NH_4NO_3-labelled treatment for both stands of the one-year experiment suggests that the contribution of NH_4^+ to N_2O emissions in the NH_4NO_3 treatment was about 25% whereas NO_3^- contributed about 75%. This result is in line with findings by Ambus et al. (2006) who showed that 62% of N_2O emissions in 11 European forests were derived from NO_3^- and 34% were derived from NH_4^+.

4.4 Contribution of N deposition to N_2O emissions

The calculated EF depended on the method used, which also emerged from the compilation of published studies that determined the impact of N input on N_2O fluxes in temperate forests (Table 5). The EF_R (calculated by the regression method) of 25% for the beech stand is probably an artefact of the regression approach since N deposition depends on other factors which also control N_2O fluxes and this will lead to systematic errors. The reduction of the calculated EF_R from 25% to 8.6% if soil temperature was considered in the regression approach, illustrates this problem. It is therefore no surprise that emission factors obtained from this approach are among the highest reported: a study in southern Germany calculated an EF_R of 10% for a beech stand (5.1 kg N_2O-N ha^{-1} yr^{-1}) and 0.5% for a spruce stand (1.4 kg N_2O-N ha^{-1} yr^{-1}) using the regression approach (Papen and Butterbach-Bahl, 1999; Table 5). Denier van der Gon and Bleeker (2005) combined results from several studies conducted in temperate forests and found EF_R of 6.3% for deciduous forests and of 1.4% for coniferous forests. A positive correlation of $N_2O + NO$ fluxes with N deposition was further detected for 15 European forests exposed to different rates of N deposition (Pilegaard et al., 2006). Between 2% and 32% of total N deposition were emitted as $N_2O + NO$.

Title Page
Abstract Introduction
Conclusions References
Tables Figures
◀| |▶
◀ ▶
Back Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

Our EF_F obtained by the fraction method of 0.9% (spruce) and 13% (beech) are also higher than the EF_{15N} derived from our ^{15}N tracer study of 0.1% (spruce) and 0.6% (beech) in our study. A higher EF_{FB} of 1.6% was further calculated for a long-term fertilisation experiment at the same stand where 140 kg N ha^{-1} yr^{-1} were applied over a 5–6 years period (Table 5; Brumme and Beese, 1992). The EF_{FB} considers background emissions from unfertilised plots in fertilisation experiments or from a forest site exposed to lower N deposition than another site in experiments were similar forest sites are compared. Emission factors EF_F show a wide range from <0.1% to 22% for both forest types (Table 5). Using the fraction method, Denier van der Gon and Bleeker (2005) found an average EF_F of 2.4% for coniferous forests and of 6.5% for deciduous forests. The EF_{FB} based on fertilised plots ranged from 0.1% to 0.9% for the coniferous forests and from 0.03% to 1.6% for the deciduous forests (Table 5).

What causes the large differences in calculated EFs when applying different methods? The relative small proportion of N_2O derived from inorganic N deposition of 13% in our spruce stand and 6% in our beech stand shows that most of the nitrogen in N_2O emissions is not derived from direct N deposition, but results from N actively cycling in the forest ecosystem, which was not deposited during the past year (background emissions). As a result, the EF_F of the fraction method will overestimate direct N_2O emissions from deposition because it does not correct for possible N_2O background emissions. Emission factors EF_{FB} based on fertilised plots are in the same order of magnitude as our EF_{15N} obtained by the ^{15}N tracer method. However, strong increases in N_2O fluxes from the fertilised plots compared to the unfertilised control plots have been reported (Table 5), which may be a result from the high mineral N concentrations following fertilizer application, which does no reflect conditions during atmospheric N deposition (Sitaula et al., 1995; Skiba and Smith, 2000) and may cause a positive priming effect (e.g. Fenn et al., 1998). The implicit assumption of the regression approach as well is that the N_2O emissions from N cycling are not affected by N deposition. However, this assumption may be violated and lead to artificially high estimates since both N deposition and N_2O fluxes are correlated e.g. to rainfall amount: at times of high rainfall (and thus, high N deposition), high soil water content will increase denitrification and therefore N_2O fluxes, which will originate mainly from N cycling in the soil. We demonstrated that only the EF_{15N} obtained by the ^{15}N tracer study detect the direct emissions whereas the other approaches overestimated the direct emissions and did not comprise the background emissions. The results of EF estimations can be summarised as follows:

$$EF_{15N} \approx EF_{FB} \ll EF_F \approx EF_R \tag{5}$$

The low direct contribution of atmospheric deposition to N_2O emission raised the question whether the background emissions are affected by decade long atmospheric deposition, which may have been accumulated in the ecosystem. N cycling rates can vary considerable in similar forest stands, which may be the result of long-term N deposition (Corre et al., 2007). With the methods that we applied the effect of accumulated anthropogenic N on the background N_2O emissions cannot be determined. Our literature compilation shows, however, that in general soils with a similar forest type which are exposed to higher N deposition are also characterised by higher N_2O emissions than soils exposed to lower N deposition (Table 5). Assuming that N_2O emissions were negligible before the onset of anthropogenic activity, the total contribution of N deposition to N_2O emission considering direct and background emissions is probably best described by the fraction of N_2O to current N deposition.

5 Conclusions

The ^{15}N tracer method turned out to be a precise approach for quantifying the direct contribution of atmospheric N deposition on the emission of N_2O. The technique allows the simulation of atmospheric throughfall N deposition without artificial fertilisation and provides the possibility to investigate the impact of N deposition on N_2O emissions in forests also when direct emissions are low. In contrast to the regression approach, the method does not result in artefacts caused by controlling parameters (like rainfall or temperature), which influence both N_2O fluxes and N deposition. There are no uncertainties resulting from the comparison of different sites (as when regarding deposition gradients), which are usually not completely comparable in edaphic and soil conditions. Furthermore, EFs from fertilised plots may results in artificially high N_2O emissions because the pulse in mineral N concentrations do not reflect conditions during atmospheric N deposition and may cause positive priming effects. Finally, in comparison to the fraction method, only direct emissions are considered using the ^{15}N technique. Most of the other methods overestimate the direct emissions and are not able to estimate the background emissions just as the ^{15}N tracer method.

Acknowledgements. The authors thank Henning Meesenburg and Norbert Lamersdorf for providing the precipitation and deposition data. This work was financially supported by the German Research Foundation (BR 1524/6-1).

References

Aber, J., McDowell, W., Nadelhoffer, K., Magill, A., Berntson, G., Kamakea, M., McNulty, S., Currie, W., Rustad, L., and Fernandez, I. J.: Nitrogen saturation in temperate forest ecosystems, Bioscience, 48(11), 921–934, 1998.

Ambus, P., Zechmeister-Boltenstern, S., and Butterbach-Bahl, K.: Sources of nitrous oxide emitted from European forest soils, Biogeosciences, 3, 135–145, doi:10.5194/bg-3-135-2006, 2006.

Ball, B. C., Smith, K. A., Klemedtsson, L., Brumme, R., Sitaula, B. K., Hansen, S., Prieme, A., MacDonald, J., and Horgan, G. W.: The influence of soil gas transport properties on methane oxidation in a selection of northern European soils, J. Geophys. Res., 102(D19), 23309–23317, 1997.

Beier, C., Rasmussen, L., Pilegaard, K., Ambus, P., Mikkelsen, T., Jensen, N. O., Kjoller, A., Prieme, A., and Ladekarl, U. L.: Fluxes of NO_3^-, NH_4^+, NO, NO_2, and N_2O in an old Danish beech forest, Water Air Soil Poll., 1(1), 187–195, 2001.

Berge, E., Bartnicki, J., Olendrzynski, K., and Tsyro, S. G.: Long-term trends in emissions and transboundary transport of acidifying air pollution in Europe, J. Environ. Manage., 57(1), 31–50, 1999.

Bowden, R. D., Melillo, J. M., Steudler, P. A., and Aber, J. D.: Effects of nitrogen additions on annual nitrous oxide fluxes from temperate forest soils in the northeastern United States, J. Geophys Res., 96(D5), 9321–9328, 1991.

Bredemeier, M., Blanck, K., Lamersdorf, N., and Wiedey, G. A.: Response of soil water chemistry to experimental "clean rain" in the NITREX roof experiment at Solling, Germany, Forest Ecol. Manag., 71, 31–44, 1995.

Bredemeier, M., Blanck, K., Dohrenbusch, A., Lamersdorf, N., Meyer, A. C., Murach, D., Parth, A., and Xu, Y.-J.: The Solling roof project – site characteristics, experiments and results, Forest Ecol. Manag., 101, 281–293, 1998.

Brumme, R. and Beese, F.: Effects of liming and nitrogen fertilization on emissions of CO_2 and N_2O from a temperate forest, J. Geophys. Res., 97(D12), 12851–12858, 1992.

Brumme, R., Borken, W., and Finke, S.: Hierarchical control on nitrous oxide emission in forest ecosystems, Global Biogeochem. Cy., 13(4), 1137–1148, 1999.

Brumme, R., Verchot, L. V., Martikainen, P. J., and Potter, C. S.: Contribution of trace gases nitrous oxide (N_2O) and methane (CH_4) to the atmospheric warming balance of forest biomes, in: The carbon balance of forest biomes, edited by: Griffiths, H. and Jarvis, P. G., Thomson Publishing Services, Hampshire, UK, 293–317, 2005.

Brumme, R. and Khanna, P. K.: Ecological and site historical aspects of N dynamics and current N status in temperate forests, Glob. Change Biol., 14(1), 125–141, 2008.

Brumme, R. and Khanna, P. K. (Eds.): Functioning and management of European beech ecosystems, Ecol Stud, 208, Springer-Verlag, Berlin Heidelberg, 490 pp., 2009a.

Brumme, R. and Khanna, P. K.: Chapter 23: Stand, soil and nutrient factors determining the functioning and management of beech forest ecosystems: A Synopsis, in: Functioning and management of European beech ecosystems, edited by: Brumme, R. and Khanna, P. K., Springer-Verlag, Berlin Heidelberg, 459–490, 2009b.

Butterbach-Bahl, K., Gasche, R., Huber, C., Kreutzer, K., and Papen, H.: Impact of N-input by wet deposition on N-trace gas fluxes and CH_4-oxidation in spruce forest ecosystems of the temperate zone in Europe, Atmos. Environ., 32(3), 559–564, 1998.

Butterbach-Bahl, K., Stange, F., Papen, H., and Li, C. S.: Regional inventory of nitric oxide and nitrous oxide emissions for forest soils of southeast Germany using the biogeochemical model PnET-N-DNDC, J. Geophys. Res., 106(D24), 34155–34166, 2001.

Butterbach-Bahl, K., Breuer, L., Gasche, R., Willibald, G., and Papen, H.: Exchange of trace gases between soils and the atmosphere in Scots pine forest ecosystems of the northeastern German lowlands, 1. Fluxes of N_2O, NO/NO_2 and CH_4 at forest sites with different N-deposition, Forest Ecol. Manag., 167(1–3), 123–134, 2002.

Castro, M., Steudler, P., Melillo, J., Aber, J., and Millham, S.: Exchange of N_2O and CH_4 between the atmosphere and soils in spruce-fir forests in the northeastern United States, Biogeochemistry, 18(3), 119–135, 1993.

Corre, M. D. and Lamersdorf, N. P.: Reversal of nitrogen saturation after long-term deposition reduction: impact on soil nitrogen cycling, Ecology, 85(11), 3090–3104, 2004.

Corre, M. D., Brumme, R., Veldkamp, E., and Beese, F. O.: Changes in nitrogen cycling and retention processes in soils under spruce forests along a nitrogen enrichment gradient in Germany, Glob. Change Biol., 13(7), 1509–1527, 2007.

Crawley, M. J. (Ed.): The R Book, John Wiley & Sons Ltd, Chichester, 942 pp., 2007.

Crutzen, P. J.: The Role of NO and NO_2 in the chemistry of the troposphere and stratosphere, Annu. Rev. Earth Pl. Sc., 7(1), 443–472, 1979.

Denier van der Gon, H. and Bleeker, A.: Indirect N_2O emission due to atmospheric N deposition for the Netherlands, Atmos. Environ., 39(32), 5827–5838, 2005.

Feng, Z., Brumme, R., Xu, Y.-J., and Lamersdorf, N.: Tracing the fate of mineral N compounds under high ambient N deposition in a Norway spruce forest at Solling/Germany, Forest Ecol. Manag., 255, 2061–2073, 2008.

Fenn, M. E., Poth, M. A., Aber, J. D., Baron, J. S., Bormann, B. T., Johnson, D. W., Lemly, A. D., McNulty, S. G., Ryan, D. F., and Stottlemyer, R.: Nitrogen excess in North American ecosystems: Predisposing factors, ecosystem response, and management Strategies, Ecol. Appl., 8(3), 706–733, 1998.

Galloway, J. N., Townsend, A. R., Erisman, J. W., Bekunda, M., Cai, Z. C., Freney, J. R., Martinelli, L. A., Seitzinger, S. P., and Sutton, M. A.: Transformation of the nitrogen cycle: Recent trends, questions, and potential solutions, Science, 320(5878), 889–892, 2008.

Gueorguieva, R. and Krystal, J. H.: Move over ANOVA, progress in analyzing repeated-measures data and its reflection in papers published in the archives of general psychiatry, Arch. Gen. Psychiat., 61, 310–317, 2004.

IPCC: Chapter 4: Atmospheric chemistry and greenhouse gases, in: Climate Change 2001: Working Group I: The scientific basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., Dai, X., Maskell, K., and Johnson, C. A., Cambridge University Press, Cambridge, UK and New York, USA, 2001.

IPCC: Chapter 11: N_2O Emissions from managed soils, and CO_2 emissions from lime and urea application, in: IPCC guidelines for national greenhouse gas inventories, edited by: Eggleston, H. S., Buendia, L., Miwa, K., Ngara, T., and Tanabe, K., IGES, Hayama, Japan, 2006.

IPCC: Changes in atmospheric constituents and in radiative forcing, in: Climate Change 2007: The physical science basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge University Press, Cambridge, UK and New York, USA, 2007.

Kitzler, B., Zechmeister-Boltenstern, S., Holtermann, C., Skiba, U., and Butterbach-Bahl, K.: Nitrogen oxides emission from two beech forests subjected to different nitrogen loads, Biogeosciences, 3, 293–310, doi:10.5194/bg-3-293-2006, 2006a.

Kitzler, B., Zechmeister-Boltenstern, S., Holtermann, C., Skiba, U., and Butterbach-Bahl, K.: Controls over N_2O, NO_x and CO_2 fluxes in a calcareous mountain forest soil, Biogeosciences, 3, 383–395, doi:10.5194/bg-3-383-2006, 2006b.

Klemedtsson, L., Klemedtsson, A. K., Moldan, F., and Weslien, P.: Nitrous oxide emission from Swedish forest soils in relation to liming and simulated increased N-deposition, Biol. Fert. Soils, 25(3), 290–295, 1997.

Kroeze, C., Mosier, A., and Bouwman, L.: Closing the global N_2O budget: A retrospective analysis 1500–1994, Global Biogeochem. Cy., 13(1), 1–8, 1999.

Loftfield, N., Flessa, H., Augustin, J., and Beese, F.: Automated gas chromatographic system for rapid analysis of atmospheric trace gases methane, carbon dioxide, and nitrous oxide, J. Environ. Qual., 26, 560–564, 1997.

Macdonald, J. A., Skiba, U., Sheppard, L. J., Ball, B., Roberts, J. D., Smith, K. A., and Fowler, D.: The effect of nitrogen deposition and seasonal variability on methane oxidation and nitrous oxide emission rates in an upland spruce plantation and moorland, Atmos. Environ., 31(22), 3693–3706, 1997.

Meesenburg, H., Eichhorn, J., and Meiwes, K. J.: Chapter 15: Atmospheric deposition and canopy interactions, in: Functioning and management of European beech ecosystems, edited by: Brumme, R. and Khanna, P. K., Springer-Verlag, Berlin Heidelberg, 265–302, 2009.

Oura, N., Shindo, J., Fumoto, T., Toda, H., and Kawashima, H.: Effects of nitrogen deposition on nitrous oxide emissions from forest floor, Water Air Soil Poll., 130, 673–678, 2001.

Papen, H. and Butterbach-Bahl, K.: A 3-year continuous record of nitrogen trace gas fluxes from untreated and limed soil of a N-saturated spruce and beech forest ecosystem in Germany – 1. N_2O emissions, J. Geophys. Res., 104(D15), 18487–18503, 1999.

Papen, H., Daum, M., Steinkamp, R., and Butterbach-Bahl, K.: N_2O and CH_4-fluxes from soils of a N-limited and N-fertilized spruce forest ecosystem of the temperate zone, J. Appl. Bot., 75(3–4), 159–163, 2001.

Pataki, D. E., Ehleringer, J. R., Flanagan, L. B., Yakir, D., Bowling, D. R., Still, C. J., Buchmann, N., Kaplan, J. O., and Berry, J. A.: The application and interpretation of Keeling plots in terrestrial carbon cycle research, Global Biogeochem. Cy., 17(1), 1022–1029, 2003.

Pilegaard, K., Skiba, U., Ambus, P., Beier, C., Brüggemann, N., Butterbach-Bahl, K., Dick, J., Dorsey, J., Duyzer, J., Gallagher, M., Gasche, R., Horvath, L., Kitzler, B., Leip, A., Pihlatie, M. K., Rosenkranz, P., Seufert, G., Vesala, T., Westrate, H., and Zechmeister-Boltenstern, S.: Factors controlling regional differences in forest soil emission of nitrogen oxides (NO and N_2O), Biogeosciences, 3, 651–661, doi:10.5194/bg-3-651-2006, 2006.

R Development Core Team: R: A language and environment for statistical computing, R Foundation for Statistical Computing, Vienna, Austria, ISBN 3-900051-07-0, available at: http://www.R-project.org, 2009.

Rennenberg, H., Kreutzer, K., Papen, H., and Weber, P.: Consequences of high loads of nitrogen for spruce (*Picea abies*) and beech (*Fagus sylvatica*) forests, New Phytol., 139(1), 71–86, 1998.

Sitaula, B. K., Bakken, L. R., and Abrahamsen, G.: N-Fertilization and Soil Acidification Effects on N_2O and CO_2 Emission from Temperate Pine Forest Soil, Soil Biol. Biochem., 27(11), 1401–1408, 1995.

Skiba, U., Sheppard, L., Pitcairn, C. E. R., Leith, I., Crossley, A., van Dijk, S., Kennedy, V. H., and Fowler, D.: Soil nitrous oxide and nitric oxide emissions as indicators of elevated atmospheric N deposition rates in seminatural ecosystems, Environ. Pollut., 102, 457–461, 1998.

Skiba, U., Sheppard, L. J., Pitcairn, C. E. R., van Dijk, S., and Rossall, M.: The effect of N deposition on nitrous oxide and nitric oxide emissions from temperate forest soils, Water Air Soil Poll., 116, 89–98, 1999.

Skiba, U. and Smith, K. A.: The control of nitrous oxide emissions from agricultural and natural soils, Chemosphere, 2, 379–386, 2000.

Tilsner, J., Wrage, N., Lauf, J., and Gebauer, G.: Emission of gaseous nitrogen oxides from an extensively managed grassland in NE Bavaria, Germany II. Stable isotope natural abundance of N_2O, Biogeochemistry, 63(3), 249–267, 2003.

Venterea, R. T., Groffman, P. M., Verchot, L. V., Magill, A. H., Aber, J. D., and Steudler, P. A.: Nitrogen oxide gas emissions from temperate forest soils receiving long-term nitrogen inputs, Glob. Change Biol., 9(3), 346–357, 2003.

Vitousek, P. M., Aber, J. D., Howarth, R. W., Likens, G. E., Matson, P. A., Schindler, D. W., Schlesinger, W. H., and Tilman, D. G.: Human alteration of the global nitrogen cycle: sources and consequences, Ecol. Appl., 7(3), 737–750, 1997.

Wolf, I. and Brumme, R.: Contribution of nitrification and denitrification sources for seasonal N_2O emissions in acid German forest soil, Soil Biol Biochem, 34, 741–744, 2002.

Zechmeister-Boltenstern, S., Hahn, M., Meger, S. and Jandl, R.: Nitrous oxide emissions and nitrate leaching in relation to microbial biomass dynamics in a beech forest soil, Soil Biol. Biochem., 34(6), 823–832, 2002.

Table 1. Soil characteristics of the spruce and beech stands at the Solling site.

Soil depth and forest site		*n*	pH(H_2O)	*n*	Total organic carbon [mg C g^{-1}]	Total nitrogen [mg N g^{-1}]	C/N ratio	*n*	Bulk density [g cm^{-3}]	Humus mass [Mg ha^{-1}]
Organic layer	Spruce	3	3.6 (0.1)^a	13	447 (13)	16.5 (0.4)^a	27.2 (0.6)^a	6		67.1 (4.4)^a
	Beech	3	4.0 (0.0)^b	13	443 (12)	18.6 (0.7)^b	24.1 (0.7)^b	6		35.6 (1.9)^b
0–5 cm	Spruce	6	3.3 (0.0)	14	112 (7)	4.6 (0.3)	24.3 (0.8)^a	4	0.76 (0.03)	
	Beech	6	3.4 (0.0)	14	98 (11)	4.4 (0.3)	21.7 (1.4)^b	6	0.89 (0.02)	
5–10 cm	Spruce	5	3.5 (0.0)	13	39 (2)^a	1.7 (0.1)	22.9 (0.8)^a	5	1.00 (0.02)	
	Beech	6	3.6 (0.0)	13	32 (2)^b	1.7 (0.1)	19.8 (1.1)^b	6	1.08 (0.04)	

At each layer, means (±SE) followed by different letters indicated differences among the spruce and beech stands (independent t-test or Mann-Whitney U-test at $P \leq 0.05$).

Table 2. Emission factors (EFs) of N_2O during the first week and the first three weeks following tracer application in the spruce and beech stands. The emission factors were derived from the NH_4^+-labelled and NO_3^--labelled treatments of the short-term experiment ($n = 4$).

	EF (1 week) [%]			EF (3 weeks) [%]		
Tracer	NH_4^+	NO_3^-	$NH_4NO_3^*$	NH_4^+	NO_3^-	$NH_4NO_3^*$
Spruce	0.01 (0.00)	0.03 (0.01)	0.02 (0.01)	0.02 (0.01)	0.05 (0.02)	0.04 (0.01)
Beech	0.12 (0.09)	0.31 (0.18)	0.22 (0.10)	0.43 (0.33)	0.62 (0.33)	0.54 (0.22)

Means (±SE); no significant differences were detected among stands, labellings, and EFs.

[*] The EF for the NH_4NO_3-labelled treatment was obtained by calculating the mean of the NH_4^+-labelled and NO_3^--labelled treatments of the short-term experiment ($n = 8$).

Table 3. Cumulative N_2O and $^{15}N_2O$ emissions, one-year emission factor (EF), N_2O derived from deposition (N_2O-NDFD), ratio of N_2O-NDFD to total N_2O emissions, and recovery of ^{15}N tracer in N_t of the organic layer and the upper 10 cm of mineral soil. Results are from the NH_4^+-labelled and NH_4NO_3-labelled treatments of the one-year experiment.

	Labelling	N_2O [kg N ha^{-1} yr^{-1}]	$^{15}N_2O$ [g ^{15}N ha^{-1} yr^{-1}]	EF [%]	N_2O-NDFD [g N ha^{-1} yr^{-1}]	N_2O-NDFD/ N_2O-N* [%]	Recovery in N_t [%]
Spruce	NH_4^+	0.26 (0.06)^{aA}	0.12 (0.04)^{aA}	0.03 (0.01)^{aA}	9.4 (3.0)^{aA}	3.6 (1.1)^{aA}	59.2 (11.6)^{aA}
	NH_4NO_3	0.30 (0.13)^{aA}	0.91 (0.47)^{aB}	0.12 (0.06)^{aA}	37.9 (19.3)^{aA}	12.5 (6.4)^{aA}	36.6 (6.6)^{aA}
Beech	NH_4^+	3.15 (1.26)^{bA}	1.79 (1.10)^{bA}	0.46 (0.29)^{aA}	79.6 (49.0)^{aA}	2.5 (1.6)^{aA}	41.7 (7.2)^{aA}
	NH_4NO_3	1.75 (1.90)^{bA}	4.28 (2.18)^{bA}	0.58 (0.29)^{aA}	99.3 (50.5)^{aA}	5.7 (2.9)^{aA}	20.7 (3.1)^{aB}

Means (±SE) ($n = 5$ for N_2O, $^{15}N_2O$, EF, N_2O-NDFD and N_2O-NDFD/N_2O-N and $n = 4$ for recovery in N_t) followed by the same lower-case letters indicated no significant differences among both stands and the same labelling treatments. Means followed by the same capital letters indicated no significant differences among different labelling treatments within one stand (independent t-test at $P \leq 0.05$).

[*] The ratio N_2O-NDFD/N_2O-N represented the weighted harmonic mean with corresponding SE.

Table 4. Results of the regression analyses of N_2O-N fluxes and throughfall N deposition (total N, NH_4^+-N, NO_3^--N, and organic N deposition) and precipitation.

	N_{total}			NH_4^+-N			NO_3^--N			Norg			Precipitation		
	r^2	*P*	*df*	r^2	*P*	*df*	r^2	*P*	*df*	r^2	*P*	*df*	r^2	*P*	*df*
Spruce	–	ns	–	–	ns	–	–	ns	–	–	ns	–	0.30	0.042	12
Beech	0.53	0.001	14	0.53	0.001	14	–	ns	–	–	ns	–	–	ns	–

ns is not significant

Table 5. Compilation of published studies that investigated the impact of N input on N_2O fluxes from deciduous and coniferous temperate forest soils. Studies are divided into fertilisation, deposition, and ^{15}N tracer experiments. The emission factor (EF) for N_2O is given for one year if not differently indicated.

Forest type	Type of N input	N_2O [µg N m^{-2} h^{-1}]	Emission factor [%]	Method used	Treatment	Reference
Deciduous	Fertilisation	0.68	0.03[e]	EF_{FB}	NH_4NO_3-fertilised (120 (1. year) and 150 (2. year) kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 2. year; USA)	Bowden et al. (1991)
		0.57	0.06[e]	EF_{FB}	NH_4NO_3-fertilised (37 (1. year) and 50 (2. year) kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 2. year; USA)	
		0.23			Unfertilised (USA)	
		89	1.6	EF_{FB}	$(NH_4)_2SO_4$-fertilised (140 kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 6. and 7. year; Germany)	Brumme and Beese (1992)
		64	16[e]	EF_F	Unfertilised (35 kg N ha^{-1} yr^{-1} deposition; Germany)	
		< 10	< 0.3	EF_F	NH_4NO_3-fertilised (150 kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 13. year; USA)	Venterea et al. (2003)
		< 10	< 0.3	EF_F	NH_4NO_3-fertilised (50 kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 13. year; USA)	
		< 10	< 0.3	EF_F	Unfertilised (8 kg N ha^{-1} yr^{-1} wet + dry deposition; USA)	
	Deposition	5.7	2[e]	EF_F	25.6 kg N ha^{-1} yr^{-1} throughfall + stemflow dep. (Denmark)	Beier et al. (2001)
		83.3	22[e]	EF_F	33 kg N ha^{-1} yr^{-1} wet deposition (Germany)	Brumme et al. (1999)
		9.1	2.4[e]	EF_F	33 kg N ha^{-1} yr^{-1} wet deposition (Germany)	
		1.9	0.6[e]	EF_F	28 kg N ha^{-1} yr^{-1} throughfall deposition (Germany)	
		7.2	2.4[e]	EF_F	26 kg N ha^{-1} yr^{-1} throughfall deposition (Germany)	
		4.7	1.9[e]	EF_F	21 kg N ha^{-1} yr^{-1} throughfall deposition (Germany)	
		7.3–9.0	~ 3.5[e]	EF_F	20.2 kg N ha^{-1} yr^{-1} wet deposition (Austria)	Kitzler et al. (2006a)[a]
		5.9–7.4	~ 4.6[e]	EF_F	12.6 kg N ha^{-1} yr^{-1} wet deposition (Austria)	
		2.3	1.27	EF_F	15.7 kg N ha^{-1} yr^{-1} throughfall deposition (Japan)	Oura et al. (2001)
		58.4	10	EF_R	20 kg N ha^{-1} yr^{-1} wet deposition (Germany)	Papen and Butterbach-Bahl (1999)
		29.7	25; 13	EF_R, EF_F	20 kg N ha^{-1} yr^{-1} throughfall + stemflow dep. (Germany)	This study
	^{15}N tracer	29.7	0.6	EF_{15N}	20 kg N ha^{-1} yr^{-1} throughfall + stemflow dep. (Germany)	This study

Table 5. Continued.

Forest type	Type of N input	N_2O [µg N m^{-2} h^{-1}]	Emission factor [%]	Method used	Treatment	Reference
Coniferous	Fertilisation	4.0	0.1^e	EF_{FB}	NH_4NO_3-fertilised (120 (1. year) and 150 (2. year) kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 2. year; USA)	Bowden et al. (1991)
		3.0	0.5^e	EF_{FB}	NH_4NO_3-fertilised (37 (1. year) and 50 (2. year) kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 2. year; USA)	
		0.8			Unfertilised (USA)	
		3.11	0.35	EF_F	NH_4Cl-fertilised (31.4 kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the beginning of the 3. year (only growing season), estimated annual N_2O flux of 0.11 kg N ha^{-1} yr^{-1}); USA)	Castro et al. (1993)
		−1.12			Unfertilised (10 kg N ha^{-1} yr^{-1} wet + dry deposition; measurement only in the growing season; USA	
		1.1 (drained); 2.9 (wet)	0.1 (drained) 0.6 (wet)^e	EF_{FB}	NH_4NO_3-fertilised (35 kg N ha^{-1} yr^{-1} in small doses; chronic N addition exp.; measurement after 2 years; Sweden)	Klemedtsson et al. (1997)
		0.8 (drained); 1.3 (wet)			Unfertilised (12 kg N ha^{-1} yr^{-1} deposition; Sweden)	
		0.9	$< 0.1^e$	EF_F	$(NH_4)_2SO_4$-fertilised (150 kg N ha^{-1}; single dose; measurement in the first 3 years; Germany)	Papen et al. (2001)
		−1.0			Unfertilised (Germany)	
		45.8	0.93 (1 month)	EF_{FB}	NH_4NO_3-fertilised (90 kg N ha^{-1}; single dose; measurement over 1 month; Norway)	Sitaula et al. (1995)[b]
		21.7	0.94 (1 month)	EF_{FB}	NH_4NO_3-fertilised (30 kg N ha^{-1}; single dose; measurement over 1 month; Norway)	
		8.3			Unfertilised (Norway)	
		5.7	0.6^e	EF_{FB}	Acid mist-fertilised (96 kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement after 2 years; UK)	Skiba et al. (1998, 1999)
		0.5	0.2^e	EF_{FB}	Acid mist-fertilised (48 kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 3. year, UK)	
		−0.3			Unfertilised (6.7 kg N ha^{-1} yr^{-1} wet + dry deposition; UK)	
		< 10	< 0.3	EF_F	NH_4NO_3-fertilised (150 kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 13. year; USA)	Venterea et al. (2003)
		< 10	< 0.3	EF_F	NH_4NO_3-fertilised (50 kg N ha^{-1} yr^{-1}; chronic N addition exp.; measurement in the 13. year; USA)	
		< 10	< 0.3	EF_F	Unfertilised (8 kg N ha^{-1} yr^{-1} wet + dry deposition; USA)	

Table 5. Continued.

Forest type	Type of N input	N_2O [µg N m^{-2} h^{-1}]	Emission factor [%]	Method used	Treatment	Reference
	Deposition	2.9	0.6^e	EF_F	41 kg N ha^{-1} yr^{-1} throughfall deposition (Germany)	Brumme et al. (1999)
		2.4	0.7^e	EF_F	31 kg N ha^{-1} yr^{-1} throughfall deposition (Germany)	
		14.8	6.5^e	EF_F	20 kg N ha^{-1} yr^{-1} throughfall deposition without N_{org} (Germany)	
		4–15	$1.2–4.4^e$	EF_F	30 kg N ha^{-1} yr^{-1} wet deposition (Germany)	Butterbach-Bahl et al. (1998)[a]
		−0.5 – 2.1	$\sim 1.3^e$	EF_F	5–6 kg N ha^{-1} yr^{-1} wet deposition (Ireland)	
		16–32	$\sim 7 – 13^e$	EF_F	20–22 kg N ha^{-1} yr^{-1} wet deposition (Germany)	Butterbach-Bahl et al. (2002)[a]
		5–10	$\sim 4.4^e$	EF_F	~ 15 kg N ha^{-1} yr^{-1} wet deposition (Germany)	
		3.4–4.7	2.5–3.5	EF_F	10.6–11.9 kg N ha^{-1} yr^{-1} bulk deposition (Austria)	Kitzler et al. (2006b)[c]
		3.8	~ 1	EF_{FB}	~ 46.2 kg N ha^{-1} yr^{-1} total deposition (UK)	Macdonald et al. (1997)
		1.3			6.4 kg N ha^{-1} yr^{-1} total deposition (UK)	
		4.3	1.23	EF_F	30.6 kg N ha^{-1} yr^{-1} throughfall deposition (Japan)	Oura et al. (2001)
		16.4	0.5	EF_R	30 kg N ha^{-1} yr^{-1} wet deposition (Germany)	Papen and Butterbach-Bahl (1999)
		56	6	EF_{FB}	80.1 kg N ha^{-1} yr^{-1} NH_3 dep from poultry farm; 30 m downwind from farm (UK)	Skiba et al. (1998, 1999)[d]
		13			17.4 kg N ha^{-1} yr^{-1} NH_3 dep from poultry farm; 250 m down-wind from farm (UK)	
		3.4	0.9	EF_F	33 kg N ha^{-1} yr^{-1} throughfall deposition (Germany)	This study
	^{15}N tracer	3.4	0.1	EF_{15N}	33 kg N ha^{-1} yr^{-1} throughfall deposition (Germany)	This study

EF_F: Emission factor represented the fraction of N input that is re-emitted: EF_F = flux/N input · 100. EF_{FB}: Emission factor calculated for N fertilisation experiments or N deposition gradients, where N_2O background emissions are considered. Calculated for N fertilisation experiments: EF_{FB} = (flux from fertilised plot – flux from control plot)/N amount in fertiliser · 100. Calculated for N deposition gradients: EF_{FB} = (flux at high N input – flux at background N input/high N input – background N input) · 100. EF_R: Emission factor were derived from regression analysis between N_2O fluxes and N deposition rates. EF_{15N}: Emission factor were derived from ^{15}N tracer experiment; for calculations see this study.

[a] The compared sites were similar in climatic and edaphic conditions.

[b] Lysimeter study with re-established soil profiles from Scots pine forest.

[c] Spruce-fir-beech forest.

[d] Mixed woodland of pine, birch, oak, rowan, and elder.

[e] Emission factor was calculated by the authors of this study.

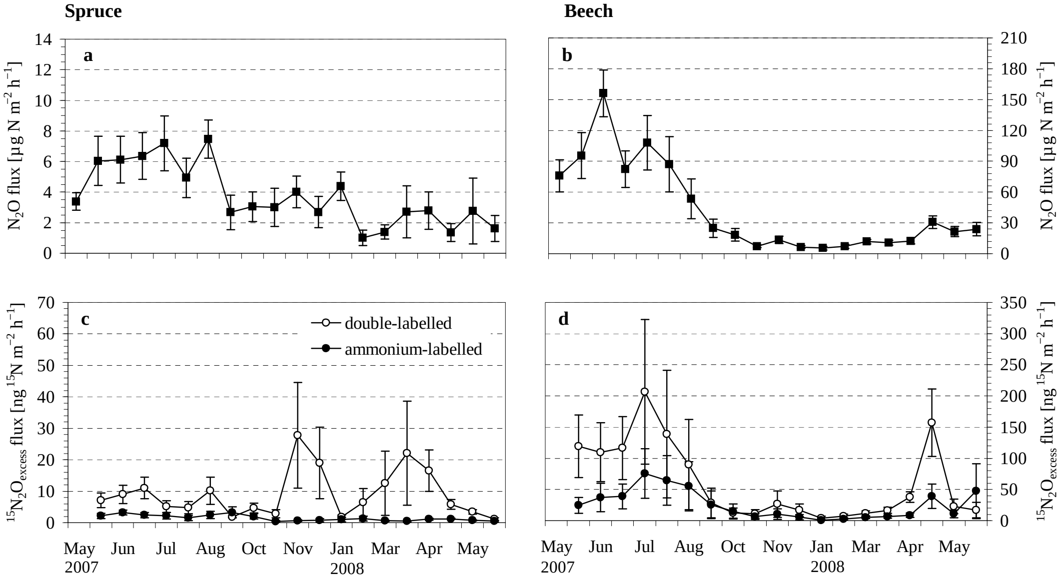

Fig. 1. Mean (±SE, $n = 13$) N_2O fluxes **(a, b)** and mean (±SE, $n = 5$) $^{15}N_2O$ excess fluxes in the NH_4NO_3 (double)-labelled and NH_4^+-labelled treatments **(c, d)** of the spruce (a, c) and beech stands (b, d). Please note the different scales.

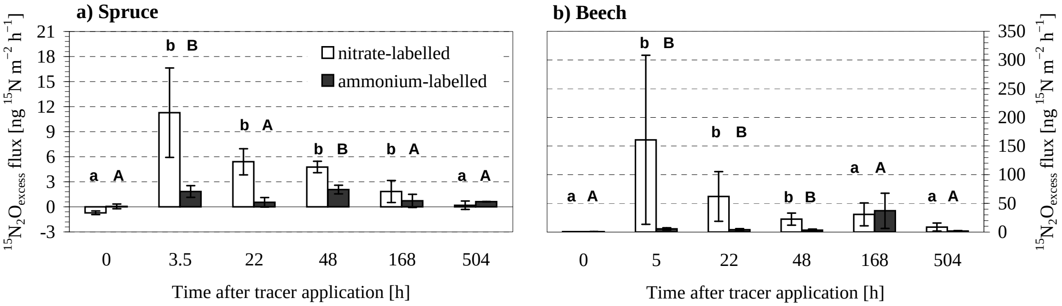

Fig. 2. Mean (±SE, $n = 4$) $^{15}N_2O$ excess fluxes in the spruce **(a)** and beech stands **(b)** during the short-term experiment. Fluxes are given for the NO_3^--labelled and NH_4^+-labelled treatments before (zero hours) and after the tracer application. Means followed by the same letter indicated no significant differences in $^{15}N_2O$ excess fluxes of one labelling treatment (NO_3^-- or NH_4^+-labelled) between zero hours and the time points after the tracer application (mixed effects models with linear contrasts at $P \leq 0.05$). Please note the different scales.

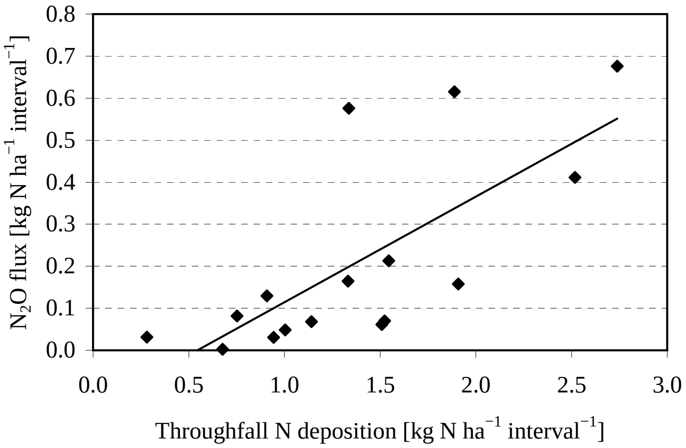

Fig. 3. Relation between N_2O fluxes (y) and total throughfall N deposition (x) in the beech stand from May 2007 to June 2008 ($y = x \cdot 0.25\ (\pm 0.06) - 0.14\ (\pm 0.09)$, $r^2 = 0.53$, $P = 0.001$, $df = 14$).

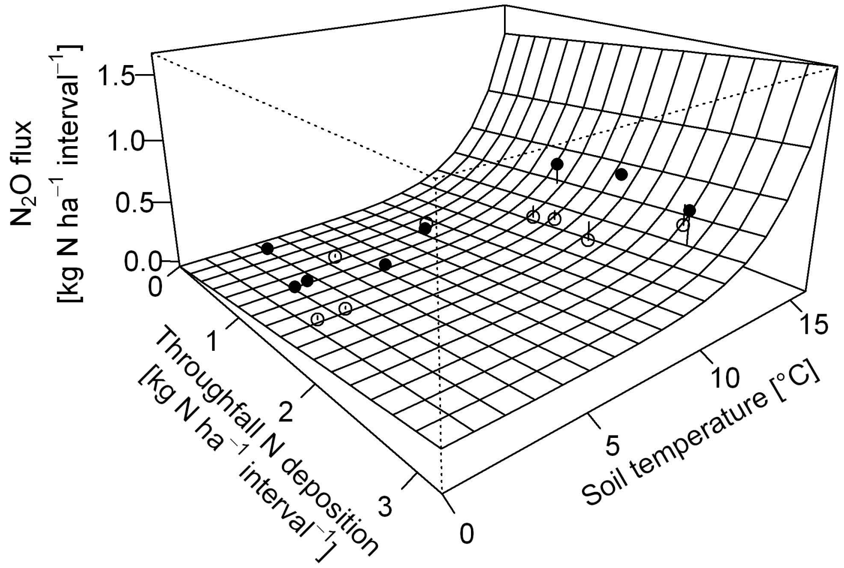

Fig. 4. Relation of N_2O fluxes (y) to throughfall N deposition ($x1$) and soil temperature in 5 cm depth ($x2$) ($y = 0.086\ (\pm 0.063) \cdot x1 + 0.001\ (\pm 0.000) \cdot \exp(0.468\ (\pm 0.242) \cdot x2) - 0.042\ (\pm 0.073)$; $r^2 = 0.77$, $P < 0.0001$, $df = 13$). The measured data are indicated as points, where black points are located above the surface area and white points are located below the surface area. The solid lines indicate the deviations of measured data from the model.

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
◀I
▶I
◀
▶
Back
Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion