Spatial variability of nitrous oxide emissions in an unmanaged old-growth beech forest

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Abstract

Nitrous oxide (N₂O) is a high-impact greenhouse gas. Due to the scarcity of unmanaged forests in Central Europe, its long-term natural background emission level is not entirely clear. We measured soil N₂O emissions in an unmanaged, old-growth beech forest in the Hainich National Park, Germany, at 15 plots over a 1-year period. The average annual measured N₂O flux rate was (0.49 \pm 0.44) kg N ha⁻¹ y⁻¹. The N₂O emissions showed background-emission patterns with two N₂O peaks. A correlation analysis shows that the distance between plots (up to 380 m) does not control flux correlations. Comparison of measured data with annual N₂O flux rates obtained from a standard model (Forest-DNDC) without site-specific recalibration reveals that the model overestimates the actual measured N₂O flux rates mainly in spring. Temporal variability of measured N₂O flux was better depicted by the model at plots with high soil organic C (SOC) content. Modeled N₂O flux rates were increased during freezing only when SOC was > 0.06 kg C kg⁻¹. The results indicate that the natural background of N₂O emissions may be lower than assumed by most approaches.

Key words: old-growth forest / nitrous oxide / model / spatial variability / background emissions

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1 Introduction

The atmospheric abundance of the greenhouse-gas nitrous oxide (N₂O) has been rising since industrialization and intensification of agriculture and anthropogenic emissions need to be reduced to counteract global warming (Denman et al., 2007). Consequently, anthropogenic emissions need to be separated from natural background emissions for inventories and better scientific understanding. This is not a trivial task. The standard background emission rate of 1.0 kg N₂O-N ha⁻¹ y⁻¹ by Bouwman (1996) is only valid for agricultural soils which are in any case highly influenced by man. Therefore, fluxes from forest soils which are less influenced by man should better reflect natural background emissions. Using a third version of PnET-N-DNDC, Kesik et al. (2005) proposed an N2O-emission factor for European forests of 0.55 to 0.62 kg N ha⁻¹ y⁻¹ and for German forests the mean value of 0.72 kg N ha⁻¹ y⁻¹. Schulte-Bisping et al. (2003) estimated that the average forest-soil N₂O-emission rate in Germany is 0.32 kg N ha-1 y-1. These estimations are below the background-emission value proposed by Bouwman (1996) for agricultural soils. Most unfertilized agricultural sites are not sites that were never fertilized and like forest site, they are being fertilized by atmospheric depositions. Therefore, natural emissions rates without any anthropogenic impact cannot be measured in Central Europe. To determine realistic natural background-emission rates anyhow joint measurement and modeling approaches for little-fertilized ecosystems are needed. Nitrogen fertilization is one of the main drivers for elevated N_2O emissions and forests to react to increased atmospheric N depositions (Eickenscheidt et al., 2011).

Anthropogenic influence is not restricted to N fertilization (Ruser et al., 1998; Flessa et al., 2002a; Teepe et al., 2004) and plowing (Mosier et al., 1996), and it is unclear after which period of withholding fertilization emission rates return back to their natural background level, if at all. Therefore, heavily biased estimates for natural background emission rates are likely derived from "former" agricultural land due to longer lasting anthropogenic influences. Even soil compaction due to the use of heavy machinery will highly likely have a long-term influence as soil structure controls soil aeration which is a main driver of N_2O emission.

Actual natural background fluxes may only be determined from undisturbed, unmanaged ecosystems that exhibit vegetation close to the potential natural one. However, such ecosystems have become very rare across the globe (Groombridge and Jenkins, 2000). For the greater part of Central Europe, the potential natural vegetation is beech (Fagus sylvatica L.) forest (Ellenberg and Leuschner, 2010). Hence, natural background emission rates representative for Central



Europe should be recorded in pristine beech forests preferably at contrasting sites, e.g., at different soils and under different climate. However, there are hardly any unmanaged beech forests and there are certainly no pristine beech forests left in Central Europe. Nevertheless, in unmanaged old forest ecosystems anthropogenic influences may be assumed to be low and the N status is not elevated besides the unavoidable atmospheric deposition.

Ecosystems in the core zone of the Hainich National Park (NP) are among the closest to natural that are available in all of Central Europe. Thus, they provide the opportunity to determine the closest estimate to the potential natural background emission in Central Europe.

Furthermore, if N₂O release can be redrawn by a biogeochemical model without local recalibration, the model can be used for determining to a certain degree "near-natural N₂O fluxes" for similar ecoregions in Central Europe. By using a publicly accessible model without site-specific recalibration, this assessment would be applicable to these areas. In any case, the outcome of a measurement and modeling approach of a "long-term" unmanaged beech forest will add to the understanding of N₂O fluxes from natural ecosystems. A wellknown phenomenon associated with gas-flux measurement from soils is the high spatial and temporal variability (Folorunso and Rolston, 1984; Huang et al., 2011). An issue that needs to be considered in this respect is the question of spatial autocorrelation (Jurasinski et al., 2012). What is the optimal distance between flux-measurement chambers to avoid autocorrelation (e.g., pseudoreplication)?

The objectives of this study were: (1) to determine N₂O fluxes within an unmanaged beech forest site in the Hainich National Park at 15 plots during a 1-year period; (2) to calculate if distance between the plots matters to avoid autocorrelation; (3) to test if the set of 15 individually varying soil parameters helps to explain the spatial variation of the N₂O fluxes; (4) to evaluate if a model (Forest-DNDC) without recalibration, as needed for any regionalization, individually run with the set of 15 soil input parameters, can account for N₂O emissions of this near-natural ecosystem. Thereby the relevance of spatial variability for the assessment of N₂O fluxes in unmanaged temperate forests will be elucidated.

2 Materials and methods

2.1 Study site

Our research site is located within the Hainich National Park (NP) (51°04′46″ N, 10°27′08″ E, 440 m asl) in Thuringia, Germany. The Hainich NP was established in 1997 to protect one of the largest beech forests in Central Europe. Due to a unique history as a military base for more than 60 years prior to 1997, a large part of the forest has been taken out of management and developed with little disturbance. In the centuries before, the forest at the Hainich research site was used by the local village population as a coppice with standard systems and therefore has not been exposed to clearcut (Gleixner et al., 2009). As a consequence, the trees cover a wide range of age classes with a maximum up to 250 y. The forest is dominated by beech (65%). The aboveground stem C pool is \approx 130 t C ha⁻¹ (Gleixner et al., 2009). Maximum tree height varies between 30 and 35 m with a maximum leafarea index (LAI) of 5.0 m² m⁻². The long-term mean annual air temperature is 7.5°C–8°C, and the mean annual precipitation is 750–800 mm. The latest estimates for wet and dry deposition for the Hainich area are 25 kg N ha y⁻¹ for 2007 (Builtjes et al., 2010) and show no strong trend over more than a decade.

Within the same area, an intensive soil survey was undertaken for detail soil-C analyses (Schrumpf et al., 2011). The soils are rather shallow and very clayey Eutric Cambisols related to Vertisols. The clay contents in the deeper horizons (20–60 cm) are well > 60% and in the topsoil \approx 50% (Schrumpf, personal communication). The litter layers usually are gone by the end of summer, and by the onset of spring shallow Oi and Oe horizons appear. The mean C stocks for the top 20 cm are 6.92 kg C m⁻² and 4.97 kg C m⁻² for 20 to 60 cm (Schrumpf et al., 2011).

2.2 Field measurements and N₂O-flux analysis

Measurements of N₂O fluxes were carried out using a closed-chamber technique employing 15 cylindrical polyvinyl chloride (PVC) frames (30 cm in diameter and 15 cm tall). The collars were randomly distributed throughout the footprint of an eddy covariance tower (run by the Max Planck Institute for Biogeochemistry, MPI, Jena) as well as in a small valley just outside this fetch. The frames had been set up in the topsoil 2 weeks before starting the gas sampling. The location of each frame was selected stratified randomly.

 N_2O exchange was measured 34 times within the period from November 2005 to November 2006 by placing a PVC lid (30 cm in diameter and 30 cm tall) at each frame and taking five gas samples from the chamber headspace using gastight syringes (60 mL) after 0, 5, 10, 15, and 20 minutes of closure. N_2O concentration was analyzed in the laboratory using an automated gas-chromatograph (GC) system (GC-14B, Shimadzu, Germany) equipped with flame ionization and an electron-capture detector. A detailed description of the GC system is given by Loftfield et al. (1997). For calibration, three certified standards were used (303 ppb, 1000 ppb, 1998 ppb). As no saturation effects were found, fluxes were calculated from the linear slope of the concentration change over time taking into account the headspace temperature and the coefficient of determination for each regression.

During all N₂O measurements at the GC, we regularly performed measurements of the standard gases. Forty replicate measurements of the 303 ppb N₂O standard gas were used to calculate the analytical uncertainty range of the N₂O measurements of 1%. Annual flux rates were calculated by summing up the assumed steady fluxes (1) until the next measurement and (2) since the last measurement, and applying the arithmetic means of both values following Jungkunst et al. (2004).

2.3 Soil and model input parameters

Meteorological data, in this case daily minimum and maximum air temperature as well as daily precipitation, were observed 2 m above ground at a station located outside the forest installed by the MPI.

Additionally, we determined soil temperature and soil moisture at each plot at the time of gas sampling. Soil temperature was measured using a mobile temperature sensor (Testo 110, Testo, Germany), and soil moisture was determined gravimetrically from the top 10 cm of soil following removal of litter by oven-drying at 105°C for 36 h. To compare the gravimetric soil water content with the soil moisture displayed by the Forest-DNDC model (see subsection 2.4) we calculated the water-filled pore space (WFPS) according to Parton et al. (2001) by

WFPS =
$$\theta \left(\frac{BD}{WD}\right) \frac{BD}{1 - \frac{BD}{PD}},$$
 (1)

where θ is the gravimetric soil water content, BD is the bulk density, WD is the density of water, and PD is the particle density of the average soil material (quartz) with a value of 2.65 g cm⁻³.

Bulk density, soil organic C (SOC), and soil pH (H_2O) were determined at the beginning of the measurement period in November 2005. To determine soil bulk density, undisturbed soil samples from 0 to 5 cm depth were taken using stainless-steel soil cores of known volume (100 cm³). Then the samples were oven-dried at 105°C for 24 h. Concentration of SOC was determined by the mean difference of 5 g (air-dried) of the soil sample and 5 g dried at 430°C in a muffle furnace (until constant weight was achieved). For the determination of soil pH, the soil was homogenized and afterwards measured in a soil-towater suspension (1:2.5) using a glass electrode.

2.4 Forest-DNDC

For all 15 plots, we simulated the N₂O fluxes for the Hainich research site using the model Forest-DNDC (http:// www.dndc.sr.unh.edu/). Forest-DNDC simulates C and N dynamics in soil as well as trace-gas exchange (N₂O, CH_4 , N₂, NO, and NH₃) between soil and atmosphere from wetland and upland ecosystems. Driving parameters for the biological processes are climate, physical and chemical soil parameters, vegetation, and anthropogenic activity (Li et al., 2000; Li, 2000; Stange et al., 2000). The general structure was adapted from two existing models: an upland forest biogeochemical model (PnET-N-DNDC) and a hydrology-driven model (Wetland-DNDC; Zhang et al., 2002). For comparison of modeled with measured fluxes, we restricted the analysis to Forest-DNDC-generated daily values for the days of field mesurements. Annual flux rates were calculated in two ways: first identically to the measured fluxes taking the simulated fluxes of the days when measurements had been done and secondly by taking the value calculated by Forest-DNDC by simply adding up the daily modeled fluxes (for modeled daily fluxes see Tab. 1).

Table 1: Overview of N_2O flux rates, mean N_2O flux rates, and annual N_2O flux rates for the measured N_2O fluxes, modeled N_2O fluxes, and daily modeled N_2O fluxes.

	range / μg N m ^{−2} h ^{−1}	annual N₂O flux rate / kg N ha ⁻¹ y ⁻¹
measured fluxes	-4.54 - 40.5	0.49 ± 0.44
modeled fluxes	2.54 – 81.72	1.77 ± 1.82
modeled fluxes (daily) 1.4 – 133.09	1.56 ± 0.006

2.5 Statistical analyses

Further, we tested whether correlations between N₂O flux rates at different plots depend on spatial distance between the plots. We first extracted the spatial coordinates of all 15 plots and computed their mutual distances dist (i, j) for all distinct pairs {i, j}, where i, j are chosen from {1, 2, ..., 15}, yielding $15 \times 14/2 = 105$ distances. For each of the 105 possible pairs of plots, we also computed the cross-correlation coefficient C_{flux} (i, j) of their flux time series (34 time points each). The resulting correlation between the distances and flux correlations was tested for significance using a bootstrap permutation. We repeatedly associated the flux values to randomly chosen plots, again computed the cross-correlation coefficient C_{rand} (i, j), and generated a histogram of 20 000 such randomly occurring correlations obtained from random association of actual fluxes to actual plot locations.

An analogous bootstrap analysis (based on 20 000 random permutations) was performed for accessing the significance of correlations between soil parameters (pH value, SOC content, clay content, and bulk density) and N₂O flux rates.

3 Results

3.1 Physical and chemical soil parameters

Physical and chemical soil parameters (BD, soil pH, SOC, clay content) at the 15 plots are summarized in Tab. 2. Although forest structure and land-use history in the research area are homogeneous (Jurasinski et al., 2012), soil parameters are less so. Bulk density ranged from 0.79 g cm⁻³ (plot 2) to 0.92 g cm⁻³ (plot 4). Soil pH showed the lowest value at plot 2 (4.8) and the highest soil pH at plot 4 (6.7). The clay content and the soil organic carbon showed even larger variability. The clay content varied from 30.8% to 51%, and the SOC ranged from 0.032 to 0.087 kg C (kg soil)⁻¹.

3.2 Soil climate

Figure 1 presents the daily soil temperature and soil moisture simulated with Forest-DNDC and the mean measured soil temperature and soil moisture measured at the time of gas sampling from plot 1. All other plots showed a similar pattern (Tab. 2). The modeled soil temperature matched well with the measured soil temperature except for soil temperature near freezing. The modeled soil water content shows good agreement in the period from October 2005 to June 2006, but in the period from July to October 2006 the Forest-DNDC model overestimates the soil water content (Fig. 1). The modeled

Table 2: Plot-specific soil parameters (soil pH, soil organic carbon [SOC], clay content, bulk density [BD], and measured and modeled N₂O fluxes) of all 15 plots of the Hainich research site.

Plot	рН	SOC	Clay	BD	<mark>Ν₂Ο</mark> / μg N m ⁻² h ⁻¹			
			/ %	/gcm-3				
		/kg C (kg soil)−1			measured		modeled	
					average	SD	average	SD
1	4.9	0.033	36.2	0.9	1.44	21.88	16.31	32.61
2	4.8	0.032	30.8	0.86	-0.49	10.25	17.79	36.04
3	5.3	0.05	45.3	0.92	12.68	22.05	13.50	25.40
4	6.7	0.087	50.5	0.88	0.22	15.26	79.63	66.26
5	5.8	0.04	42.0	0.84	3.85	16.33	11.05	22.42
6	5.4	0.036	39.3	0.86	2.95	17.33	14.49	28.94
7	5.3	0.041	36.4	0.87	7.95	18.86	15.73	31.33
8	6.2	0.047	41.2	0.86	10.47	23.28	7.27	15.64
9	6.0	0.064	51.0	0.89	13.38	19.42	17.30	21.80
10	6.6	0.062	48.6	0.79	6.53	11.03	17.30	21.80
11	5.7	0.052	44.2	0.79	5.98	13.41	8.58	17.60
12	5.3	0.054	35.0	0.81	5.03	16.75	14.48	26.97
13	6.4	0.05	44.5	0.86	3.54	22.33	5.12	11.25
14	6.1	0.041	38.7	0.87	2.81	17.20	8.70	18.03
15	6.2	0.046	40.2	0.91	-1.61	20.30	6.81	14.68
Average	6.1	0.049	41.6	0.86	4.98 (n = 510) 16.93 (n = 510)		510)	
SD	0.6	0.014	5.8	0.04	18.42		33.41	



Figure 1: (a) Daily soil temperature (0–12 cm depth) from plot 1 simulated by Forest-DNDC. Solid circles represent the soil temperature (integral between 0 and 12 cm) measured at the time of gas-flux sampling at plot 1. (b) Daily soil WFPS (0–12 cm depth) from plot 1 simulated by Forest-DNDC. Solid circles represent the WFPS calculated with the gravimetric soil water content (0–10 cm) measured at the time of gas-flux sampling at plot 1.

soil water content showed no values below 0.57 WFPS. In contrast, the measured soil water content ranged between 0.28 and 0.71 WFPS.

3.3 Measured N₂O flux rates

Figure 2 presents an overview of air temperature, precipitation, and both the average measured and modeled N₂O flux rates obtained from 15 plots distributed over the Hainich research site for the years 2005 and 2006. The mean measured N₂O flux rates were obtained from averaging flux rates of all 15 plots. A seasonal pattern of measured N₂O exchange was lacking. Average measured N₂O-N flux rates (November 2005 to November 2006) exhibited small amplitudes between -4.54 and 40.5 μ g m⁻² h⁻¹, but most average measured N₂O flux rates do not significantly differ from zero (t-test, p = 0.05) (see Fig. 1). A portion of 38% of all (n = 510) observed N₂O flux rates shows negative values. The highest measured N₂O flux rates occurred between January and February 2006. During this time, there was an extended frost period with soil temperature below -0.5°C. This period contributes 40% to the annual measured N₂O flux rate. A second period with mean measured N₂O flux rates significantly different from zero started at the end of June 2006 (June 21-28, 2006) and contributes 15% to the annual measured N₂O flux rate. During this period, the air temperature strongly increased (see Fig. 1). The annual measured N₂O flux rate for the Hainich research site for the 1-year measuring period

(November 2005 to November 2006) was (0.49 \pm 0.44) kg N ha-1 y-1. The 15 individual plots showed measured N₂O flux rates between -87.6 and 121.6 µg N m⁻² h⁻¹. During periods with higher mean N₂O emissions, only small negative fluxes occurred (see Fig. 3) and these fluxes are within the uncertainty range. During the frost period, nearly all plots exhibited positive measured N₂O flux rates. Also the spatial variability showed high values in this period (range: -17.5 to 94.6 µg N m⁻² h⁻¹, mean: 27.5 μ g N m⁻² h⁻¹). In the second period (end of June) with N₂O flux rates that were significantly different from zero, the spatial variability was also high (range: -18.0 to 121.6 μg N m⁻² h⁻¹, mean: 30.1 μg m⁻² h⁻¹). During the periods with a background emission pattern (Brumme et al., 1999), both negative and positive N₂O fluxes occurred at similar ratio (range: -54.3 to 88.8 μ g N m⁻² h⁻¹, mean: 3.4 µg N m⁻² h⁻¹).

3.4 Spatial correlation of measured N₂O flux rates between different plots

There is a weak positive correlation (correlation coefficient 0.085) suggesting that flux correlations between plots tend to be larger at the more distant plots (Fig. 4a). However, flux correlations are broadly distributed in the range between -0.3 and +0.77 and the best linear fit (least square regression) shows only a low average increase of flux correlations with distance at 0.17 km⁻¹. Together with the small correlation coefficient, this suggests that the correlation between the



Figure 2: (a) Daily mean air temperature for 2005 and 2006 recorded 2 m above the ground and (b) daily precipitation for 2005 and 2006. (c) The black dots are the mean field N₂O flux rates with (n = 15) for the Hainich research site, the gray squares are the mean modeled N₂O flux rates (n = 15), and the gray line shows the mean daily modeled N₂O flux rates. The error bars on each individual data point are the standard deviation.



Figure 3: Range of N_2O fluxes between the individual plots: The dark gray zone presents the range within measured N_2O fluxes occurred and the light gray zone presents the range within modeled N_2O fluxes occurred. The inset shows the course of simulated N_2O fluxes from selected plots.



Figure 4: Interplot flux correlations do not significantly correlate with intersite distances. (a) Intersite correlations of nitrous oxide fluxes plotted versus intersite distances. The weakly positive trend is indicated by the least-squares linear fit (line, rate of change in correlation is 0.17 km⁻¹). (b) Histogram of 20 000 correlations from location-randomized bootstrap samples indicate a weakly positive correlation of 0.084 (vertical black line), i.e., flux correlations between distant sites trends to be larger than between nearby sites. However, this positive trend is not significant.

 N_2O flux rates at any two plots is almost independent of the distance between them. Indeed, a randomized (bootstrap) sampling of the given data (see section 2) yields some insight into whether this slightly positive correlation might be significant. We therefore created 20 000 random permutation samples by randomly associating plots with flux time series, obtaining the distribution shown in Fig. 4b. We observe that 25.7% of the correlations between N_2O flux correlations and randomized plot locations are larger than the observed value of 0.084%, and 74.3% are lower. This strongly indicates that there is no significant correlation between the actual interplot distances and the actual interplot N_2O flux correlations at spatial distances < 400 m.

3.5 Modeled N₂O flux rates

The modeled N₂O flux rates of the 15 individual plots showed no negative values. They ranged between 0.0 and 255.7 μg N m⁻² h⁻¹ (see inset in Fig. 3). The 15 individual plots exhibited the highest variability in modeled N₂O flux rates from each other (high standard deviation) on May 23, 2006 (Fig. 3). Plot 4, and, to a lesser degree, plot 9 and plot 10, with high N₂O fluxes, drive this high standard deviation (Tab. 2).

Mean modeled N₂O-N flux rates (2.54 to 81.72 μ g m⁻² h⁻¹) were typically higher than the actual measured N₂O-N flux rates (-4.54 to 40.5 μ g m⁻² h⁻¹). During this period, modeled

 N_2O flux rates (1.40–133.09 μg N m⁻² h⁻¹) were up to three times larger than the measured N_2O flux rates. Mean modeled N_2O flux rates showed lowest values from November 2005 to March 2006 and from September to November 2006. Highest values occurred at the end of April 2006 (see Fig. 1). Furthermore, the daily modeled N_2O flux rates showed a weak seasonal pattern. The model does not account for N_2O uptake. The annual modeled N_2O flux rate at measurement intervals and the annual modeled N_2O flux in daily resolution for the Hainich research site for the 1-year measuring period (November 2005 to November 2006) were (1.77 \pm 1.82) and (1.56 \pm 0.006) kg N ha⁻¹ y⁻¹, respectively.

4 Discussion

4.1 Comparison with other studies

Except for the recent study by Guckland et al. (2010), all previously published studies focusing on N₂O exchange from soils in beech-forest ecosystems were performed in managed forest ecosystems (references in Tab. 3). Still our results agree well with the results of Brumme et al. (1999) and Brumme and Borken (2009) for forests dominated by the background-emission type. They found that soils in beech forests with a mull organic horizon usually show background emissions sometimes interrupted by event emissions like frost and thaw. The annual N₂O flux rates ([0.49 \pm 0.44] kg N

 $ha^{-1} y^{-1}$) were within the lower range of values reported for temperate beech-forest soils (see Tab. 3).

Contrasting to the other studies, the core zone of the Hainich NP is a close to natural deciduous forest which is unique in Central Europe. This could be a reason why most N₂O fluxes did not significantly differ from zero. It may also well be that N is immobilized as a part of rising soil organic-matter stocks (Gleixner et al., 2009). In any case, it clearly supports the notion that most natural ecosystems do not emit significant amounts of N₂O and natural background emissions are somewhat lower than the simulated average value with PnET-N-DNDC for forests in Germany (Kesik et al., 2005). Guckland et al. (2010) found even lower emission rates of N₂O ranging from -31.4 to 167.8 μ g N m⁻² h⁻¹ at a site covered to 59% with beech also located in the Hainich NP. These measurements were performed on loess soils (Luvisols) with lower clay content than in this study resulting in an annual N₂O flux rate of (0.19 \pm 0.16) kg N ha⁻¹ y⁻¹. The N₂O fluxes measured by Guckland et al. (2010) during freezing and thawing amounted to 94% of the emissions of the first year. Our values of 38% are closer to the values observed by Papen and Butterbach-Bahl (1999) who estimated a contribution up to 39% to the total annual N₂O emission caused by freezing and thawing at a 96-y-old beech plantation in the Höglwald (S Germany). Therefore, the bulk of N₂O emissions at the Hainich site is related to frost. Similar results were also observed by Butterbach-Bahl et al. (2002), Papen and Butter-

Table 3: Compilation of published annual N₂O flux rates from soils of temperate beech forests.

Site	Annual N₂O flux / kg N ha ⁻¹ y ⁻¹	Observation period	Reference
Hainich ^a , Germany	0.49 ± 0.44	2005–2006	this study
Hainich ^p , Germany	0.11 ± 0.11	2005–2007	Guckland et al. (2010)
Solling, Germany	1.93 ± 0.63	1990–2000	Brumme and Borken (2009)
Solling, Germany	0.54 ± 0.14	2000–2001	Borken and Beese (2006)
Göttinger Wald, Germany	0.17 ± 0.03	1993–1995	Brumme et al. (1999)
Zierenberg, Germany	0.41 ± 0.12	1991–1992	Brumme et al. (1999)
Schottenwald, Austria	4.03 ± 1.37	1996–1998	Zechmeister-Boltenstern et al. (2002)
Klausenleopoldsdorf, Austria	$\begin{array}{c} 6.82 \pm 0.5 \\ 7.63 \pm 0.5 \end{array}$	2002–2003 2003–2004	Kitzler et al. (2006) Kitzler et al. (2006)
Schottenwald, Austria	$\begin{array}{c} 10.42 \pm 0.6 \\ 10.15 \pm 0.4 \end{array}$	2003–2004 2003–2004	Kitzler et al. (2006) Kitzler et al. (2006)
Tuttlingen, Germany:			
SW exposed, control SW exposed, thinned NE exposed, control NE exposed, thinned NW exposed, control NW exposed, thinned	0.07 0.05 0.05 0.04 0.22 0.86	2003–2004 2003–2004 2003–2004 2003–2004 2003–2004 2003–2004	Dannemann et al. (2006) Dannemann et al. (2006)
Börnhöved Lake region, Germany	0.4	1993	Mogge et al. (1998)
Soroe, Denmark	0.5	1997	Beier et al. (2001)
Höglwald, Germany	5.1	1995–1996	Papen and Butterbach-Bahl (1999)
Höglwald, Germany	3.8 ± 1.3	1995–1997	Butterbach-Bahl et al. (2002)

a main soil texture: clay; b main soil texture: silt (loess)

bach-Bahl (1999), Brumme et al. (1999), and Teepe et al. (2004). The absence of a peak of N₂O emission during thawing could be related to insufficiently high sampling frequency (Flessa et al., 2002b). We observed no seasonal pattern of N₂O fluxes that would be detectable with our temporal resolution and is in accordance with the results from Brumme et al. (1999) and Brumme and Borken (2009) who also reported no seasonal pattern of N₂O emissions < 1 kg N ha⁻¹ y⁻¹.

Significantly higher annual N₂O flux rates usually derive from sites showing a clear seasonal pattern with low N₂O fluxes during the winter months and high N₂O fluxes during the summer. Seasonal emission patterns like the one observed by Zechmeister-Boltenstern et al. (2002) for a 140-y-old beech stand located at Schottenwald (Austria) report (3.6 ± 1.0) kg N ha⁻¹ y⁻¹ in 1996, (4.2 ± 1.3) kg N ha⁻¹ y⁻¹ in 1997, and (4.3 \pm 1.8) kg N ha⁻¹ y⁻¹ in 1998. Kitzler et al. (2006) found N₂O flux rates ranging between -6.3 and 75.4 µg N m-2 h-1 for site with a 142-y-old beech stand at Schottenwald (Austria) and between -1.0 and 82.8 µg N m⁻² h⁻¹ in a 62-y-old beech forest at Klausenleopoldsdorf (Austria) in 2002. The seasonal variations in N₂O emissions at these sites followed mainly forest management and the annual changes in soil temperature, soil moisture, and the availability of N in the soil.

4.2 Measured vs. modeled N₂O exchange

Our results reveal that measured and modeled soil temperatures fit well except for the period with soil temperature near the freezing point. The soil cooled down slower and warmed up faster than the model simulation.

Similar results were found by Szyska et al. (2008) who applied the original version of DNDC designed for agricultural ecosystems and not Forest-DNDC. In contrast, measured soil moisture differed significantly from modeled soil moisture, which does not fall to WFPS values < 0.57. The reason for this is the model settings, particularly the wilting point, for the default soil texture "clay loam". However, both parameters significantly affect the N₂O exchange and Forest-DNDC was designed with a strong soil-moisture control on N₂O emissions (Lamers et al., 2007; Zhang et al., 2002). Only a few studies report about the match between measured and Forest-DNDC-simulated soil water contents. Kröbel et al. (2010) found an overestimation of the soil water content by DNDC (version DNDC89). At a site in Scotland on a glacial till with very low hydraulic conductivity in the subsoil which makes drainage very slow, Frolking et al. (1998) reported DNDC to underestimate soil moisture. Beheydt et al. (2007) also mentioned an underestimation of WFPS for different investigated sites in their study using the DNDC version 8.3P. Saggar et al. (2004) reported a poor match between measured and simulated WFPS for a silt-loam soil in New Zealand using NZ-DNDC. However, a successful simulation of soil temperature and soil water conditions is necessary for a successful simulation of N₂O flux rates (Saggar et al., 2004).

The annual N₂O flux rate of the Forest-DNDC simulation overestimated the annual measured N₂O flux rate. Forest-DNDC also fails to correctly simulate the actual fluctuations

of measured N₂O flux rates except for the plot with the highest SOC content (plot 4), which reflected a part of the fluctuations. Forest-DNDC failed to show the N₂O emissions during frost expect for plot 4, plot 9, and plot 10. These plots showed higher N₂O emissions during frost. This suggests that the ground was not frozen deeply and that the measured N2O release is related to processes typically related to frost-thaw cycles as a sudden increase of bioavailable C and N (Röver et al., 1998) or spontaneous release of previously produced N₂O (Goodroad and Keeney, 1984). These small-scale differences can be triggered by differences in soil C content and the model seems to be very sensitive to the parameter setting for SOC, because all three plots exhibit SOC contents > 0.06 kg C (kg soil)⁻¹. At the plots with < 0.06 kg C (kg soil)-1 SOC, no increasing N₂O emissions during frost took place. The model produced thaw-induced elevated N₂O flux rates at the end of April. This event contributes to a great part to the annual N₂O flux rate. However, we did not observe a thawing peak in the field, but due to the short duration of such peaks, this is not surprising. Consequently, this is in line with Stange et al. (2000), who also reported disagreements between modeled and measured N₂O flux rates especially during freezing and thawing. The second N₂O peak in summer 2006 did not appear in the simulated data. This confirms the findings by Saggar et al. (2004), who remarked that the DNDC model had limited success in predicting the size and timing of very high emissions. Abdalla et al. (2009) also found that DNDC poorly described those fluxes from zero-fertilizer treatments.

As it is impossible to provide the data needed for calibration of large and insufficiently examined areas, N₂O fluxes for these cannot be achieved by parameter-intensive models. As these input parameters can only be derived by referring to ubiquitously available proxies as digital elevation models (Glatzel and Bareth, 2006), it is unlikely to achieve realistic mechanistic upscales in the near future. This does not apply to the simulation of natural N₂O fluxes from clean-air ecosystems that do not exist anymore in Central Europe as the prediction of reactions of the ecosystem reaction to external (climatic) and internal (soil) may very well be simulated best by calibrated models.

As the parameter settings for denitrification in versions of Forest-DNDC without site-specific recalibration do not permit N₂O uptake, all modeled N₂O fluxes had a positive sign. 38% of the measured N₂O flux rates were negative and also other studies observed negative N₂O flux rates (Kitzler et al., 2006; Chapuis-Lardy et al., 2007; Guckland et al., 2010). However, the measured N₂O flux rates are usually small and the standard errors of these fluxes are high. Although the PnET-N-DNDC version presented by Stange (2001) did simulate N₂O uptake, the small involved amounts are a challenge to modeling N₂O field fluxes.

4.3 Spatial variability of N₂O flux rates

The spatial variability in N_2O emissions is naturally large (Folorunso and Rolston, 1984; Mathieu et al., 2006; Turner et al., 2008) but so are underlying soil variables. This applies to measurements derived from low spatial representation

(small chambers or taken from small soil augers) and was confirmed by our data. Our measured N₂O flux rates observed at the 15 individual plots show their highest spatial variability when high N₂O flux rates occur. However, also in periods with low mean N₂O flux rates the variability was pronounced. The modeled N_2O flux rates simulated with individual soil parameters from the 15 plots also exhibited a high spatial variability. This is due to the differences in the parameter settings for SOC. N₂O fluxes at plots with low (0.032-0.054 kg C [kg soil]-1) SOC content fluctuated less than plots with high (0.062–0.087 kg C [kg soil]⁻¹) SOC concentrations. The measured N₂O flux rates did not display this effect. Therefore, the differences in SOC do not explain differences found in N₂O as model results frequently suggest. Nevertheless, also the modeled N₂O flux rates showed their highest variability when high N₂O flux rates occur.

4.4 Spatial correlation of N₂O fluxes

One may assume that N₂O fluxes from nearby sites are more strongly correlated than N₂O fluxes from more distant sites, but we are not aware of any studies on this issue. We observed no autocorrelation of N₂O flux rates at the Hainich research site. This confirms the results of Jurasinski et al. (2012), who found soil CO₂ efflux to lack autocorrelation except for summertime fluxes with dominant modeled ash fine-root biomass at the same site as the one examined in this contribution. For N₂O, studies about spatial variability usually focus on differences of emissions caused by differences in soil properties (Ambus and Christensen, 1995; Röver et al., 1999). For our 15 different plots at the Hainich research site, we found no relationships between individual soil parameters and N₂O flux rates.

A potential reason for lacking autocorrelation between the plots could be that the minimum distance is not short enough. However, in our opinion it is more likely that driving parameters as C : N ratio (Klemedtsson et al., 2005), N input (Ambus and Robertson, 2006), and soil water status (Jungkunst et al., 2008) vary in a short range as much as in longer distances. The latter two are very well determined by the heterogeneity of interception which hardly ever is determined. Also, Don et al. (2012) discovered that small-scale heterogeneity of SOC (a major prerequisite for N₂O emissions) was so high that paired sampling did not significantly reduce the number of required samples. Therefore, there are no indications given that "realistic" mean values for one ecosystem type are best derived by wide and randomly spread measurements. Several chambers in close proximity to each other are more pragmatic for a measurement team and most likely produce a similar variability as more distanced chamber measurements. Unfortunately, the spatial variability of driving factors does not really explain spatial variability of N₂O fluxes. The reason for that should be investigated in more detail. For the moment, mean ecosystem values better explain mean fluxes for which Forest-DNDC and others have been designed anyhow. Still, Forest-DNDC without site-specific recalibration could not account for N₂O emissions of this near-natural ecosystem. This is mainly due to the inability of Forest DNDC to account for event-based peaks.

5 Conclusions

Like other forest ecosystems that lack strong seasonal patterns and show few event-induced occasions of N₂O release, the beech forest examined in this study displayed annual N₂O flux rates < 0.5 kg N ha⁻¹ y⁻¹. The core area of the Hainich NP is the closest approximation of a natural ecosystem in Central Europe. Therefore, we propose a maximum background emission factor of 0.5 kg ha⁻¹ N y⁻¹. Our study also indicates that Forest-DNDC without recalibration is not appropriate for simulating annual fluxes of N₂O for zero-fertilizer treatments. Therefore, a Forest-DNDC-based regionalization of N₂O fluxes without a wealth of site-specific recalibration parameters is inappropriate. A broader validation of the model seems to be necessary especially for sites with low SOC values.

The absence of spatial correlations of N₂O flux indicates that within one site the distance between each chamber is secondary. This is valuable for designing measurement plots because larger distances between individual chambers are not required. The latter of course needs to be further verified by additional studies. It remains a challenge for future measurements and modeling to satisfactorily reproduce the spatial variability between sites and within sites of natural N₂O emissions.

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