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Control of nitrous oxide emissions in European beech, Norway spruce and Scots pine forests

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Abstract. Elevated nitrogen deposition has increased tree growth, the storage of soil organic matter, and nitrate leaching in many European forests, but little is known about the effect of tree species and nitrogen deposition on nitrous oxide emission. Here we report soil N_2O emission from European beech, Scots pine and Norway spruce forests in two study areas of Germany with distinct climate, N deposition and soils. N_2O emissions and throughfall input of nitrate and ammonium were measured biweekly during growing season and monthly during dormant season over a 28 months period. Annual N_2O emission rates ranged between 0.4 and 1.3 kg N ha^{-1} $year^{-1}$ among the stands and were higher in 1998 than in 1999 due to higher precipitation during the growing season of 1998. A 2-way-ANOVA revealed that N_2O fluxes were significantly higher ($p < 0.001$) at Solling than at Unterlüß while tree species had no effect on N_2O emissions. Soil texture and the amount of throughfall explained together 94% of the variance among the stands, indicating that increasing portions of silt and clay may promote the formation of N_2O in wet forest soils. Moreover, cumulative N_2O fluxes were significantly correlated ($r^2 = 0.60$, $p < 0.001$) with cumulative NO_3^- fluxes at 10 cm depth as an indicator of N saturation, however, the slope of the regression curve indicates a rather weak effect of NO_3^- fluxes on N_2O emissions. N input by throughfall was not correlated with N_2O emissions and only 1.6–3.2% of N input was released as N_2O to the atmosphere. Our results suggest that elevated N inputs have little effect on N_2O emissions in beech, spruce and pine forests.

Introduction

Nitrous oxide (N_2O) has a lifetime of about 120 years in the atmosphere and contributes to global warming and to the destruction of ozone in the stratosphere (IPCC 2001). The atmospheric concentration of N_2O has increased from 270 ppb to 314 ppb during the past 250 years and continues to increase with an annual rate of 0.8 ppb. Globally, terrestrial soils are the largest source of N_2O, though the uncertainty of the estimates for the individual biomes is extremely high. Temperate forest soils have been estimated to emit between 0.1 and 2.0 Tg N_2O–N $year^{-1}$ which is equivalent to a range of 0.1–1.7 kg N_2O–N ha^{-1} $year^{-1}$ (IPCC 2001).

N_2O is an intermediate product of aerobic nitrification and anaerobic denitrification and the production rates may be controlled directly or indirectly by a combination of site and climatic parameters such as soil pH, temperature,

water potential, texture and aggregation of soils, gas diffusivity, and substrate availability. The temporal dynamic of N_2O emissions follows generally the seasonal trend in soil temperature, soil moisture and substrate availability (Li et al. 2000). N_2O emissions may be higher in wet-warm years as compared to dry years because maximum N_2O production can be reached at water filled pore space (WFPS) of about 50–70% while an increase or decrease in WFPS reduces the production of N_2O (Davidson et al. 2000). Moreover, intensive dry/wet and frost/thaw cycles can cause large N_2O emissions of soils, indicating that additional factors may control N_2O production during these extreme conditions (Davidson 1992; Papen and Butterbach-Bahl 1999).

In contrast to the temporal dynamic, little is known about the combinatorial effect of site specific parameters on N_2O emissions in forest ecosystems. Hence, there is a lack of information about the control of N_2O emissions on regional scales. The heterogeneity of soils may play an important role in the release of N_2O through the influence of the physical soil structure on the O_2 supply of soil organisms. Bollmann and Conrad (1998) reported increasing N_2O production by denitrification with decreasing O_2 partial pressure whereas maximum N_2O production by nitrification was reached at 0.1–0.5% O_2. Because the gas transport within the pore space is associated with texture and aggregation of soils (Ball et al. 1997), higher N_2O emissions may be expected in fine textured soils as compared with coarse textured soils at high water contents (Bollmann and Conrad 1998).

Tree species are considered to affect N_2O fluxes of forest ecosystems through modification of litter quality, the buildup of organic horizons, throughfall input, and chemical, biological and physical soil properties. Only few studies were performed to assess the effect of tree species on N_2O emissions at same soil and climate. Annual N_2O emission rate of a beech stand was higher than the emission rate of an adjacent spruce stand at Solling (Brumme et al. 1999). According to Butterbach-Bahl et al. (2002a), mean annual N_2O emission rate of a beech stand was three times as high as in the spruce stand at the Höglwald, Germany. One reason for elevated N_2O emissions in beech forests is the laminar structure of beech leaves which allows the creation of anaerobic habitats in the water film between the leaves (Brumme et al. 1999). Moreover, thick and moist organic horizons from beech litter promote anaerobic conditions in the mineral soil by reduction of O_2 diffusion into the mineral soil. N_2O emissions of beech forests with mull-type humus forms are low (Brumme et al. 1999), indicating that the effect of tree species cannot be generalized.

Another parameter that affects the production of N_2O in soils is the turnover of ammonium (NH_4^+) and nitrate (NO_3^-) by nitrification and denitrification, respectively (Firestone and Davidson 1989). Most temperate forest soils have no or low NH_4^+ concentrations in soil solution due to efficient nitrification and/or immobilization of NH_4^+ by plants and microorganisms. By contrast, NO_3^- concentrations are often very high in soil solution and, thus, leaching of NO_3^- to the aquifer has become an environmental problem in some regions with high N deposition during past years.

Annual emissions of reactive N have been reduced from 696 to 500 Gg NH_3–N $year^{-1}$ and from 1020 to 485 Gg NO_x–N during the period from 1987 to 2001 in Germany (UBA 2003). Despite these reductions, the input of inorganic N by throughfall remained on a high level and long-term study sites showed only a weak tendency towards lower N inputs since the late 1980s (Wright et al. 2001; Matzner et al. 2004a). Borken and Matzner (2004) reported inorganic N inputs by throughfall of 6.5–35.4 kg N ha^{-1} $year^{-1}$ in 57 forest sites in Germany. Elevated inputs of NH_4^+ and NO_3^- could have enhanced the production of N_2O by both nitrification and denitrification in most forest soils. The conceptual model of Aber et al. (1998) predicts an exponentially increase in N_2O emissions with gradual N saturation of forest ecosystems. This conceptual model was supported by the results of Butterbach-Bahl et al. (2002b) showing that N_2O emissions of pine forest soils increased along a gradient from low to high N deposition. Hence, N_2O emissions could potentially increase with increasing N deposition as in many Central European forests nitrogen input exceeds the need of trees for their growth.

Our objectives were (1) to assess the effect of tree species on N_2O fluxes and (2) to investigate the effect of inorganic N input and N fluxes in seepage on N_2O fluxes of European beech, Norway spruce and Scots pine at Solling and Unterlüß two study areas with contrasting climate, N deposition rates and soil properties. We used inorganic N fluxes in seepage as an indicator of N saturation rather than as direct driver of nitrification and denitrification. European beech, Norway spruce and Scots pine stands were chosen because these tree species represent the dominant tree species in German forests.

Materials and methods

Study sites

The experiment was carried out in European beech (*Fagus sylvatica*), Norway spruce (*Picea abies*) and Scots pine stands (*Pinus sylvestris*) at Solling and at Unterlüß in Lower Saxony, Germany (Table 1). All sites are characterized by soil acidification with a base saturation of less than 10% in the soil profile from 5 cm down to 100 cm depth. Aluminum is the dominating exchangeable cation of the soil matrix. In 1988, soils at Solling were limed to mitigate soil pH in the O horizon and increase the base saturation of the upper mineral soil at all sites.

The 150-year-old beech stand and the 115-year-old spruce stand at the Solling plateau above 500 m elevation have a mean annual air temperature of 7.2 °C and an annual precipitation of 1038 mm, evenly distributed throughout the year. The 103-year pine stand at Solling is located at an elevation of 270 m and has a mean annual temperature of 7.5 °C and an annual precipitation of 900 mm. The soils of these sites are developed on 30–80 cm thick solifluction deposits, overlaying weathered Triassic Sandstone. Texture of the soils at 0–20 cm depth was dominated by the silt fraction (46–58%) with varying clay

Table 1. Some characteristics of the study sites.

Site	Tree species	Geographical location	Elevation (m)	MAT (°C)	MAP (mm)	Stand age (year)	Soil texture		
							% clay	% silt	% sand
Solling	Beech	51°46′ N, 9°35′ E	504	7.2	1038	150	19	54	27
	Spruce	51°46′ N, 9°34′ E	508	7.2	1038	115	28	58	14
	Pine	51°34′ N, 9°40′ E	270	7.5	900	103	15	46	39
Unterlüß	Beech	52°50′ N, 10°18′ E	117	8.4	837	131	8	16	77
	Spruce	52°50′ N, 10°17′ E	115	8.4	837	90	3	23	74
	Pine	52°50′ N, 10°16′ E	110	8.4	837	54	4	16	81

and sand contents. The soils were classified as well-drained to poorly-drained Dystric Cambisols according to FAO soil taxonomy (FAO 1998).

The 131-year-old beech, 90-year-old spruce and 54-year-old pine stands at Unterlüß, were close to each other at an elevation of 110 m above sea level (Table 1). The long-term average of mean annual air temperature is 8.4 °C and the mean annual precipitation is 837 mm. The soils are developed from fluvio-glacial sand and gravel deposited over a terminal moraine during the Warthe-stadium of the Saale/Riss ice age. The soils contain about 74–81% sand, 16–23% silt and 3–8% clay at 0–20 cm depth. The soils have been classified as well-drained Cambisols (FAO 1998) with moder-type O horizons. The beech stand has no ground vegetation whereas the pine stand has a dense cover of grass and *Vaccinium* species.

N_2O measurements

In each stand five cylindrical PVC columns, 30 cm in diameter and 25 cm tall, were inserted into the O-horizon down to 5 cm depth. The columns were left in the soil for the duration of the experiment. N_2O fluxes were measured by placing a PVC lid over each column and taking three gas samples from the chamber headspace (11–15 l) using a sampling device and evacuated glass bottles (100 ml) after 0, 20 and 40 min of closure. N_2O concentrations were analyzed in the laboratory on an automated gas chromatograph (Carlo Erba Instruments, GC 6000, Vegas Series 2), equipped with an electron capture detector (ECD) (Loftfield et al. 1997). Four certified N_2O standards (332, 756, 1110, and 2535 ppbv N_2O in N_2, Deuste Steininger, Mühlhausen, Germany) were used for calibration. Repeated measurements of certified N_2O standards resulted in a precision of 2% for our GC system. N_2O fluxes were calculated, using chamber air temperature, barometric air pressure and the slope of the temporal change in N_2O concentration within the chamber headspace. The minimum detectable flux was 2 μg N_2O–N m^{-2} h^{-1}. Gas samples were taken between 10.00 and 14.00 hours on each sampling date, i.e. monthly from September 1997 to April 1998 and biweekly from May 1998 to December 1999.

Ammonium, nitrate and water fluxes

In the spring of 1997, four suction lysimeters equipped with ceramic P-80 cups were installed at 10 cm mineral soil depth in each of three subplots at all six forest stands. Except for dry periods, a vacuum pressure between −0.7 and −0.3 bar, generated by a vacuum pump, insured continuous sampling of seepage in 1 or 2 l glass jars. Subsamples (100 ml) of seepage water were discontinuously collected from summer 1997 throughout March 2000 and were stored at 4 °C until chemical analysis. Throughfall water was collected biweekly to monthly using 10 rain gauges installed at a height of 1 m above the forest floor. During the winter seasons from November to March, five 10 l buckets were used to collect snow and rain. All water samples were filtered using 0.45 μm membrane filters (Schleicher and Schuell, Dassel). Ammonium and nitrate were colorimetrically measured in a continuous flow system (Skalar Analytic GmbH, Erkelenz) with a detection limit of 0.15 mg N l^{-1} for NH_4^+ and NO_3^- (König and Fortmann 1996).

Daily volumetric water contents and seepage fluxes at 5 and 10 cm depth were obtained from a soil water balance model that was previously described by Borken et al. (2002a). N fluxes in seepage were calculated by multiplying seepage fluxes with mean NH_4^+ and NO_3^- concentration in seepage.

Modelled volumetric soil water contents θ (cm^3 cm^{-3}) at 5–10 cm mineral soil depth were converted into water filled pore space (WFPS) as follows:

$$\mathrm{WFPS} = \theta/(1 - \mathrm{BD}/\mathrm{PD}),$$

where BD is the bulk density (g cm^{-3}) and PD is the particle density (g cm^{-3}) of the mineral soil which was estimated as 2.65 g cm^{-3}.

Data analysis

A 2-way ANOVA was performed to test the significance of tree species (beech, spruce, pine) and study area (Solling and Unterlüß,) on cumulative N_2O fluxes for the entire sampling period using five replications from each stand. Linear regressions were performed to evaluate the relationships between N_2O fluxes, the amount of throughfall, N input by throughfall and N seepage fluxes at 10 cm depth. Cumulative N_2O flux rates were calculated assuming linear changes between two sampling occasions.

Results

N_2O emissions

N_2O emissions were mostly below 20 μg N m^{-2} h^{-1} in all stands during the entire experimental period (Figures 1a–6a). Only at few occasions the soils at

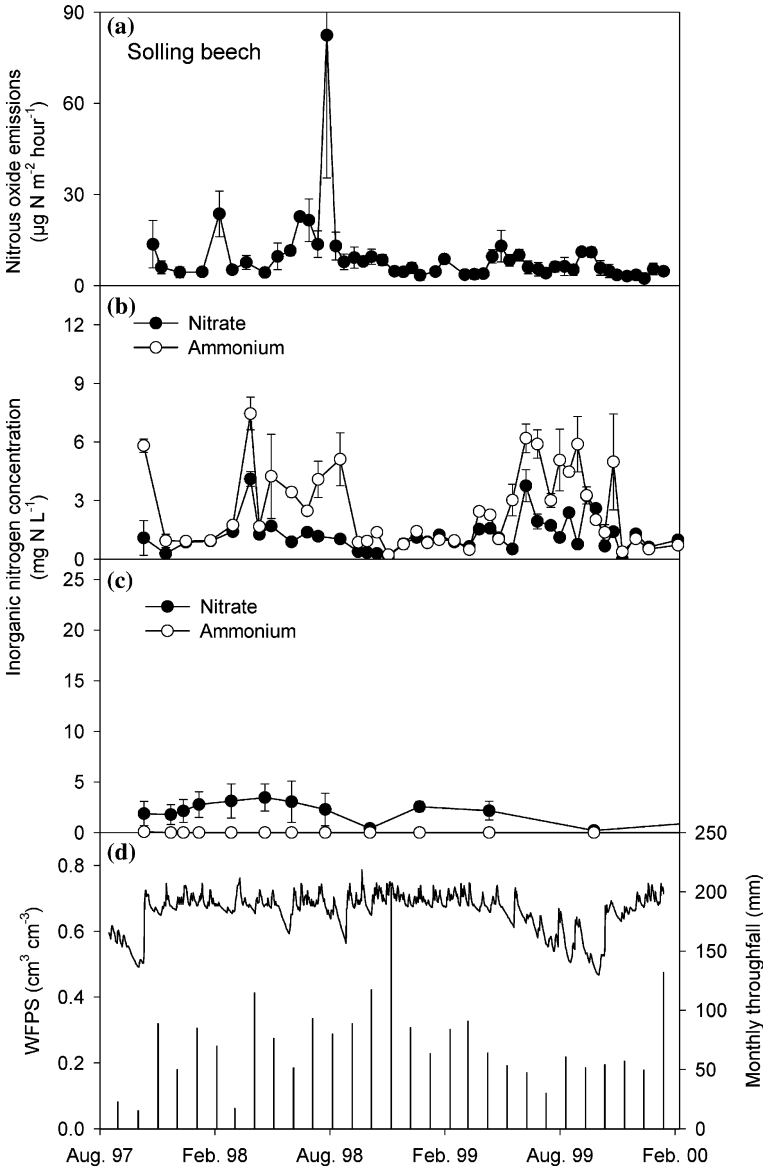

Figure 1. Nitrous oxide emission rates (a), ammonium and nitrate concentration of throughfall (b) and seepage at 10 cm depth (c), daily WFPS at 10 cm depth and monthly throughfall (d) in the beech forest at Solling. Error bars represent the standard error of the mean.

Solling emitted up to 80 μg N m^{-2} h^{-1} in 1998, which was a rather wet year as compared to 1997 and 1999. Annual N_2O emissions ranged between 0.6 and 1.3 kg N ha^{-1} $year^{-1}$ in 1998 and between 0.4 and 0.6 kg N ha^{-1} $year^{-1}$ in 1999. N_2O emissions were positively correlated ($y = 0.001x - 0.26$, R^2

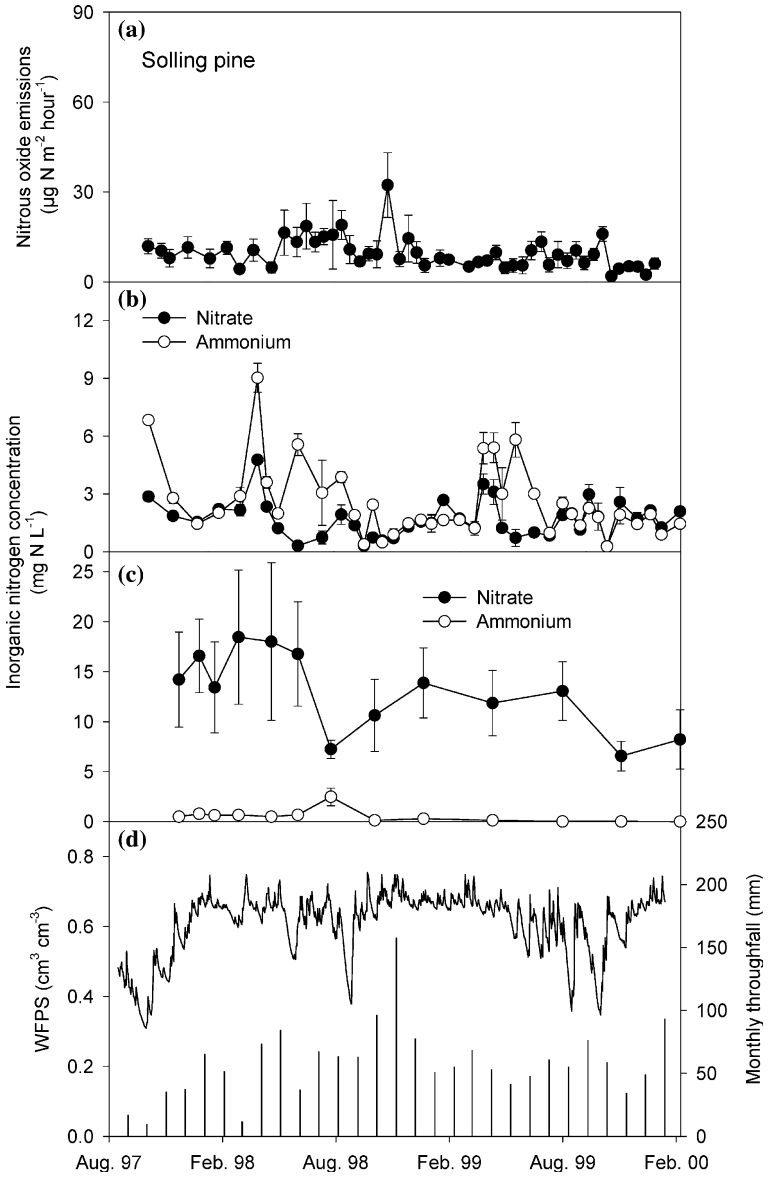

Figure 2. Nitrous oxide emission rates (a), ammonium and nitrate concentration of throughfall (b) and seepage at 10 cm depth (c), daily WFPS at 10 cm depth and monthly throughfall (d) in the pine forest at Solling. Error bars represent the standard error of the mean.

adj. = 0.64, $p = 0.001$) with annual amount of throughfall (Figure 7). The results indicate that the amount of throughfall during the growing seasons affected annual N_2O emission rates at Solling, but to much lesser extent at Unterlüß. Soils at Solling were always wetter during growing seasons than at

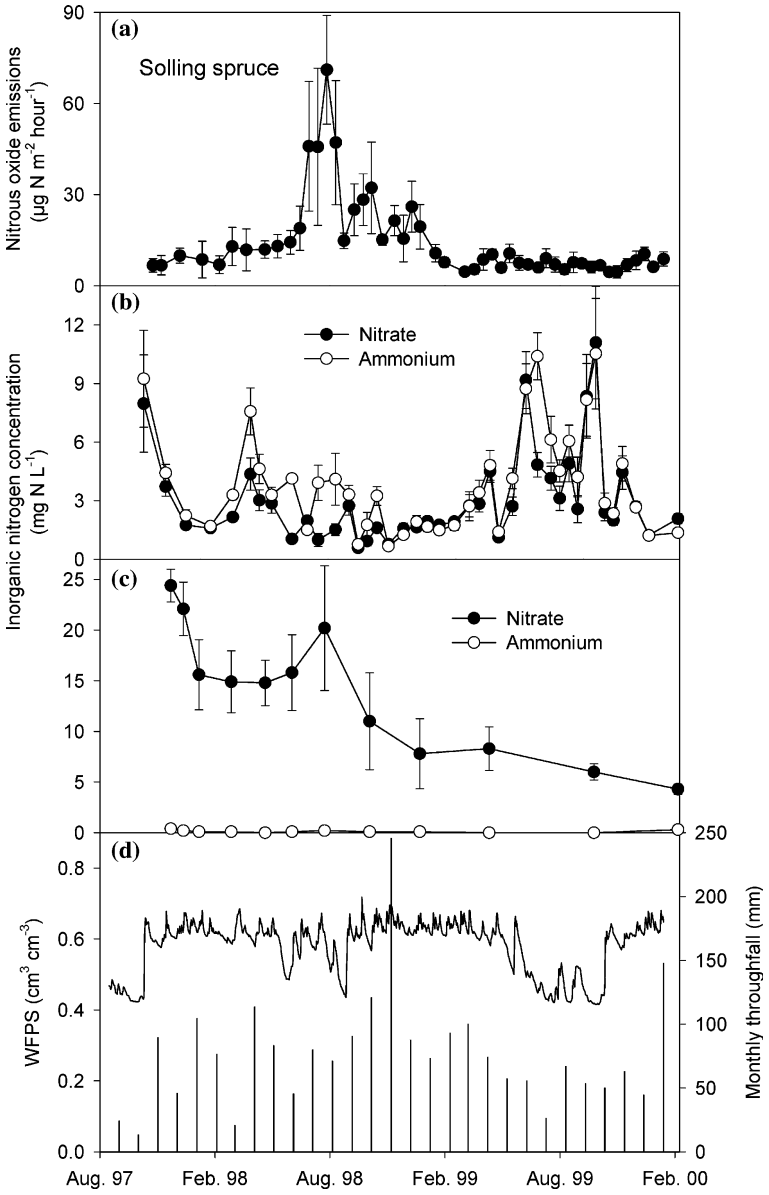

Figure 3. Nitrous oxide emission rates (a), ammonium and nitrate concentration of throughfall (b) and seepage at 10 cm depth (c), daily WFPS at 10 cm depth and monthly throughfall (d) in the spruce forest at Solling. Error bars represent the standard error of the mean.

Unterlüß, due to higher amounts of throughfall (Figures 1d–6d) and higher water holding capacities of the silt soil texture (Table 1). Mean WFPS decreased in the order Solling > Unterlüß, and beech > pine > spruce.

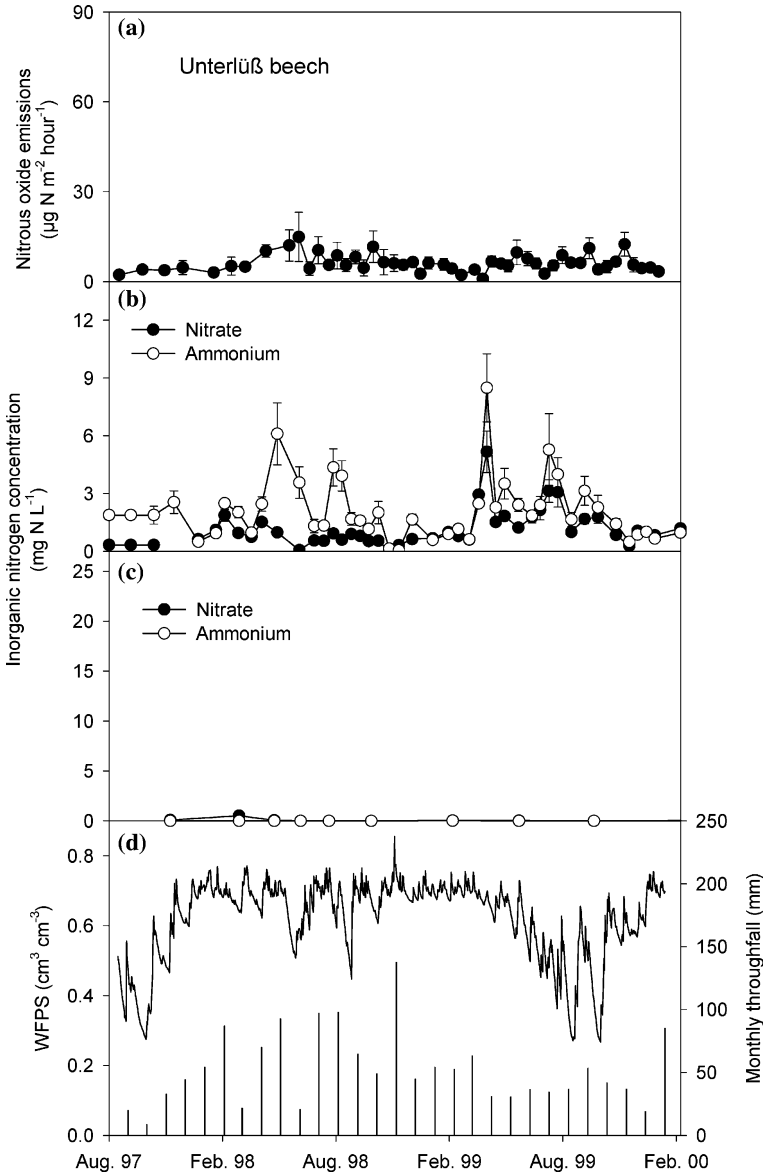

Figure 4. Nitrous oxide emission rates (a), ammonium and nitrate concentration of throughfall (b) and seepage at 10 cm depth (c), daily WFPS at 10 cm depth and monthly throughfall (d) in the beech forest at Unterlüß. Error bars represent the standard error of the mean.

A two-way ANOVA revealed a strong effect of the study area ($p < 0.001$) on cumulative N_2O emission rates. A reason for the difference in N_2O emissions between Solling and Unterlüß is the difference in soil texture which explained 87% ($p = 0.004$) of the variation among the six stands (Figure 8). A multiple

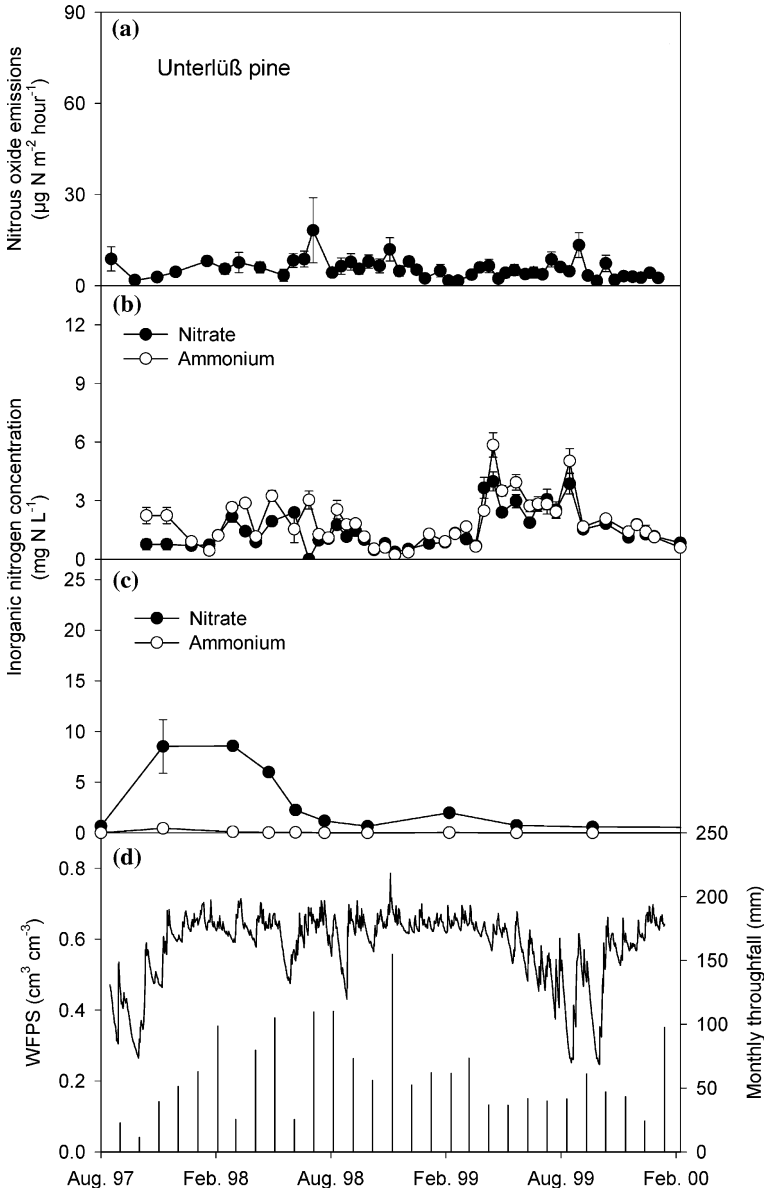

Figure 5. Nitrous oxide emission rates (a), ammonium and nitrate concentration of throughfall (b) and seepage at 10 cm depth (c), daily WFPS at 10 cm depth and monthly throughfall (d) in the pine forest at Unterlüß. Error bars represent the standard error of the mean.

regression with soil texture and the amount of throughfall as independent variables explained 94% ($p = 0.007$) of the variation in N_2O emissions. Overall, the stands at Solling emitted between 1.7 and 2.1 kg N ha^{-1} and the

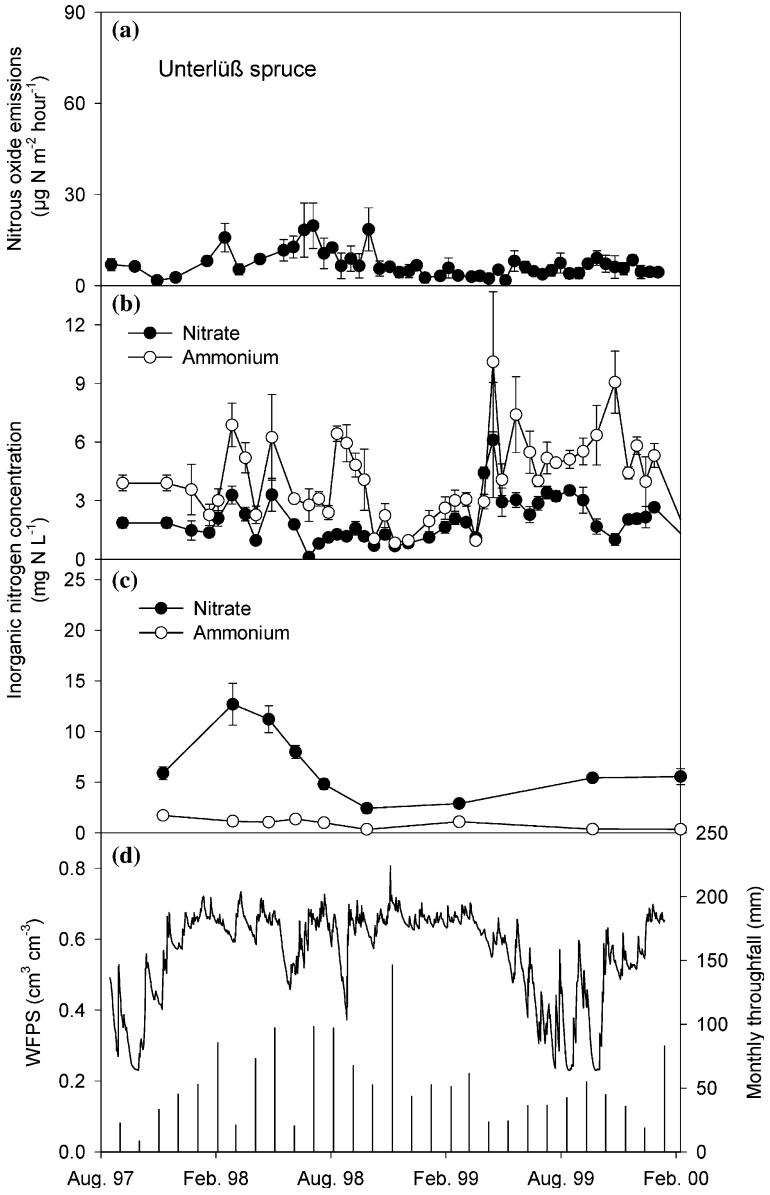

Figure 6. Nitrous oxide emission rates (a), ammonium and nitrate concentration of throughfall (b) and seepage at 10 cm depth (c), daily WFPS at 10 cm depth and monthly throughfall (d) in the spruce forest at Unterlüß. Error bars represent the standard error of the mean.

stands at Unterlüß, emitted between 1.1 and 1.4 kg N ha^{-1} during the entire experimental period (Table 2). Tree species had no effect on N_2O emissions and the interaction between study area and tree species was not significant.

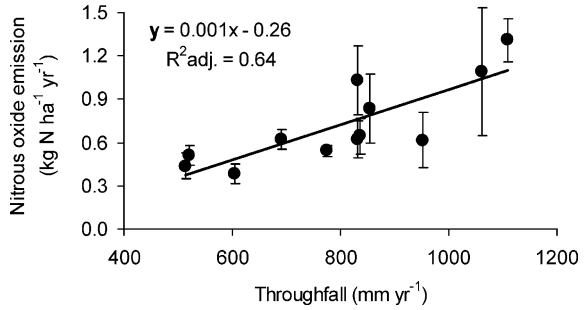

Figure 7. Relationship between mean annual nitrous oxide fluxes and annual amounts of throughfall in the beech, spruce and pine stands at Solling and Unterlüß in 1998 and 1999. Error bars represent the standard deviation of five replications.

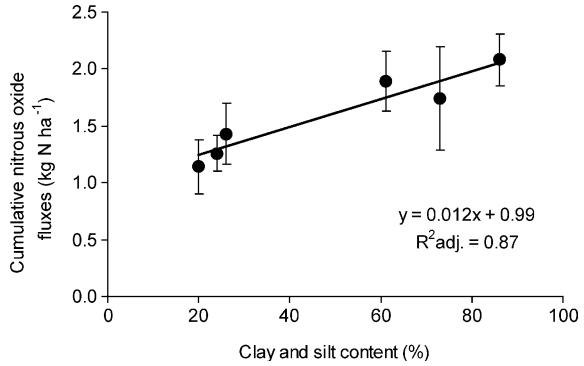

Figure 8. Relationship between mean cumulative nitrous oxide fluxes and soil texture (sum of clay and silt content) of the top soil. Error bars represent the standard deviation of five replications.

Table 2. Nitrous oxide fluxes of European beech, Norway spruce and Scots pine forests at Solling and Unterlüß during different time periods. Values in parentheses represent the standard error of the mean.

Site	Forest type	1997[1]	1998	1999	Sum
		(kg N ha^{-1})	(kg N ha^{-1} $year^{-1}$)	(kg N ha^{-1} $year^{-1}$)	(kg N ha^{-1})
Solling	Beech	0.1 (0.0)	1.1 (0.2)	0.5 (0.0)	1.7 (0.2)
	Pine	0.3 (0.0)	1.0 (0.1)	0.6 (0.1)	1.9 (0.1)
	Spruce	0.2 (0.0)	1.3 (0.1)	0.6 (0.1)	2.1 (0.1)
Unterlüß,	Beech	0.2 (0.0)	0.6 (0.1)	0.5 (0.1)	1.3 (0.1)
	Pine	0.1 (0.0)	0.6 (0.1)	0.4 (0.1)	1.1 (0.1)
	Spruce	0.2 (0.0)	0.8 (0.1)	0.4 (0.1)	1.4 (0.1)

[1] In 1997, nitrous oxide fluxes were measured from August to December at Unterlüß, (136 days) and from October to December (90 days) at Solling.

NH_4^+ and NO_3^- concentration in throughfall followed a seasonal pattern with maximum concentrations during growing season and lowest concentrations during dormant season (Figures 1b–6b). Cumulative NH_4^+ fluxes ranged between 29 and 67 kg N ha^{-1} and were about 8–31 kg N ha^{-1} higher than NO_3^- fluxes in the six forest stands (Table 3). The spruce soils received 43–60 kg N ha^{-1} more inorganic nitrogen than the respective beech and pine soils at Solling and Unterlüß. Comparing same tree species, inorganic nitrogen input by throughfall was higher at Solling than at Unterlüß.

In contrast to throughfall, NH_4^+ concentration in seepage was very low or below the detection limit (Figures 1c–6c). Cumulative NH_4^+ flux rates of 0.1–8.0 kg N ha^{-1} indicate that NH_4^+ was almost completely removed by biotic and/or abiotic processes throughout the year (Table 3). Nitrate concentration in seepage ranged between 0 and 24 mg N l^{-1} and were often higher than in throughfall (Figures 1c–6c). With one exception, cumulative NO_3^- fluxes in seepage at 10 cm soil depth were 1.6–5.4-times higher than in throughfall (Table 3). Only the beech stand at Unterlüß, had very low NO_3^- fluxes in seepage. The study area as well as the tree species had significant effects ($p = 0.02$) on NO_3^- fluxes and followed the order Solling > Unterlüß, and spruce > pine > beech.

Cumulative N_2O fluxes were positively correlated with cumulative NO_3^- fluxes in seepage at 10 cm depth ($y = 0.005x + 1.25$, R^2adj. = 0.61, $p = 0.04$) (Figure 9). Despite the significant correlation, the N_2O emissions are small compared to NO_3^- fluxes in seepage. Ammonium, nitrate and total nitrogen input by throughfall were not correlated with N_2O fluxes.

Discussion

Annual N_2O emission rates of 0.4–1.3 kg ha^{-1} $year^{-1}$ in the beech, pine and spruce stands at Solling and Unterlüß were in the range of N_2O emissions (0.1–1.7 kg N ha^{-1} $year^{-1}$) estimated globally for temperate forest soils (IPCC 2001). The emissions were significantly higher at Solling than at Unterlüß, suggesting that soil texture and the amount of throughfall together had a strong effect on the release of N_2O since both parameters were significantly correlated with N_2O emissions. In the wetter soils at Solling, the average O_2 supply within the soil pore space was apparently lower and the formation of N_2O higher as compared with the soils at Unterlüß. Assuming that the moderate variation of soil texture within the study sites (Table 1) did not affect N_2O emissions, it can be deduced from the 2-way ANOVA that tree species had no significant effect on N_2O emissions at Solling and Unterlüß.

In contrast to our results, Brumme et al. (1999) and Butterbach-Bahl et al. (2002a) found higher N_2O emission rates in beech forests than in adjacent spruce forests. In beech forests with low soil pH and low bioturbation, beech

Table 3. Cumulative ammonium and nitrate fluxes of throughfall and seepage at 10 cm depth of European beech, Norway spruce and Scots pine forests at Solling (807 days) and at Unterlüß (856 days) and the ratios of cumulative N_2O fluxes and N input by throughfall. Values in parentheses represent the standard error of the mean.

Site	Tree species	Throughfall		Seepage		N_2O flux/N input (%)
		(kg NH_4–N ha^{-1})	(kg NO_3–N ha^{-1})	(kg NH_4–N ha^{-1})	(kg NO_3–N ha^{-1})	
Solling	Beech	43.9 (0.8)	19.4 (0.5)	0.1 (0.1)	30.1 (14.1)	2.7
	Pine	36.1 (0.5)	22.7 (1.0)	3.0 (0.5)	120.9 (31.9)	3.2
Spurce	67.3 (8.1)	51.2 (7.7)	1.1 (0.3)	149.6 (35.5)	1.8	
Unterlüß	Beech	28.7 (2.2)	13.2 (0.8)	0.1 (0.0)	0.7 (0.6)	3.0
	Pine	29.1 (1.6)	21.6 (0.9)	0.9 (0.3)	39.8 (4.6)	2.3
	Spruce	58.2 (5.1)	27.2 (1.9)	8.0 (1.4)	45.2 (1.8)	1.7

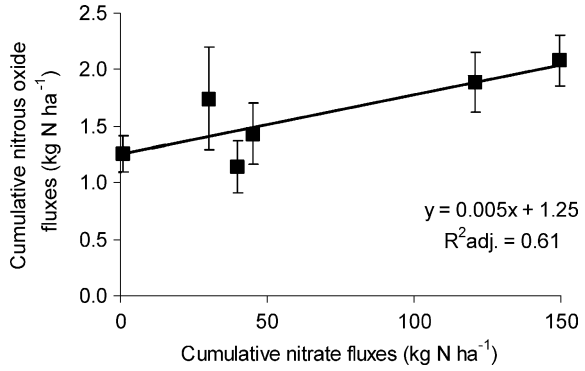

Figure 9. Relationship between mean cumulative nitrous oxide fluxes and mean cumulative nitrate fluxes in seepage at 10 cm mineral soil depth. Error bars represent the standard deviation of five replications.

litter may build up thick organic horizons with a laminar structure, where low gas diffusivity can promote anaerobic microsites at high water contents. Both beech stands at Solling and Unterlüß, have thick organic horizons, but the N_2O emission rates were much lower than reported N_2O emissions rates for similar beech stands (Brumme et al. 1999; Butterbach-Bahl et al. 2002a). The low N_2O emissions of the beech stand at Solling may be explained by the application of lime (28 Mg ha^{-1} in 1988) and the moderate increase in soil pH. An increase in soil pH affects the denitrification rate and the N_2O/N_2 ratio towards N_2 (Granli and Bøckman 1994). In agreement with previous results, heavy lime doses may reduce N_2O production of acidic forest soils (Borken and Brumme 1997). Furthermore, beech forests with soil pH within the silicate or carbonate puffer range had low N_2O emission rates (Brumme et al. 1999), indicating that only few beech forests have the potential to emit large amounts of N_2O to the atmosphere.

The low N_2O emissions of the beech stand at Unterlüß may be explained by low NO_3^- concentration in seepage and rather low WFPS during growing seasons as compared to Solling. Our results show that precipitation did not only affect site variation of N_2O emissions but also interannual variation of N_2O emissions. N_2O emission rates of all stands were higher in 1998 at high amounts of throughfall (range of 832–1108 mm) as compared to 1999 with low amounts of throughfall (range of 514–831 mm). Annual N_2O emission rates were positively correlated with annual amounts of throughfall (Figure 7), suggesting that future changes in precipitation and evapotranspiration, particularly during growing season, can affect N_2O emission rates. Butterbach-Bahl et al. (2002a) reported an annual variation of 1.6–6.6 kg N_2O–N ha^{-1} $year^{-1}$ for a beech forest and 0.8–3.1 kg N_2O–N ha^{-1} $year^{-1}$ for an adjacent spruce forest at the Höglwald, Germany, during a 3-years period. High emission rates during an intensive frost/thaw period in the winter of 1996 contributed largely to the interannual variation at the Höglwald. However, soil

frost was not observed at our sites during the entire experimental period (Borken et al. 2002a).

With one exception, our forest stands may be defined as N saturated ecosystems as considerably fluxes of NO_3^- were measured at 10 cm depth (Table 3) and below the rooting zone (Borken et al. 2004). The beech forest at Unterlüß, was the only stand where both NH_4^+ and NO_3^- concentrations and fluxes in seepage were low or not measurable. Compared with the other forests, this stand received the lowest inorganic N input by throughfall (17.9 kg N ha^{-1} yr^{-1}) which is above the range of what beech forests need to maintain their growth. Matzner et al. (2004b) calculated a N uptake by above- and belowground biomass increment of 14.0 kg N $year^{-1}$ for a beech stand with similar N input by throughfall than the beech stand at Unterlüß. Throughfall N input of the other stands exceeded the need of N for growth and, thus, elevated NH_4^+ and NO_3^- availability may have promoted the production of N_2O. In fact, cumulative N_2O fluxes were positively correlated with cumulative NO_3^- fluxes at 10 cm depth as an indicator of N saturation (Figure 9). Because cumulative N_2O fluxes were relatively low (1.1–2.1 kg N ha^{-1}) compared with cumulative NO_3^- fluxes (0.7–149.6 kg N ha^{-1}) we conclude that the site variation in NO_3^- fluxes had little effect on N_2O emissions. Hence, it is not surprising that N input by throughfall did not explain the variation in N_2O emissions among the stands. The ratio of N_2O emissions and N input by throughfall (Table 3) shows that 1.7–3.2% of the N input was released as N_2O in our stands. Tree species had obviously an effect on this ratio as both spruce stands emitted only 1.7 and 1.8% of the N input to the atmosphere whereas the ratios of the beech and pine stands ranged between 2.3 and 3.2%.

Butterbach-Bahl et al. (2002b) reported a strong impact of N deposition on N_2O fluxes in Scots pine forests of the northeastern German lowlands, suggesting a much stronger increase in N_2O emissions with increasing inorganic N input as indicated by our results. Their relationship, however, is based on some short-term campaigns < 1 month, which could have resulted in an overestimation of the N deposition effect on N_2O emissions, considering annual time scales. On a regional scale, inorganic N input is positively correlated with the amount of throughfall (Borken and Matzner 2004), suggesting that the level of N_2O emissions cannot only be attributed to N input, but also to the amount of throughfall.

N-fertilization studies might provide insight into the response of N_2O emissions to elevated N deposition although additional effects such as changes in the osmotic potential must be considered when soils were treated with large amounts of fertilizer. N_2O emissions of an adjacent beech stand at Solling increased by about 40% following an annually repeated (NH_4SO_4) application of 140 kg N ha^{-1} (Brumme and Beese 1992). Only 1.6% of the added nitrogen was released as N_2O during the study which is in agreement with a similar study at the Harvard Forest, USA, where 0.2% of the fertilizer N was emitted as N_2O (Bowden et al. 1991). Even after more than 12 years of NH_4NO_3 fertilization (50 and 150 kg N $year^{-1}$) N_2O emissions at the Harvard Forest

were generally < 10 μg N m^{-2} h^{-1} and only at few occasions significantly higher than the emissions in the control plots (Venterea et al. 2003). A similar result was reported from a fertilization study in a spruce forest in Sweden, where 35 kg N ha^{-1} $year^{-1}$ were gradually applied to soil by chemical manipulation of throughfall water (Klemedtsson et al. 1997). Although the background N input by throughfall was rather low (12 kg N ha^{-1} $year^{-1}$) as compared to our sites, only 0.2% of the added N was released as N_2O in the Swedish spruce forest. As mentioned above, the ratios between inorganic N input and N_2O emissions (Table 3) agree well with the N fertilization studies.

Conclusion

Our results suggest that elevated N deposition has little effect on N_2O emissions and that other factors such as soil texture and the amount of precipitation may have a stronger effect on site and interannual variation of N_2O emissions. Hence, a reduction of N deposition would not reduce the N_2O emissions of most German forest soils. A previous study revealed that a 10-year reduction of N input in throughfall from about 35 to 11 kg N ha^{-1} $year^{-1}$ did not affect N_2O emissions of a spruce soil at Solling (Borken et al. 2002b). More studies are needed to improve our understanding and the prediction of the fate of inorganic N in soils because the potential of forests to store surplus nitrogen is limited.

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