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Inventories of N_2O and NO emissions from European forest soils

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Abstract. Forest soils are a significant source for the primary and secondary greenhouse gases N_2O and NO. However, current estimates are still uncertain due to the still limited number of field measurements and the herein observed pronounced variability of N trace gas fluxes in space and time, which are due to the variation of environmental factors such as soil and vegetation properties or meteorological conditions. To overcome these problems we further developed a process-oriented model, the PnET-N-DNDC model, which simulates the N trace gas exchange on the basis of the processes involved in production, consumption and emission of N trace gases. This model was validated against field observations of N trace gas fluxes from 19 sites obtained within the EU project NOFRETETE, and shown to perform well for N_2O (r^2=0.68, slope=0.76) and NO (r^2=0.78, slope=0.73). For the calculation of a European-wide emission inventory we linked the model to a detailed, regionally and temporally resolved database, comprising climatic properties (daily resolution), and soil parameters, and information on forest areas and types for the years 1990, 1995 and 2000. Our calculations show that N trace gas fluxes from forest soils may vary substantial from year to year and that distinct regional patterns can be observed. Our central estimate of NO emissions from forest soils in the EU amounts to 98.4, 84.9 and 99.2 kt N yr^{-1}, using meteorology from 1990, 1995 and year 2000, respectively. This is <1.0% of pyrogenic NO_x emissions. For N_2O emissions the central estimates were 86.8, 77.6 and 81.6 kt N yr^{-1}, respectively, which is approx. 14.5% of the source strength coming from agricultural soils. An extensive sensitivity analysis was conducted which showed a range in emissions from 44.4 to 254.0 kt N yr^{-1} for NO and 50.7 to 96.9 kt N yr^{-1} for N_2O, for year 2000 meteorology.

The results show that process-oriented models coupled to a GIS are useful tools for the calculation of regional, national, or global inventories of biogenic N trace gas emissions from soils. This work represents the most comprehensive effort to date to simulate NO and N_2O emissions from European forest soils.

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1 Introduction

The atmospheric concentration of nitrous oxide (N_2O) has been increasing in recent decades with a rate of approx. 0.25% yr^{-1} (IPCC, 2001). Among other sources, forest soils have been acknowledged to represent significant sources of this potent greenhouse gas (e.g. Schmidt et al., 1988; Skiba et al., 1994). Emissions of N_2O from forest soils have most likely increased in recent decades and will probably increase in the future due to the anthropogenic perturbation of the global N cycle (Galloway et al., 2004) and hence high rates of atmospheric N deposition to many forest ecosystems in Europe, North America and Asia (Aber et al., 1989; Bowden et al., 1991). In a series of recent

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publications evidence has been provided that N deposition to forest ecosystems are positively correlated with N_2O emissions (Brumme and Beese, 1992; Butterbach-Bahl et al., 1998; Zechmeister-Boltenstern et al., 2002).

On a global scale soils have been identified as sources of atmospheric NO_x of a comparable magnitude like combustion processes (Davidson and Kingerlee, 1997). This especially applies to acidic forest soils, which have been shown to act primarily as sources of NO, a reactive trace gas involved in the production of tropospheric ozone (Williams et al., 1992; Butterbach-Bahl et al., 2002a). However, due to its reactivity only a part of the NO emitted from forest soils will reach the atmosphere, whereas some of the NO will react with ozone to NO_2, associated with a partial re-deposition of NO_2 to plant and soil surfaces or an uptake by plant tissues (Duyzer and Fowler, 1994; Meixner, 1994; Gessler et al., 2001; Dorsey et al., 2004). As for N_2O, atmospheric N deposition to forest ecosystems has been shown to be closely related to the source strength of forest soils for NO (Gasche and Papen, 1999; Pilegaard et al., 1999; Van Dijk and Duyzer, 1999).

The emission of NO and N_2O from forest soils is mainly the result of simultaneously occurring production and consumption processes, most of which are directly linked to the microbial N turnover processes of nitrification and denitrification (Conrad, 1996, 2002). With regard to NO also the abiotic process of chemo-denitrification, during which biologically produced nitrite is chemically decomposed to NO, has been shown to be an important production process in soils at pH values lower than 4.0 (Van Cleemput and Baert, 1984). Like most other biological processes, microbial turnover processes vary largely on spatial and temporal scales, since they are significantly influenced by a number of environmental factors such as climate and meteorological conditions, soil and vegetation properties or human management of the land surface. Due to this also the emission of N trace gases from forest soils have been observed to vary over several orders of magnitudes between seasons, years or measuring sites (Papen and Butterbach-Bahl, 1999; Brumme et al., 1999; Butterbach-Bahl et al., 2002b). This variability is the reason for the still high uncertainty of current regional and global estimates of N trace gas emissions from soils. In recent years it has been proposed that linking process-oriented models, which are able to simulate the processes involved in N trace gas emissions from soils, to detailed GIS databases, holding explicit spatial information on major drivers of microbial processes, can serve as tools to improve current estimates of the magnitude of terrestrial sources and sinks of atmospheric trace gases (Brown et al., 2002).

This approach, i.e. the use of process-oriented biogeochemical models for calculating inventories of N trace gas emissions from soils, was also followed within the EU funded project NOFRETETE (**N**itrogen **O**xides Emissions **fr**om **E**uropean Fores**t** **E**cosys**te**ms). This manuscript provides details of model modifications, model testing and the establishment of a GIS database which was finally used to initialize and drive the PnET-N-DNDC model for the calculation of inventories of N trace gas emissions of forest soils of Europe. It represents the most comprehensive effort to date to simulate NO and N_2O emissions from European forest soils.

2 Materials and methods

2.1 The PnET-N-DNDC model

For the calculation of N_2O and NO emission inventories of European forest soils the biogeochemical model PnET-N-DNDC was used. The model has been already applied for regional emission inventories for temperate forest ecosystems (Butterbach-Bahl et al., 2001, 2004), and recently, after some adaptation, also for the calculation of a N_2O emission inventory for tropical rainforests in Australia (Kiese et al., 2005). The PnET-N-DNDC model was developed to predict soil carbon and nitrogen biogeochemistry in temperate forest ecosystems and to simulate the emissions of N_2O and NO from forest soils (Li et al., 2000; Stange et al., 2000). The model is mainly based on the PnET model (= **P**hotosy**n**thesis-**E**vapo**t**ranspiration-Model), the DNDC model (=**De**n**i**trification-**Dec**omposition-Model) and a nitrification module. The PnET model is a forest physiology model used for predicting forest photosynthesis, respiration, organic carbon production and allocation, and litter production. It was originally developed by Aber and Federer (1992). This model has already been used in regional studies in order to predict the sensitivity of forest production to climate variability and site quality (e.g. Goodale et al., 1998). DNDC is a soil biogeochemistry model used for predicting soil organic matter decomposition, nitrogen turnover and N_2O production in agricultural soils (Li et al., 1992). This model has also been used to predict regional N_2O emissions from agriculture in US, China and UK (Li et al., 1996, 2001, 2004; Brown et al., 2002). The nitrification module was developed by Stange (2000) in order to simulate nitrification rates, the growth of nitrifier populations and the nitrification induced N_2O and NO emissions associated with nitrification.

In the PnET-N-DNDC model N_2O and NO emissions from soils are directly influenced by environmental factors, such as soil temperature and moisture, pH and substrate availability (C- and N-content). These environmental factors are driven by different ecological drivers, namely climate, soil properties, vegetation and anthropogenic activities. Five modules for predicting forest growth, soil climate, decomposition, nitrification and denitrification are linked to translate the environmental factors and ecological drivers into predicted N_2O and NO emissions. The functions of the different modules are as follows: a) the soil climate module is used to convert daily climate data into soil temperature and moisture profiles and to calculate soil oxygen availability in the forest soil

profile; b) the forest growth module simulates forest growth as a function of solar radiation, temperature, water and N availability. The forest growth module is linked to the soil climate and decomposition modules via litter production and water and N demand; c) the decomposition module simulates the turnover of litter and other organic matter and, hence, the production of ammonium, nitrate and dissolved organic carbon (DOC) in the soil driven by temperature, moisture and O_2 availability in the soil profile; d) the nitrification module predicts growth and death of nitrifiers, the nitrification rate as well as N_2O and NO production during nitrification depending on soil temperature, moisture, ammonium and DOC concentrations; e) depending on the population size of denitrifiers, soil temperature, moisture and substrate concentrations (DOC, NO_3^-, NO_2^-, NO and N_2O), the denitrification module simulates the individual steps of the sequential reduction of nitrate or other oxidized N compounds (NO_2^-, NO, N_2O) to the final product N_2.

The nitrification and denitrification induced N_2O and NO fluxes are calculated based on the dynamics of soil aeration status, substrate supply and gas diffusion. Chemo-denitrification, i.e. chemical decomposition of NO_2^- to NO, is considered as another source of NO production in soils. This chemical reaction is controlled by the concentration of nitrite in the soil, soil pH and temperature. To handle the problem of simultaneously occurring aerobic and anaerobic processes in adjacent microsites, the PnET-N-DNDC model uses the concept of a so-called "anaerobic balloon". Based on the O_2 diffusion from the atmosphere into the soil and the O_2 consumption during heterotrophic and autotrophic respiration the O_2 concentration is calculated for a given soil layer. The O_2 concentration is assumed to be reciprocally proportional to the anaerobic fraction within this soil layer (Li et al., 2000). For further details on the PnET-N-DNDC model we refer to Li et al. (1992, 1996, 2000), Li (2000), Stange et al. (2000), Butterbach-Bahl et al. (2001, 2004) and Kiese et al. (2005).

To simulate N trace gas emissions for a specific site the PnET-N-DNDC requires the following input parameters: daily climate data (precipitation, minimum and maximum air temperature, optional: solar radiation), soil properties (texture, clay content, pH, soil organic carbon content, stone content, humus type), and forest data (forest type and age, above-ground and below-ground biomass, plant physiology parameters). The PnET-N-DNDC is currently parameterized for 12 tree species/genera, i.e. pine, spruce, hemlock, fir, hardwoods, oak, birch, beech, slash pine, larch, cypress and evergreen oak. Whenever there are no site-specific forest data available except for the forest type and age, the model calculates with default values for each forest type taken from an internal database of literature data (Li et al., 2000).

Since several authors have discussed the importance of the forest floor humus type for N trace gas emissions (e.g. Brumme et al., 1999; Butterbach-Bahl et al., 2002a), we shortly want to discuss how the PnET-N-DNC model deals with the effect of humus type on processes involved in N trace gas emissions. In the model the effect of the humus on N trace gas production is indirect: the humus type influences the partitioning of SOC into different fractions (humus, humads, litter) with their specific decay constants (Li et al., 2000). For a forest floor with mull as humus type the SOC fractions with short turnover times are highest, medium in moder and smallest if the humus type is rawhumus. This does affect the C as well as N availability and, thus, the C and N turnover rates and finally also the processes involved in N trace gas emissions. Furthermore, the humus type is also influencing the density and the porosity of the organic layer, which also results in differences in soil climatic conditions such as water and temperature distribution in the soil layers.

Furthermore, the model needs information about inorganic N concentrations in rainfall which are used to calculate throughfall values for N. Throughfall is a surrogate of wet and dry deposition and is depending on forest type and N concentration. The equations used in PnET-N-DNDC to calculate throughfall values are described by Li et al. (2000).

No structural changes were applied to the PnET-N-DNDC model for its use within the NOFRETETE project. However, based on results from laboratory studies by Kesik et al. (2005)[1] the parameterisation of NO production by chemo-denitrification (Chem_NO) in dependency on the soil pH (soil_pH[layer]) and the nitrite concentration (NO_2[layer]) in the respective soil layers was changed as follows:

$$\text{Chem_NO}\,[\text{kg N ha}^{-1}\,\text{day}^{-1}] = 300 \times \text{NO}_2[\text{layer}] \times 16\,565 \times \exp^{(-1.62\times\text{soil_pH[layer]})} \times f1)$$

f1 is an Arrhenius type function describing the temperature dependency (temp[layer]) of chemo-denitrification:

$$f1 = \exp^{\left(\frac{-31494}{(\text{temp[layer]}+273.18)\times 8.3144}\right)} \quad \text{(Stange, 2000)}$$

It should be noted that these algorithms do not consider the pH microsite variability in soils, which have been found to be significant in some soils (Häussling et al., 1985; Strong et al., 1997; Bruelheide and Udelhoven, 2005). This means that we assume a uniform bulk soil pH. However, future model developments may be necessary to address the microsite variability of soil pH and to assess consequences of this variability on NO production via chemo-denitrification.

Furthermore, the parameters for the moisture dependency of gross nitrification and N_2O and NO production and consumption during nitrification and denitrification (see Li et al., 2000) were optimised according to the results from laboratory studies (see Schindelbacher et al., 2004; Kesik et al., 2005[1]).

[1]Kesik, M., Blagodatsky, S., Papen, H., and Butterbach-Bahl, K.: Effect of pH, temperature and substrate on N_2O, NO and CO_2 production by Alcaligenes faecalis p. J. Applied Microb., submitted, 2005.

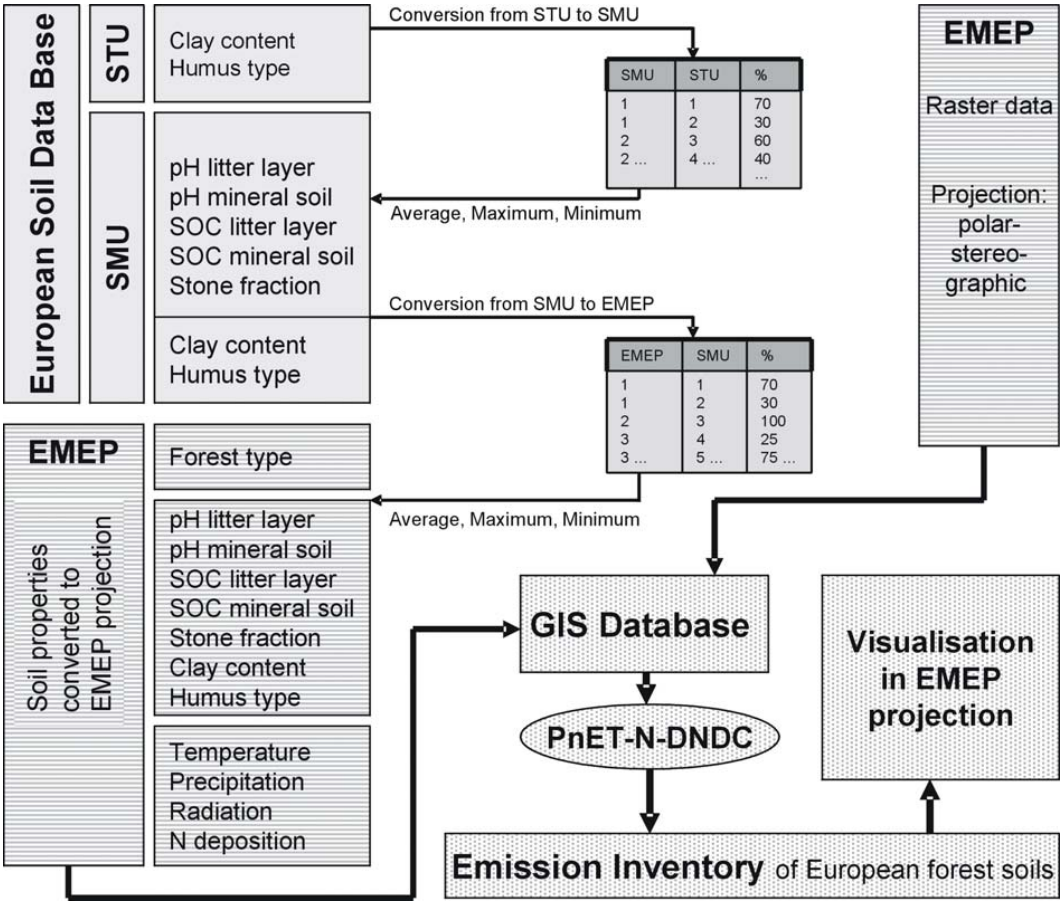

Fig. 1. Scheme for data aggregation and visualisation used for the calculation of a regional inventory of N trace gas emissions from forest soils.

2.2 Evaluation sites

The capability of the PnET-N-DNDC model to simulate N trace gas emissions from forest soils was tested by comparing model results with results from field measurements at 19 different field sites across Europe and US (Table 1). Most of these sites were measuring sites within the NOFRETETE project. The testing sites were well distributed across Europe, with a boreal forest site at Hyytiälä, Finland, forest sites in temperate maritime climate (e.g. Speulderbos, Netherlands, Sorø, Denmark, and Glencorse, UK) and temperate continental climate (Matrafüred, Hungary) and forest sites exposed to Mediterranean climate (San Rossore, Parco Ticino, Italy) (Table 1). For all sites information on model input parameters (see Sect. 2.1) as well as on N trace gas fluxes were aggregated in a database for model testing.

2.3 GIS database

A detailed GIS database covering all EU states plus Romania, Bulgaria, Switzerland and Norway with all relevant initialization and driving parameters and variables was created for the regionalization of N trace gas emissions by use of the PnET-N-DNDC. Spatially resolved information included soil, forest and climate properties. Within the NOFRETETE project information on meteorological data and on atmospheric N deposition was provided by the Norwegian Meteorological Institute (MET.NO), from the inputs of the EMEP MSC-W photo-oxidant model (Sandnes-Lenschow et al., 2000; Simpson et al., 2003), on soil properties and on forest information by the Joint Research Centre (JRC) at Ispra, Italy. Since data were delivered in different formats and projections, transformations into the ArcGIS format and the Lambert Azimuthal projection on the basis of the EMEP (European Monitoring and Evaluation Program) raster were necessary. The used EMEP raster is a polar stereographic projected grid with a resolution of 50 km×50 km at 60° North (Simpson et al., 2003; http://www.emep.int).

2.3.1 Soil information

The soil data were retrieved from the Soil Geographical Data Base of Europe (SGDBE) at a scale of 1:1 000 000. This data base is part of the European Soil Data Base (http://eusoils.

Table 1. Site characteristics of the temperate forest sites used for model testing.

Site, Country	Coordinate	Forest Type	Forest Age [yr]	Humus Type	Soil Texture	Clay [%]	pH Forest floor	pH Mineral soil	SOC [kg C kg^{-1}]	Years	N Input [kg N yr^{-1}]	Annual temperature [°C]	Annual precipitation [mm]
Achenkirch, Austria[a]	47° N 11° E	Spruce	125	Mull	Loam	19	5.7	7.0	0.077	1998–99	2.7–7.7	6.8–6.9	1691–1976
										2002–03	6.8–5.0	7.5–7.0	1747–1275
Copenhagen, Denmark[b]	55° N 12° E	Spruce	30	Moder	Loamy sand	6	3.7	3.7	0.061	1992	11.3	8.6	756
Glencorse, UK	55° N 3° W	Birch	21	Moder	Silty loam	18	4.8	4.8	0.070	2002–03	12.9–9.2	9.1–7.9	1183–840
Glencorse, UK	55° N 3° W	Sitka spruce	19	Moder	Silty loam	18	4.2	4.2	0.070	2002–03	12.9–9.2	9.1–7.9	1183–840
Harvard Forest, USA[c]	42° N 72° W	Hard-woods	79	Moder	Sandy loam	9	3.3	3.8	0.076	1989	2.2	7.4	1120
Höglwald, Germany[de]	48° N 11° E	Beech	87	Mull	Loam	19	4.0	3.7	0.051	1994–97	18.3–26.0	6.1–10.1	731–1041
Höglwald, Germany	48° N 11° E	Beech	110	Mull	Sandy loam	9	4.5	4.0	0.051	2002–03	26.3–14.3	9.1–8.9	1054–571
Höglwald, Germany[de]	48° N 11° E	Spruce	96	Moder	Loam	19	3.2	3.5	0.029	1994–97	18.3–26.0	6.1–10.1	731–1041
										2002–03	26.3–14.3	9.1–8.9	1054–571
Hyytiälä, Finland	61° N 24° E	Pine	41	Moder	Sandy loam	9	3.2	3.7	0.029	2002–03	0.09–0.1	4.2–4.1	535–644
Klausenleopoldsdorf, Austria[a]	48° N 16° E	Beech	55	Moder	Sandy clay loam	27	5.2	4.5	0.051	1996–97	9.5–12.9	8.6–8.9	763–1035
										2002–03	12.0–6.4	9.0–8.3	959–515
Matrafüred, Hungary	48° N 20° E	Spruce	35	Moder	Sandy loam	9	4.5	3.9	0.019	2002–03	13.3–7.3	9.0–8.1	809–678
Matrafüred, Hungary	48° N 20° E	Oak	63	Mull	Sandy loam	9	5.7	4.3	0.036	2002–03	8.7–6.5	9.0–8.1	809–678
Parco Ticino, Italy	45° N 9° E	Hard-woods	150	Mull	Loamy sand	6	4.1	4.2	0.067	2002–03	10.7–6.0	14.3–14.5	1066–602
Parco Ticino, Italy	45° N 9° E	Poplar	13	Moder	Sandy loam	9	5.8	5.9	0.010	2002–03	10.7–6.0	14.3–14.5	1066–602
San Rossore, Italy	43° N 10° E	Pine	38	Raw humus	Sand	3	5.0	5.8	0.007	2002–03	5.5–3.7	14.4–14.7	1101–742
Schottenwald, Austria[f]	48° N 16° E	Beech	135	Moder	Silty loam	18	5.0	4.2	0.068	1996–97	25.1–34.1	9.4–9.7	718–973
										2002–03	33.6–16.3	10.3–10.0	959–467
Sorø, Denmark	55° N 12° E	Beech	80	Moder	Loamy sand	9	4.3	4.5	0.040	2002–03	45.6–23.9	8.8–8.6	1013–532
Speulderbos, Netherlands	52° N 5° E	Douglas fir	43	Moder	Sand	3	3.7	3.7	0.090	2002–03	56.7–37.6	10.6–10.2	924–613
Wildbahn, Germany[g]	53° N	Pine	65	Moder	Loamy sand	6	3.3	3.6	0.035	1997	12.3	8.3	616

[a] Data of Forstliche Bundesversuchsanstalt, Vienna, [b] Ambus and Christensen (1995), [c] Bowden et al. (1991), [d] Göttlein and Kreutzer (1991), [e] Papen and Butterbach-Bahl (1999), [f] Jandl et al. (1997), [g] Böß (2000).

jrc.it/). The SGDBE is the resulting product of a collaborative project involving the European Union and neighbouring countries. It is a simplified representation of the diversity and spatial variability of the soil coverage (CEC, 1985). The SGDBE provides typological information according to so-called Soil Typological Units (STU; N: 5306). The STU are grouped into Soil Mapping Units (SMU; N: 1650) to form soil associations (for details see CEC, 1985). The STU attribute data contain information about specific soil properties, such as textural class, humus type, water regime, etc. Additionally data for soil carbon and pH for the organic layer and the mineral soil, stone content on SMU level were needed. Therefore preliminary results from the ongoing research project (CarboInvent) were used (aggregated soil C data for organic layer and mineral soil). Regarding these data it has to be noted that the SOC content may not yet represent the final level of quality. For example, data for the continental Eastern part of Europe (e.g. Poland) appear to be too high, and in the case of the mountainous areas such as the Alps, no stratification was found although local data clearly indicate elevated SOC content (R. Baritz, personal information). To aggregate the STU- and SMU-based attributes on the scale of the EMEP raster an up-scaling strategy was applied (Fig. 1). At first the soil properties as derived from the STU were scaled to the SMU level, thereby considering the relative area covered. Secondly, a weighted average value was calculated for the individual soil parameters for each EMEP grid cell, i.e. for clay content, humus type, forest floor and mineral soil pH, stone content and organic carbon mass in the forest floor and mineral soil. Additionally the maximum and minimum value of each soil parameter was recorded for each EMEP grid cell to retrieve the range for a sensitivity analysis with a maximum and a minimum scenario (see Sect. 2.5) (Fig. 1). Even though forest information for Belarus and Moldavia was available, these countries were excluded from the calculations since details about soil properties were not available.

Figure 2 shows the distribution of forest areas (Fig. 2a) and of selected soil properties (0–0.2 m soil depth: SOC, clay content, soil pH) in Europe as derived from CORINE/PELCOM land cover data sets and the SGDBE dataset on soil properties. The maps show that soils rich in organic carbon (SOC) in the mineral soil (>75 t C ha^{-1}) predominate in Northern Europe including the UK and Ireland (Fig. 2b), whereas heavily textured soils (clay content $>20\%$) are often found in the Mediterranean and the Balkan region (Fig. 2c). Predominantly acidic soils with a low base saturation are reported for large parts of Sweden and Finland, but also for the Northern parts of the UK (Fig. 2d).

2.3.2 Forest distribution and forest stand information

Information about the distribution of forest types across Europe has recently been published by Köble and Seufert (2001). They adopted the spatial distribution of forest area for most parts of Europe from the CORINE land cover data set (CEC, 1994) or from the Pan-European Land Cover Mapping project "PELCOM" (Mücher, 2000). In addition Köble and Seufert (2001) used tree species information from the measurement network of the transnational survey (ICP Forest

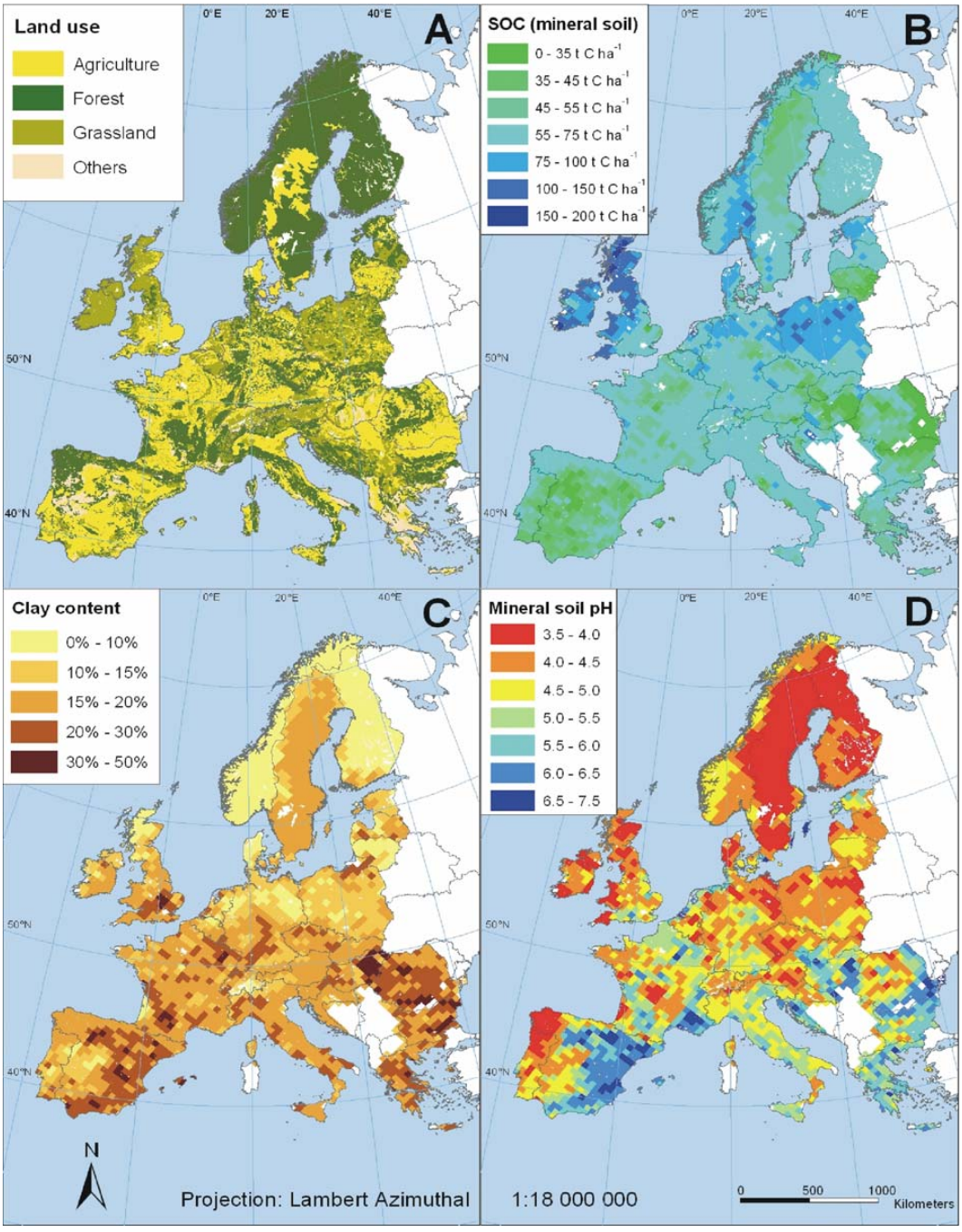

Fig. 2. Distribution of land cover types and selected soil properties across Europe. Data were derived either from CORINE or PELCOM land cover data sets or from the Soil Geographical Data Base of Europe. Note that soil property information is only valid for forest soils.

Level I) of forest condition in Europe (UN-ECE, 1998) to retrieve maps of forest type and tree species distribution on a 1 km×1 km raster format for 30 European countries. Since the PnET-N-DNDC model is currently only parameterized for the simulation of 12 forest types (see Sect. 2.1), we grouped some forest types together in order to simulate most of the forested areas in Europe. This means, that e.g. the forest types alder, ash, elm, poplar and willow were simulated with the parameterization for hardwoods. However, some forest types such as *Juniperus spec.* dominated forests were excluded from our simulations a) since such forests cover only small areas in Europe (approx. 3.5%) and b) to reduce the parameterization and computation complexity. The forest area considered in our simulations was 1 410 477 km^2,

which is in accordance with official national reports (Köble and Seufert, 2001). As there was no information about forest age available we assumed an average age of 60 years for all forest types. Forest areas in the countries of Albania, Serbia and Montenegro, Macedonia and Bosnia-Herzegovina were excluded from the simulations since no detailed forest information was available.

2.3.3 Climate and N deposition

Simulation runs were performed meteorology for the three years 1990, 1995 and 2000. Meteorological data in daily resolution was provided from the inputs of the EMEP MSC-W oxidant model (Sandnes-Lenschow et al., 2000; Simpson et al., 2003) including information about average temperature, and sum precipitation as well as photosynthetically active radiation (PAR). Figure 3 shows a map of the regional distribution of mean annual temperature and sum of annual precipitation across Europe for the year 2000 and relative differences of these parameters in 1990 as compared to the year 2000. The maps show a typical South-North gradient in temperature and reveal that e.g. in the year 2000 mean annual temperature was >10% higher in Central Europe and approx. 5% lower in Spain and Central Europe as compared to the year 1990. The variation in precipitation between the years 1990 and 2000 was pronounced and in many regions in Europe such as Central Finland, Southern UK or Portugal received >25% precipitation in 1990 than in the year 2000 (Fig. 3).

Additionally, the EMEP MSC-W model was used to calculate annual data on atmospheric N deposition (dry and wet) for each EMEP grid cell. Emissions of all pollutants were set to those of the year 2000 (Vestreng et al., 2004), whereas meteorology scenarios were taken from the years 1990, 1995 and 2000 in order to asses meteorological variability. This EMEP model's simulations of concentrations and deposition have been extensively evaluated elsewhere (e.g. Fagerli et al., 2003) and for forests in particular by Westling et al. (2005). Since the PnET-N-DNDC model does not allow consideration of dry deposition of N to forests, we only used the wet deposition values (Fig. 4). The map shows that wet deposition of N with values $>13\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$ are especially observed for the Benelux countries and neighbouring North Germany and for parts of South Germany and Northern Italy.

2.4 Coupling of the GIS database to the PnET-N-DNDC model

The forest, soil, and climate information was aggregated and linked to the EMEP raster. An individual identification number was assigned to each of the 2527 grid cells of the simulated area. By calling the ID numbers the PnET-N-DNDC model automatically received the individual initialisation and driving parameters of each grid cell. The number of model runs per grid cell depended on the number of forest types found within this cell. For example, three different forest types in one grid cell resulted in three model runs. The results of the individual model runs for one grid cell were weighted depending on the total area of each forest type in the respective grid cell.

2.5 Uncertainty analysis (Monte Carlo, MSF)

The focus of the uncertainty analysis was the assessment of the uncertainty of simulation results caused by the necessary generalisations within the GIS database on e.g. soil and vegetation properties. By using the EMEP grid with cells of $50\,\mathrm{km}\times50\,\mathrm{km}$ across Europe it was assumed that soil properties within a grid cell were uniform. However, this is of course not the case, since soil properties (e.g. pH, soil organic carbon (SOC) content) are highly variable in space. To assess the effect of sub grid cell variability in soil properties on simulated N trace gas emissions from forest soils the Most Sensitive Factor (MSF) method (Li et al., 2004) as well as a Monte Carlo approach was used with the same set of parameters. N trace gas fluxes simulated with the PnET-N-DNDC and also with the DNDC have been shown to be very sensitive to changes in soil texture, pH and SOC (Stange et al., 2000; Butterbach-Bahl et al., 2001; Li et al., 2004). After extensive sensitivity studies of the soil database of Europe we found out that there were general trends regarding the relationship between N_2O and NO emissions and the soil factors. For example, the modelled N_2O and NO emissions usually increase along with an increase in SOC content and clay fraction as well as a decrease in pH. These model reactions are in accordance with a series of results from field and laboratory observations (Li et al., 2005). The MSF method uses the generalized relationships between individual soil factors and the magnitude of N trace gas emissions by grouping a series of soil factors for which minimum and maximum values are available in such a way that N trace gas emissions are either maximized or minimized. This means that PnET-N-DNDC automatically selected the minimum organic matter mass in the forest floor and mineral soil, maximum pH in the forest floor and mineral soil, maximum stone content and minimum clay content to form a scenario which was assumed to produce a low value of N_2O and NO flux for this grid cell and the model then selected the maximum organic matter mass and minimum pH in the forest floor and mineral soil, minimum stone content and maximum clay content to form another scenario, which was assumed to produce a high value of N_2O and NO flux for the grid cell. Thus PnET-N-DNDC ran twice with the two scenarios for each grid cell to produce an upper and a lower boundary of expected N_2O and NO emission rates (three times if the average scenario is included). The calculated N trace gas emission range was assumed to be wide enough to cover the real flux with a high probability. To verify the MSF method, we also implemented a Monte Carlo routine into the PnET-N-DNDC. This allowed us to directly quantify the uncertainties derived by soil heterogeneity

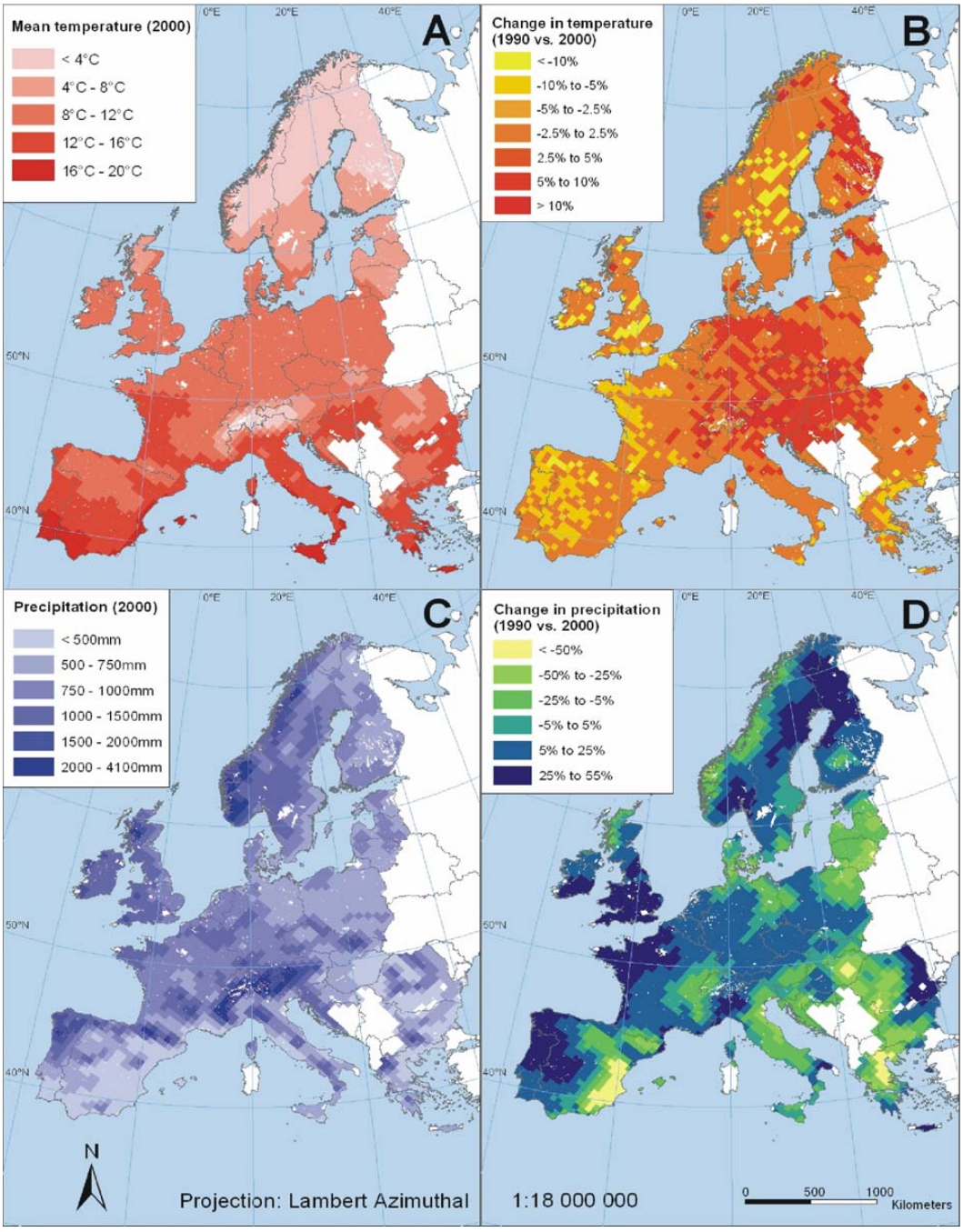

Fig. 3. Regional distribution of mean annual air temperature **(A)** and sum of annual precipitation **(C)** in the year 2000 across Europe. Panels **(B)** and **(D)** show the relative change in temperature or precipitation between the years 2000 and 1990. Meteorological data were provided by the Norwegian Meteorological Institute.

of individual EMEP grid cells (for details see Li et al., 2004). When PnET-N-DNDC ran in the Monte Carlo mode, the observed range for each soil factor in a grid cell was divided into eight intervals. For example, if the pH in a grid cell ranged from 3.5 to 5.6 the Monte Carlo approach would run with the pH values 3.5, 3.8, 4.1, 4.4 . . . , 5.6. PnET-N-DNDC selected randomly an interval of each of the six soil properties (clay content, organic mass in mineral soil and forest floor, forest floor and mineral soil pH, and stone content) to form a scenario. The process was repeated 5000 times so that 5000 N_2O and NO emission estimates were calculated for one grid cell. The results were then compared with the results

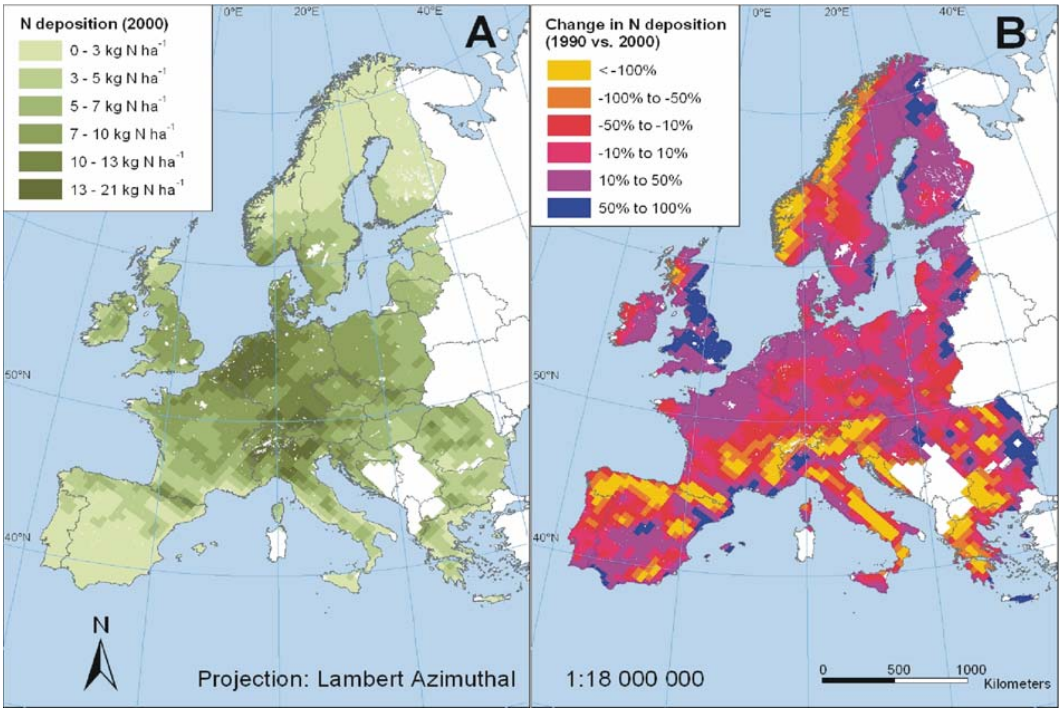

Fig. 4. Annual values of wet deposition of N in Europe in the year 2000 **(A)** and changes in wet deposition of N using 1990 meteorology versus 2000 meteorology (year 2000 emissions) **(B)**. Calculations with EMEP MSC-W Photo-oxidant model.

of the MSF method. For the Monte Carlo approach we selected randomly 50 EMEP grid cells across Europe and compared the results of the frequency distribution of N_2O and NO emissions with the ranges of N_2O and NO emissions as derived from the MSF method. The comparison of the MSF method with the Monte Carlo approach showed that the range of NO emissions calculated with the MSF method covered in average more than 79% of the variability in N trace gas emissions calculated with the Monte Carlo approach. However, this value was remarkably lower with regard to N_2O. The maximum N_2O emissions calculated with the MSF method were in average approx. 50% lower compared to the emissions using the Monte Carlo approach. The minimum N_2O emissions calculated with the MSF method were in average two fold higher than the N_2O emissions calculated with the Monte Carlo method. However, since the lower boundary of N_2O emissions ranged between 0.1 to 0.2 kg N ha^{-1} yr^{-1} this difference can be neglected for the purpose of this study. Due to the underestimation of maximum N trace gas emissions the uncertainty estimates with the MSF method are not fully satisfactory, but represent at present the best uncertainty estimate we can achieve. The full application of the Monte Carlo method (or of comparable methods) to all grid cells would be the favourable method to estimate prediction uncertainties. But for this a further optimisation of the model code with regard to the reduction of computation time is required.

Due to the lack of an uncertainty range for regional N deposition, the effect of this on N trace gas fluxes was not included in the uncertainty analysis. However, Fig. 5 shows on a site scale that variations in N deposition will significantly feedback on soil NO and N_2O fluxes even in one year simulation runs. I.e. increases in N deposition by e.g. 50% would increase simulated N_2O and NO fluxes at our 19 test sites by approx. 38% or 21% (Fig. 5).

3 Results

3.1 Model testing

The model was applied to the different field sites with identical and fixed internal parameter settings for microbial C and N turnover processes. Figure 6 shows daily simulation results for NO and N_2O emissions for the sites Höglwald (spruce, Germany), Sorø (beech, Denmark), Hyytiälä (Pine, Finland) and Glencorse (Sitka spruce, Scotland) as compared to observed N trace gas emissions. For the Höglwald spruce site simulated N_2O emissions were in average 23% higher than the observed emissions. Overestimation of N_2O emissions mainly occurred during the first of half of the year, whereas in autumn N_2O emissions tended to be underestimated in average by approx. 10–15%. The model captured the period with peak emissions in summer, but predicted the peak emission a few days earlier than observed in the field

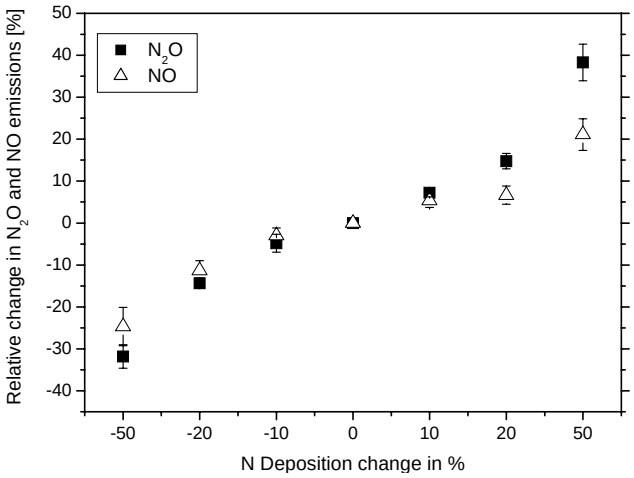

Fig. 5. Effect of changes in N deposition (−50%–+50%) on simulated N_2O and NO emissions at the 19 test sites. Given are mean values ±SE.

(Fig. 6). Simulated NO emissions for the Höglwald spruce site were in good agreement with field observations throughout the year with respect to seasonality and magnitude of fluxes. For most periods, except for three 1–2 week long periods in June, August and October, simulated results deviated only within 10–20% from observed NO emissions. However, in the short periods mentioned emissions were overestimated by a factor of two. The simulated seasonality of NO and N_2O emissions at the Höglwald site matched the seasonality as observed in the field, e.g. high NO emissions during summer versus comparably low emissions in the winter period (Fig. 6). Also the differences in magnitude of NO and N_2O emissions between both sites, which are mainly due to differences in litter quality and soil pH, were well reproduced by the model. The model also realistically predicted differences in the magnitude of N trace gas emissions for different field sites across Europe, i.e. low N_2O emissions in Hyytiälä and Glencorse, and slightly elevated N_2O emissions at Sorø. However, especially for the beech site at Sorø simulated emissions for the first few months of 2002 tended to be higher than field observations. This was mainly due to a simulation of elevated N_2O emissions during freezing-thawing events by the PnET-N-DNDC model, which were not confirmed during field measurements. However, for this period field measurements also revealed a pronounced spatial variability of N_2O emissions (Fig. 6). N_2O and sporadically performed NO emission measurements at the Hyytiälä site showed that N trace gas emissions are close to zero. A comparable result was also delivered by the PnET-N-DNDC model. For the Glencorse site the model captured the temporal variation in NO emissions during the summer period of 2002, but failed to predict the increase in NO emissions from the end of October onwards (Fig. 6). For the period during which NO field measurements had been performed the model underestimated NO emissions by approx. 30% (field mean: 4.9 g N ha^{-1} day^{-1}; simulation: 3.4 g N ha^{-1} day^{-1}).

Figures 7 and 8 and Tables 2 and 3 summarize results of model testing for all 19 field sites for which data from N trace gas emission measurements were available. The graph shows that the model was capable of capturing observed differences between high and low emitting sites, based on general information on soil and vegetation properties and by considering the local meteorological conditions. The relative variation between observed and simulated N_2O emissions was higher for sites with N trace gas emissions <3 g N ha^{-1} day^{-1} as compared to sites with N trace gas emissions >5 g N ha^{-1} day^{-1}. The linear regression of all simulated and observed mean N_2O emission rates resulted in r^2=0.68 (Fig. 7). On average over all test sites the model underestimated emissions by 24% (f(x)=0.76x). For NO the r^2 value was 0.78 (Fig. 8). Like in the case of N_2O the model also tended to underestimate NO emissions at the test sites by on average 27% (f(x)=0.73x). Given the wide range of complex processes involved in mediating soil N emissions, these results are very encouraging. These results of model testing for a wide variety of forest ecosystems across Europe (see also details in Tables 2 and 3) provided solid basis for the application of the PnET-N-DNDC model on a regional scale.

3.2 N_2O emissions from European forest soils

Figure 9a shows modelled N_2O emissions from forest soils across Europe resulting from the regional application of the GIS-coupled PnET-N-DNDC model. For the year 2000 simulated N_2O emissions from European forest soils ranged between 0.01 to 2.9 kg N ha^{-1} yr^{-1}. N_2O emissions >2.5 kg N ha^{-1} yr^{-1} were predicted for some forest ecosystems in the Netherlands. Simulated annual N_2O emissions for wide areas of Central Europe, West Spain, Slovakia and Romania were also found to be >1.0 kg N ha^{-1} yr^{-1}. Furthermore, high N_2O emissions were also predicted for soils with high amounts of organic carbon content in the forest floor in Southwest Finland and in the Northern parts of Sweden (1.0 to 1.8 kg N ha^{-1} yr^{-1}). Intermediate emissions in the range of 0.75 to 1.5 kg N ha^{-1} yr^{-1} were simulated for large parts of Poland and the Baltic states, whereas N_2O emissions <0.5 kg N ha^{-1} yr^{-1} were calculated for most Mediterranean and maritime regions including France and the UK, Ireland and Norway as well as large parts of central and northern Finland (Fig. 9a). The average N_2O emission of all forest sites across Europe calculated for the year 2000 was 0.58 kg N ha^{-1} yr^{-1} (Table 4). This average N_2O emission value changed only slightly when the model was initialized with the meteorological drivers for the years 1990 and 1995. For 1990 an average value of 0.62 kg N ha^{-1} yr^{-1} was calculated, whereas for 1995 the mean N_2O emission was 0.55 kg N ha^{-1} yr^{-1} (Table 4). However, on a regional scale the magnitude of N_2O emissions between individual years can change significantly, as shown in Fig. 9b. The

Table 2. Compilation of results for N_2O emissions from the different field sites as derived from model runs with PnET-N-DNDC and from field measurements.

Site-No.	Site-name	Year	Measuring days	Mean N_2O emission Measured	Simulated	Model performance R^2	RMSE
				$g\,N\,ha^{-1}\,day^{-1}$			$g\,N\,ha^{-1}\,day^{-1}$
1	Achenkirch	1998	18	4.0±0.6	2.3±0.4	0.48	2.55
2	Achenkirch	1999	19	2.4±0.5	2.3±0.4	0.37	1.74
3	Achenkirch	2002	122	1.1±0.1	2.9±0.1	0.00	2.43
4	Achenkirch	2003	194	0.8±0.1	3.0±0.1	0.11	2.88
5	Copenhagen	1992	17	2.3±0.5	0.5±0.1	0.02	2.88
8	Glencorse Sitka	2002	87	0.5±0.1	1.3±0.0	0.00	1.06
9	Glencorse Sitka	2003	122	0.2±0.0	0.6±0.2	0.00	0.70
10	Höglwald Beech	1994	105	2.7±0.1	7.6±0.5	0.47	6.39
11	Höglwald Beech	1995	341	10.0±0.5	11.3±0.4	0.11	9.19
12	Höglwald Beech	1996	307	18.2±1.0	15.7±0.8	0.42	13.63
13	Höglwald Beech	1997	348	5.5±0.3	9.6±0.3	0.09	7.91
14	Höglwald Beech	2002	103	2.3±0.2	1.6±0.2	0.20	2.87
15	Höglwald Beech	2003	158	3.9±0.3	7.6±0.4	0.22	5.74
16	Höglwald Spruce	1994	345	1.1±0.0	2.4±0.1	0.14	1.74
17	Höglwald Spruce	1995	359	2.1±0.1	2.8±0.9	0.01	1.92
18	Höglwald Spruce	1996	343	8.5±0.8	4.6±0.2	0.48	13.83
19	Höglwald Spruce	1997	346	1.8±0.2	2.3±0.1	0.44	2.38
20	Höglwald Spruce	2002	343	1.9±0.1	2.4±0.1	0.25	1.62
21	Höglwald Spruce	2003	340	1.0±0.1	2.0±0.5	0.27	1.41
22	Hyytiälä	2002	17	0.1±0.1	0.5±0.1	0.15	0.74
23	Hyytiälä	2003	11	0.2±0.0	0.2±0.1	0.26	0.27
24	Klausenleopoldsdorf	1996	16	6.6±1.0	3.8±0.4	0.41	4.20
25	Klausenleopoldsdorf	1997	21	5.2±1.1	3.1±0.5	0.69	4.08
26	Klausenleopoldsdorf	2002	119	1.9±0.2	3.1±0.2	0.00	2.68
27	Klausenleopoldsdorf	2003	178	1.5±0.1	3.8±0.1	0.04	3.11
28	Matrafüred Oak	2002	10	4.0±1.4	1.4±0.2	0.60	4.64
29	Matrafüred Oak	2003	27	6.6±0.8	5.3±0.5	0.00	4.91
30	Matrafüred Spruce	2002	11	4.8±1.5	0.4±0.2	0.28	6.71
31	Matrafüred Spruce	2003	28	4.8±0.7	1.3±0.1	0.29	4.97
32	P. Ticino BoscoNegri	2002	57	1.4±0.3	1.6±0.2	0.14	2.15
33	P. Ticino BoscoNegri	2003	177	0.5±0.0	1.2±0.1	0.10	1.02
34	P. Ticino Poplar	2002	48	0.5±0.2	1.0±0.1	0.04	1.38
35	P. Ticino Poplar	2003	17	0.7±0.2	0.6±0.1	0.11	0.92
36	San Rossore	2002	65	0.9±0.1	4.1±0.1	0.00	3.52
37	San Rossore	2003	183	0.2±0.0	2.1±0.1	0.14	2.21
38	Schottenwald	1996	16	15.9±2.8	9.0±1.2	0.19	11.85
39	Schottenwald	1997	21	13.4±3.3	8.3±1.1	0.66	12.43
40	Schottenwald	2002	162	11.4±0.9	8.0±0.3	0.05	11.98
41	Schottenwald	2003	252	9.8±0.5	6.4±0.2	0.00	9.29
42	Sorø	2002	19	2.4±0.5	1.7±0.2	0.04	2.18
43	Sorø	2003	156	1.5±0.1	2.5±0.1	0.21	1.70
44	Speulderbos	2002	107	0.8±0.1	1.8±0.1	0.28	1.19
45	Speulderbos	2003	216	0.4±0.0	2.7±0.1	0.32	2.51
46	Harvard Forest	1989	10	0.1±0.2	0.2±0.1	0.06	0.60
47	Wildbahn	1997	10	1.7±0.2	1.2±0.2	0.63	0.70
	Total		5971	4.2±0.1	4.7±0.1	0.51	6.50

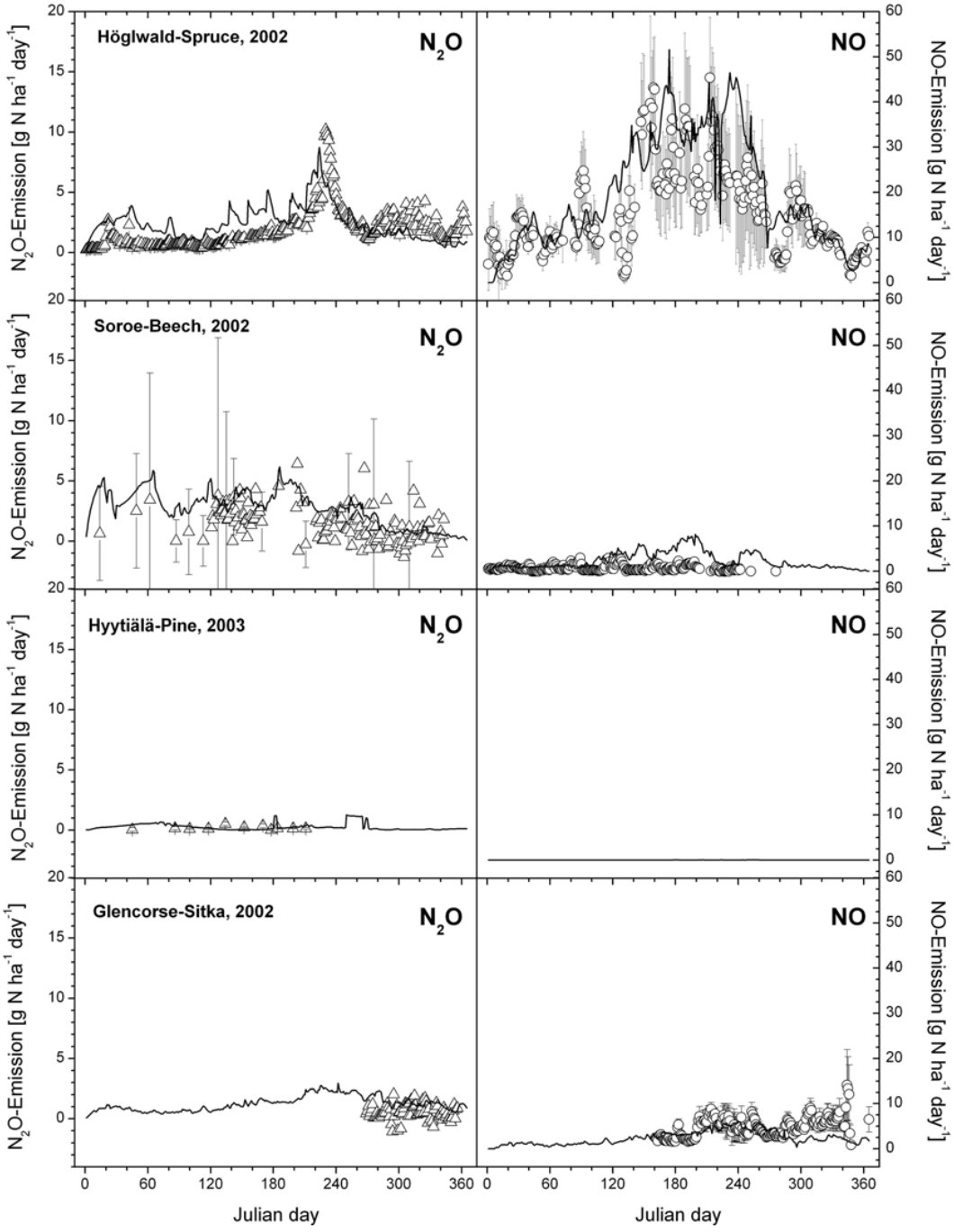

Fig. 6. Comparison of measured (triangles/circles) and simulated (solid line) N_2O and NO emissions for the forest sites Höglwald (spruce, Germany), Sorø (beech, Denmark), Hyytiälä (Pine, Finland) and Glencorse (Sitka spruce, Scotland). No measuring points are shown for NO fluxes at Hyytiälä since fluxes were always below the detection limit.

map shows that e.g. in the year 2000 the N_2O emissions from forest soils in Southern Sweden were >40% higher than in the year 1990, whereas in other areas such as the Mediterranean region N_2O emissions were 10 to >40% lower. Total N_2O emissions from forest soils across Europe for the years 1990, 1995 and 2000 were in a range of 77.6 to 86.8 kt N year^{-1} (Table 4). Due to their large forested areas Sweden and Finland contributed most to the total N_2O emissions (11.9 and 10.3 kt N yr^{-1}). However, on a per hectare basis forests in the Netherlands (1.26 kg N ha^{-1} yr^{-1}) and Romania (0.96 kg N ha^{-1} yr^{-1}) were found to be the strongest emitters (Table 4).

Table 3. Compilation of results for NO emissions from the different field sites as derived from model runs with PnET-N-DNDC and from field measurements.

Site-No.	Site-name	Year	Measuring days	Mean NO emission Measured	Mean NO emission Simulated	Model performance R^2	Model performance RMSE
				$g\,N\,ha^{-1}\,day^{-1}$			$g\,N\,ha^{-1}\,day^{-1}$
3	Achenkirch	2002	93	0.2±0.0	3.1±0.1	0.00	3.26
4	Achenkirch	2003	153	0.1±0.0	3.6±0.2	0.01	4.33
6	Glencorse Birch	2002	197	−0.1±0.1	1.6±0.1	0.00	2.03
7	Glencorse Birch	2003	176	1.0±0.1	0.7±0.1	0.23	0.99
8	Glencorse Sitka	2002	182	4.9±0.2	3.4±0.1	0.00	3.01
9	Glencorse Sitka	2003	176	7.6±0.4	1.4±0.1	0.11	7.70
10	Höglwald Beech	1994	104	2.1±0.1	3.9±0.2	0.05	2.84
11	Höglwald Beech	1995	334	6.2±0.2	5.5±0.3	0.23	4.73
12	Höglwald Beech	1996	327	7.5±0.4	4.3±0.2	0.12	7.37
13	Höglwald Beech	1997	337	9.8±0.4	6.1±0.2	0.24	7.51
14	Höglwald Beech	2002	134	2.7±0.1	2.3±0.2	0.08	2.28
15	Höglwald Beech	2003	176	6.9±0.6	3.3±0.2	0.22	8.52
16	Höglwald Spruce	1994	357	17.5±0.5	20.7±0.7	0.57	9.02
17	Höglwald Spruce	1995	332	23.6±0.7	19.3±0.7	0.45	11.51
18	Höglwald Spruce	1996	349	24.9±1.0	16.1±0.7	0.41	16.32
19	Höglwald Spruce	1997	359	19.4±0.6	17.7±0.6	0.38	10.25
20	Höglwald Spruce	2002	277	15.4±0.6	19.4±0.8	0.53	9.47
21	Höglwald Spruce	2003	209	32.2±1.9	18.9±0.8	0.54	24.29
27	Klausenleopoldsdorf	2003	63	0.2±0.0	2.4±0.2	0.22	2.63
30	Matrafüred Spruce	2002	6	0.2±0.1	0.1±0.1	0.14	0.25
31	Matrafüred Spruce	2003	13	0.5±0.2	1.6±0.3	0.15	1.47
40	Schottenwald	2002	125	3.5±0.1	7.0±0.4	0.09	5.25
41	Schottenwald	2003	132	5.6±0.3	4.4±0.3	0.02	0.02
43	Sorø	2003	231	0.8±0.0	2.7±0.1	0.03	12.26
44	Speulderbos	2002	112	15.4±1.2	9.1±0.4	0.30	15.62
45	Speulderbos	2003	229	20.4±1.0	9.7±0.4	0.51	0.38
47	Wildbahn	1997	5	2.7±0.1	2.6±0.2	0.03	2.63
	Total		5191	11.7±0.2	9.5±0.2	0.70	9.90

3.3 NO emissions from European forest soils

Figure 9c shows the modelled NO emissions from forest soils across Europe for the year 2000. As for N_2O, the highest NO emissions were simulated for forest soils in the Netherlands and neighbouring areas in Belgium and Germany. The maximum NO emission for a grid cell in this area was 7.0 $kg\,N\,ha^{-1}\,yr^{-1}$. For forest soils in most parts of Germany, Belgium, Poland and the Massif Central in France, simulated NO emissions were in a range of 1.0 to 3.0 $kg\,N\,ha^{-1}\,yr^{-1}$. Furthermore, elevated NO emissions of up to 3.0 $kg\,N\,ha^{-1}\,yr^{-1}$ were found for large areas of Sweden. This finding was mainly related to the low soil pH values usually found for forest soils in this region, causing a high NO production via chemo-denitrification in the model. Mostly low emissions of NO ($<0.5\,kg\,N\,ha^{-1}\,yr^{-1}$) were simulated for forest soils in Norway, most of Finland and the Mediterranean region (Fig. 9c). The average NO emission from forest soils across Europe in the year 2000 was calculated to be 0.7 $kg\,N\,ha^{-1}\,yr^{-1}$ and, thus, slightly higher than for N_2O (0.58 $kg\,N\,ha^{-1}\,yr^{-1}$). However, the average NO emission from forest soils varied only slightly between individual simulation years (1990: 0.7 $kg\,N\,ha^{-1}\,yr^{-1}$, 1995: 0.6 $kg\,N\,ha^{-1}\,yr^{-1}$) (Table 4). Total NO emission for all forests within the simulation area was 99.2 kt N in the year 2000 which was almost the same as in the year 1990 (98.3 kt N) and slightly lower than in the year 1995 with 84.9 kt N. The major contributors to total NO emissions from forest soils in Europe were Sweden and Germany (Table 4). The interannual variability in NO emissions due to changes in meteorological conditions was pronounced. Figure 9d shows relative changes in the NO emission strength of forest soils across Europe. The map shows that NO emissions were mostly higher in Central Europe and Northern Europe but lower in the Mediterranean Region for meteorology from the year 2000 as compared to using meteorology from the

Table 4. Average and total simulated N_2O and NO emissions from forest soils for individual European countries using meteorology for the years 1990, 1995 and 2000.

Country	Forested Area	1990				1995				2000			
		N_2O		NO		N_2O		NO		N_2O		NO	
	km^2	kg N ha^{-1} yr^{-1}	kt N yr^{-1}	kg N ha^{-1} yr^{-1}	kt N yr^{-1}	kg N ha^{-1} yr^{-1}	kt N yr^{-1}	kg N ha^{-1} yr^{-1}	kt N yr^{-1}	kg N ha^{-1} yr^{-1}	kt N yr^{-1}	kg N ha^{-1} yr^{-1}	kt N yr^{-1}
Andorra	232	0.70	1.6×10^{-2}	1.04	0.02	0.20	5.0×10^{-3}	0.28	6.6×10^{-3}	0.23	5.3×10^{-3}	0.36	8.4×10^{-3}
Austria	24 032	0.86	2.08	0.72	1.73	0.60	1.44	0.48	1.14	0.64	1.53	0.62	1.50
Belgium	7699	0.76	0.58	1.56	1.20	0.61	0.47	1.26	0.97	0.94	0.72	1.96	1.51
Bulgaria	28 494	0.95	2.71	0.80	2.27	0.58	1.64	0.64	1.82	0.70	1.99	0.56	1.61
Croatia	12 574	0.86	1.08	0.93	1.16	0.58	0.73	0.72	0.91	0.60	0.76	0.69	0.87
Czech. Republic	20 406	0.60	1.23	1.05	2.13	0.52	1.07	0.80	1.63	0.68	1.38	1.09	2.23
Denmark	18 608	0.58	1.08	0.85	1.58	0.48	0.90	0.68	1.26	0.70	1.30	0.94	1.75
Estonia	18 341	0.51	0.93	0.49	0.90	0.70	1.28	0.60	1.11	0.57	1.05	0.72	1.32
Finland	159 676	0.77	12.35	0.56	8.88	0.74	11.79	0.64	10.25	0.65	10.30	0.58	9.32
France	132 395	0.57	7.60	0.67	8.84	0.46	6.07	0.53	7.07	0.55	7.26	0.72	9.58
Germany	117 848	0.72	8.48	1.16	13.70	0.58	6.84	0.93	10.93	0.77	9.09	1.30	15.28
Gibraltar	0.43	0.55	2.4×10^{-5}	0.07	3.0×10^{-6}	0.54	2.3×10^{-5}	0.03	1.3×10^{-6}	0.55	2.4×10^{-6}	0.05	2.2×10^{-6}
Greece	30 676	0.68	2.09	0.45	1.38	0.53	1.64	0.36	1.11	0.55	1.68	0.35	1.07
Hungary	21 181	0.57	1.21	0.49	1.03	0.57	1.22	0.49	1.03	0.75	1.59	0.49	1.04
Irish Republic	5523	0.17	0.09	0.34	0.19	0.14	0.08	0.25	0.14	0.18	0.10	0.37	0.20
Italy	59 834	0.91	5.43	0.78	4.68	0.60	3.57	0.64	3.84	0.59	3.56	0.63	3.78
Latvia	28 229	0.42	1.19	0.72	2.04	0.74	2.08	0.74	2.08	0.73	2.07	0.79	2.24
Liechtenstein	89	0.87	7.7×10^{-3}	0.39	3.4×10^{-3}	0.97	8.6×10^{-3}	0.23	2.1×10^{-3}	0.77	6.8×10^{-3}	0.27	2.4×10^{-3}
Lithuania	18 843	0.63	0.55	0.43	0.82	0.43	0.81	0.42	0.79	0.53	1.01	0.52	0.98
Luxembourg	1032	0.29	0.07	0.98	0.10	0.53	0.05	0.84	0.09	0.64	0.07	1.09	0.11
Monaco	0.21	0.21	4.3×10^{-6}	0.12	2.5×10^{-6}	0.26	5.4×10^{-6}	0.22	4.7×10^{-6}	0.26	5.5×10^{-6}	0.16	3.3×10^{-6}
Netherlands	8271	0.99	0.82	2.39	1.98	0.77	0.64	1.84	1.52	1.26	1.04	3.01	2.49
Norway	159 482	0.19	2.99	0.05	0.86	0.23	3.62	0.04	0.63	0.17	2.69	0.05	0.72
Poland	76 358	0.52	4.00	1.10	8.37	0.50	3.79	0.87	6.65	0.59	4.53	1.06	8.06
Portugal	32 713	0.39	1.26	0.20	0.66	0.38	1.26	0.13	0.42	0.36	1.18	0.10	0.33
Romania	41 284	0.85	3.50	0.76	3.13	0.60	2.49	0.68	2.80	0.96	3.95	0.70	2.88
San Marino	0.35	0.27	9.4×10^{-6}	0.31	1.1×10^{-5}	0.28	9.8×10^{-6}	0.31	1.1×10^{-5}	0.30	1.0×10^{-5}	0.26	9.1×10^{-6}
Slovakia	9162	0.70	0.64	0.83	0.76	0.74	0.67	0.67	0.61	0.94	0.86	0.87	0.80
Slovenia	7881	1.14	0.90	1.30	1.02	0.67	0.52	0.87	0.69	0.82	0.65	1.10	0.87
Spain	138 484	0.65	8.97	0.37	5.19	0.60	8.36	0.29	3.98	0.57	7.94	0.27	3.80
Sweden	196 236	0.68	13.43	1.14	22.33	0.68	13.34	1.03	20.20	0.61	11.94	1.19	23.27
Switzerland	12 407	0.81	1.01	0.54	0.67	0.54	0.67	0.33	0.41	0.60	0.75	0.47	0.58
United Kingdom	22 481	0.22	0.49	0.33	0.75	0.24	0.53	0.35	0.80	0.27	0.60	0.44	0.99
Sum	1 410 477		86.78		98.37		77.59		84.89		81.59		99.20
Average		0.62		0.70		0.55		0.60		0.58		0.70	

year 1990.

Figure 10 shows the calculated NO: N_2O emission ratio for EU forests. The figure shows that for Central Europe and most of Sweden simulated NO emissions are dominating over N_2O emissions, whereas in other parts of Scandinavia, UK and South/ South East Europe N_2O emissions dominate over NO emissions. In our model simulation the NO: N_2O ratio was significantly correlated with the soil parameters SOC (r=0.129), mineral soil pH (r=−0.360) and atmospheric N deposition (r=0.356). However, all these correlations are rather weak.

3.4 Uncertainty estimates

Using the Most Sensitive Factor (MSF) method, which was re-evaluated prior to its use with a Monte Carlo approach (see Sect. 2.5), we calculated for each grid cell a minimum and a maximum scenario for the emission strength of N_2O and NO from forest soils. The cumulative sum for the minimum and the maximum scenarios as well as the results from the "average" scenario for the years 1990, 1995 and 2000 are given in Table 5. The table shows that e.g. in the year 2000 model initialisation with mean values for all grid cells resulted in a total N_2O emission from forest soils of 81.6 kt N yr^{-1}. The range of uncertainty for this respective year was 50.7 to 96.9 kt N yr^{-1}, which is equivalent to a relative range of −40 to +16% as compared to the average scenario. The uncertainty for the prediction of NO emissions was significantly higher as compared to N_2O. Here, we calculated a total NO emission of 99.2 kt N yr^{-1} (year 2000) for the average scenario with a range of uncertainty of 44.3 to 254.0 kt N yr^{-1}. The relative range as compared to the mean value thus equals to −66 to +156%. These remarkable differences in the uncertainty ranges between modelled N_2O and NO emissions were found to be mainly due to the pH sensitivity of NO production via chemo-denitrification.

Table 5. Range of uncertainty for calculated total N_2O and NO emissions from forest soils across Europe. The uncertainty ranges were calculated by use of the Most Sensitive Factor method (Li et al., 2004).

Meteorology Year	N_2O emission (kt N yr^{-1}) Minimum Scenario	Average Scenario	Maximum Scenario
1990	55.1	86.8	100.3
1995	50.0	77.6	96.2
2000	50.7	81.6	96.9
Meteorology Year	**NO emission (kt N yr^{-1}) Minimum Scenario**	**Average Scenario**	**Maximum Scenario**
1990	44.6	98.4	247.8
1995	38.3	84.9	220.2
2000	44.4	99.2	254.0

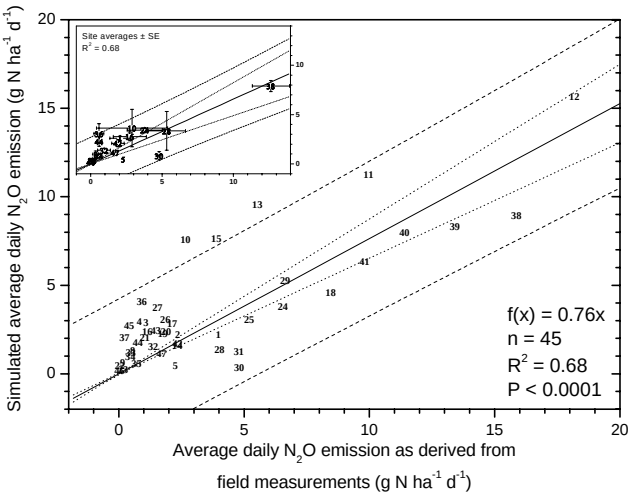

Fig. 7. Comparison of measured and simulated average daily N_2O emissions for different forest sites across Europe. The numbers refer to the individual sites and observation years as listed in Table 2. Regression line (solid line), 95% confidence limits for the slope (short dashes), and 95% prediction limits (long dashes) are provided.

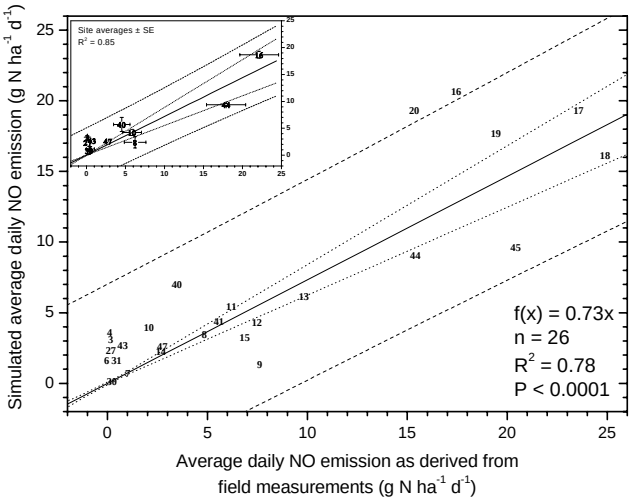

Fig. 8. Comparison of measured and simulated average daily NO emissions for different forest sites across Europe. The numbers refer to the individual sites and observation years as listed in Table 3. Regression line (solid line), 95% confidence limits for the slope (short dashes), and 95% prediction limits (long dashes) are provided.

4 Discussion

GIS coupled biogeochemical models have recently been used in a number of studies for the calculation of regional or global inventories of N_2O and NO emissions from soils (e.g. Potter et al., 1996; Davidson et al., 1998; Butterbach-Bahl et al., 2001, 2004; Brown et al., 2002; Li et al., 2001, 2004). This approach has several advantages as compared to a pure statistical approach where single or a series of field measurements at one or several site(s) are extrapolated to larger regions. Advanced biogeochemical models summarize our current understanding of environmental factors which are affecting the magnitude of trace gas emissions from soils such as meteorological conditions, soil properties (pH, SOC, texture) or – if turning especially to agricultural soils – management practices. In a series of studies all these factors have been shown to largely influence the production, consumption and emission of N trace gases (e.g. Barnard and Leadley, 2005; Conrad, 2002). By using biogeochemical models in inventory studies one assumes that the complexity of processes involved in N trace gas emissions and interacting factors, which is the main reason for the widely observed spatial and temporal variability in emissions, will at least partly be mimicked by the models (Li et al., 2000, 2001). In this paper we provided evidence that the PnET-N-DNDC model is indeed a powerful and reliable tool to simulate N trace gas

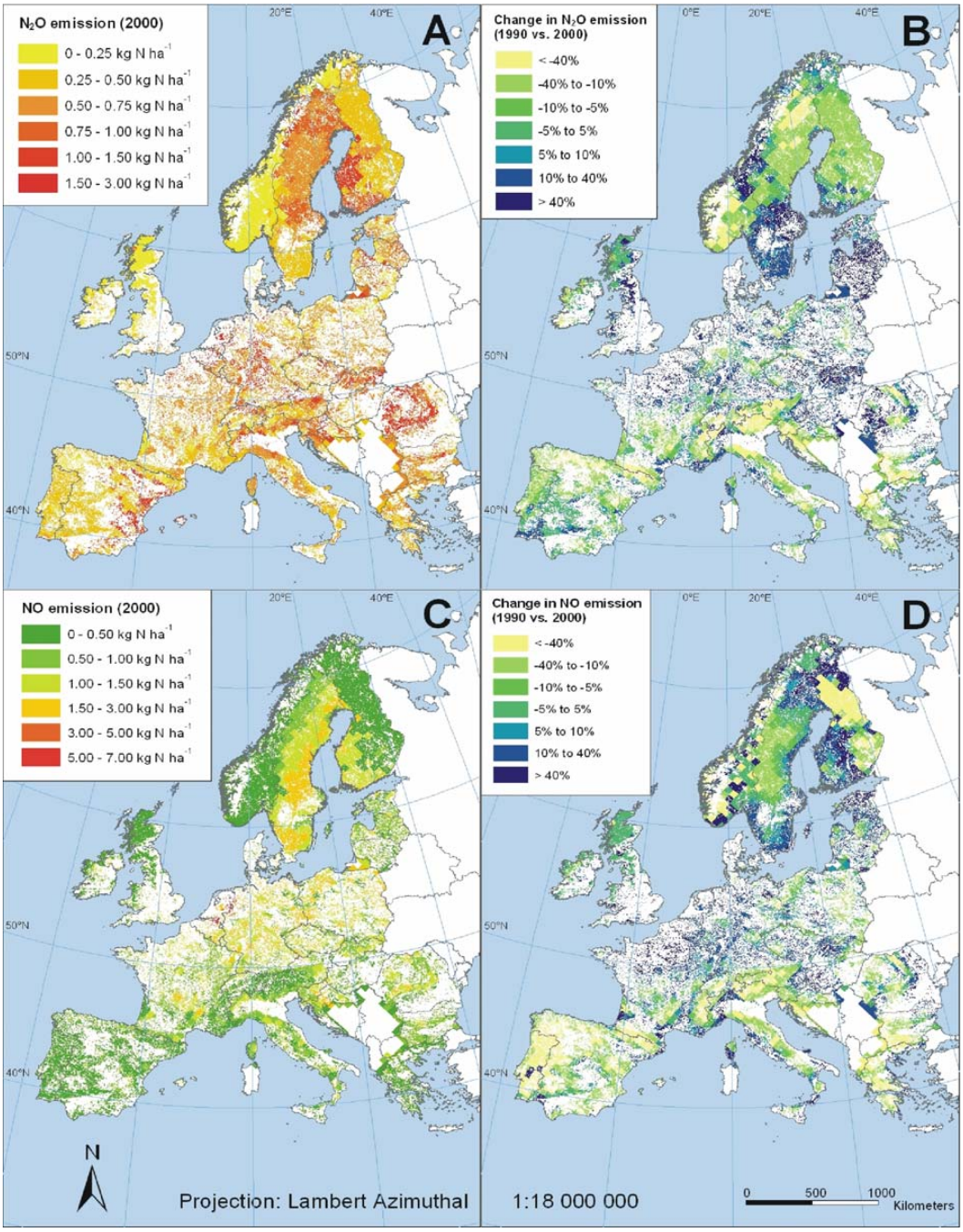

Fig. 9. Regional distribution of annual N_2O **(A)** and NO emissions **(C)** from forest soils in Europe (in kg N ha^{-1} yr^{-1}). Panels **(B)** and **(D)** show relative changes in the N_2O (B) or NO (D) emission strength if meteorological data for the year 2000 were exchanged with those of the year 1990.

emissions from forest soils. The application of the model to a range of field sites showed that the model can reproduce observed differences in N trace gas fluxes. The model was able to explain 68% of the site variability for N_2O and even 78% of the site variability for NO. However, the detailed site evaluation also showed that the model is still far from perfect, e.g. with regard to the timing of peak emissions or the magnitude of the seasonality of N trace gas emissions (see e.g. Fig. 6 and Tables 2 and 3). Further improvement requires a better understanding and parameterisation of the microbial N turnover processes, especially with regard to denitrification with an emphasis on the ratio of N_2 to N_2O

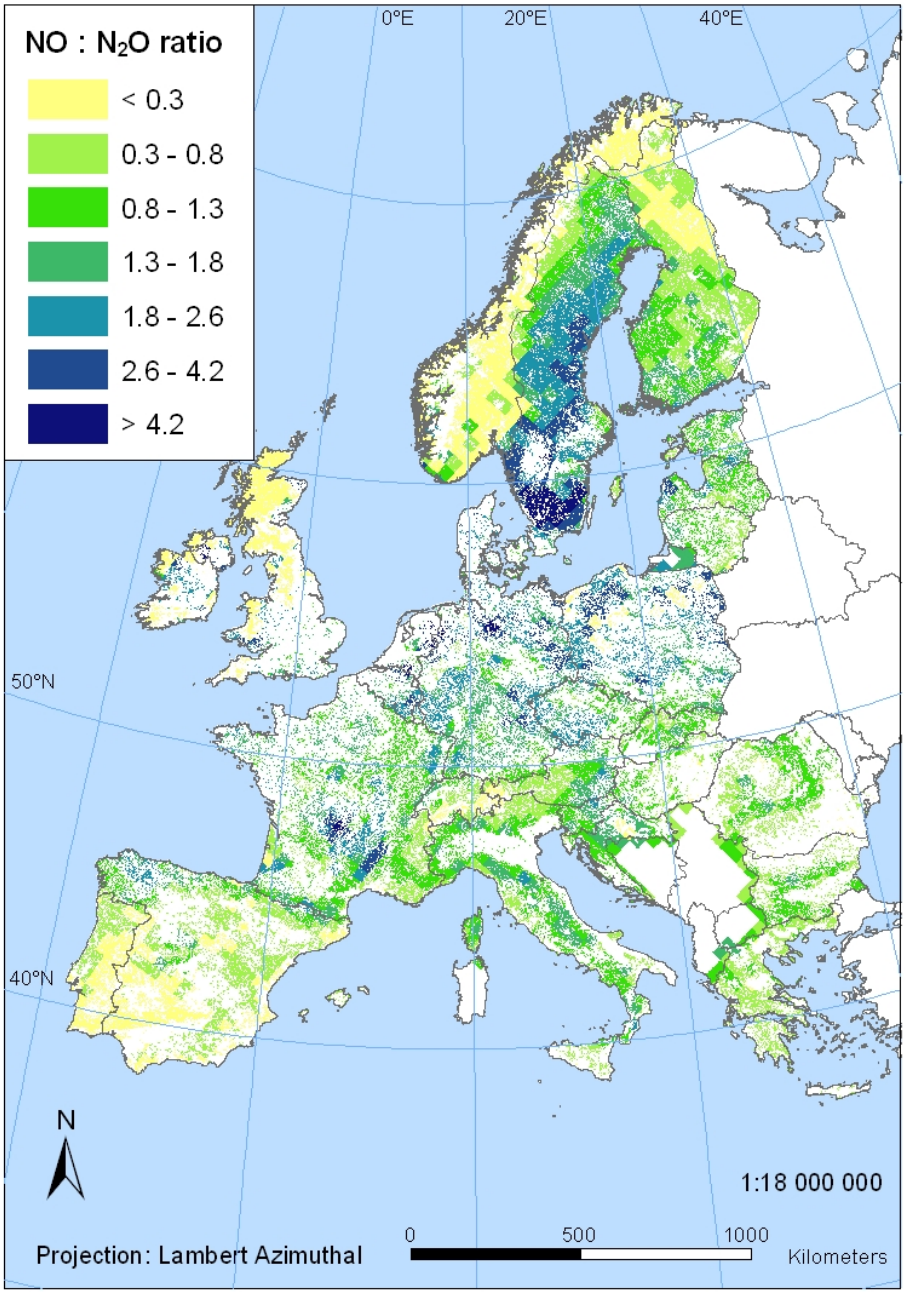

Fig. 10. Regional distribution of a NO to N_2O emission ratio from forest soils in Europe for the year 2000.

production. Furthermore, also the mechanistic description of the dynamics of plant N uptake versus microbial N uptake remains a challenge, since our understanding of the competition between the plant and microbial communities for N is still limited (Rennenberg et al., 1998). Also, more detailed site specifications such as the changes in soil properties with soil depth or the hydrological condition of a site in relation to its landscape position (e.g. with regard to the distance to the groundwater or occurrence of interflow) would further improve the basis for the simulation results. On the other hand this would require a further extension of the list of initializing and driving parameters and thus would strongly reduce the applicability of such a model on a regional basis due to the restricted availability of such data on a regional scale.

One can still find arguments that biogeochemical models such as the PnET-N-DNDC model are over-parameterised and that one may produce comparable results with more simple empirical approaches. However, due to the still limited number of field measurements and the observed importance of meteorological conditions and soil properties for the

magnitude of N trace gas emissions from soils a pure empirical approach is unlikely to work. Furthermore, empirical models can only be used for the range for which they have been validated, whereas in theory biogeochemical models can also be used as predicting tools e.g. for sites with significant different site properties as the one for which the model was tested.

There still remains an argument with regard to the availability and quality of GIS information for initializing and driving complex models. In view of e.g. the spatial inhomogeneity of soil properties and the sensitivity of the PnET-N-DNDC model to changes in e.g. SOC or soil pH (Stange et al., 2000) the uncertainty about the regional distribution of these parameters results in a significant increase of the prediction error. In this study we addressed this problem on a regional scale with the Most Sensitive Factor (MSF) method (Li et al., 2004) and with a Monte Carlo approach for selected grid cells across Europe. By this we were able to produce an uncertainty range, which most probably covers the mean N trace gas emission of a given grid cell.

4.1 N_2O emissions

In our study we estimated that total average N_2O emissions from forest soils across Europe in the years 1990, 1995 and 2000 were in the range of 77.6 to 81.6 kt N yr^{-1}. The respective uncertainty range – as calculated with the MSF method by Li et al. (2004) – is 50 to 100 kt N yr^{-1}. Furthermore, we calculated that the average N_2O emission per ha forest soil and year was 0.55 to 0.62 kg N ha^{-1} yr^{-1}, with elevated emissions in Central Europe and Western Spain but also in parts of Scandinavia where soils with high organic C content in the forest floor are found. Lower emissions were calculated for the UK and some boreal forest areas. Based on a literature review by Brumme et al. (2005) N_2O emissions from boreal forests ranged between 0.1 and 0.3 kg N ha^{-1} yr^{-1}. For most forested areas in Norway, Finland and Sweden our simulation results are in line with this estimate. However, especially for those areas in Scandinavia and the Baltic States for which elevated C stocks in the forest floor are reported, considerably higher N_2O emissions with values >0.75 kg N ha^{-1} were calculated. The estimate by Brumme et al. (2005) was mainly based on N_2O emission measurements from mineral soils in the boreal region (e.g. Martikainen, 1996), whereas estimates in other recent publications, in which N_2O emissions in the boreal zone from forest soils rich in humus were reported (von Arnold et al., 2005; Maljanen et al. 2001, 2003), resulted in annual N_2O emission rates in the range of 1.0 to 10.0 kg N ha^{-1} yr^{-1}. The highest N_2O emissions from boreal forest soils have been reported from peat soils, which have been used for agriculture prior to forestation (Maljanen et al., 2003). In the contrary, nutrient poor organic forest soils have been reported to emit negligible amounts of N_2O to the atmosphere (Regina et al., 1996). The huge discrepancy between both estimates is obvious and cannot be further clarified at present. We only can assume that C-rich soils from former peatlands, which have widely been drained in Fennoscandia for improving forest growth (Paavilainen and Päivänen, 1995) are indeed a stronger source for atmospheric N_2O than other soils poorer in C content in this area. In agreement with field studies, also other modelling studies dealing with effects of management practices such as no-till on N_2O emissions from agricultural soils, show that the magnitude of N_2O emissions is most likely positively correlated with SOC (Six et al., 2004; Li et al., 2005). However, further field studies on soils, differing in SOC but also in the ratio of C:N, are needed to further evaluate this interrelation and to proof the model algorithms and predictions.

N_2O emission measurements from temperate forest soils in Europe have been reported to vary substantially over a wide range from 0 to 20 kg N ha^{-1} yr^{-1} (see e.g. data compilation by Papen and Butterbach-Bahl, 1999) with a mean range of 0.2 to 2.0 kg N ha^{-1} yr^{-1}. The variability in the emission strength was found to be influenced by soil properties such SOC, pH, N deposition and forest stand properties (e.g. Papen and Butterbach-Bahl, 1999; Brumme et al., 1999; Zechmeister-Boltenstern et al., 2002; Jungkunst et al., 2004). Furthermore, the occurrence of high winter N_2O emissions during freezing and thawing events was acknowledged as a major factor determining the magnitude of annual N_2O emissions (Butterbach-Bahl et al., 2002b; Teepe and Ludwig, 2004). To reduce the uncertainty in estimates of N_2O emissions from temperate forest ecosystems different approaches from empirical based stratifications (Brumme et al., 1999) towards the use of process-oriented models (Butterbach-Bahl et al., 2001, 2004) have been followed. Using a stratification approach in combination with functions for N_2O production in dependency from soil water content and temperature, Schulte-Bisping and Brumme (2003) estimated that the average N_2O emission from forest soils in Germany is 0.32 kg N_2O N ha^{-1} yr^{-1}. This estimate may represent the lower boundary of emissions since neither N deposition effects nor freezing-thawing events were considered in this approach (Schulte-Bisping and Brumme, 2003). Both of these factors were considered in the studies by Butterbach-Bahl et al. (2001, 2004) who used an older version of the PnET-N-DNDC model for estimating the regional emission strength of forest soils in South Germany and Saxony. Their estimate of a mean annual N_2O emission of approx. 2 kg N ha^{-1} yr^{-1} is significantly higher than the one of Schulte-Bisping and Brumme (2003). It is also higher than estimates calculated with the recent version of PnET-N-DNDC for Germany as presented in this paper (0.6 to 0.8 kg N ha^{-1} yr^{-1}, see Table 4), which is partly due to a) an improved parameterisation of processes in the new model which was based on laboratory studies (Schindlbacher et al., 2004; Kesik et al., 2005[1]), b) the aggregation of site information on the EMEP grid (50 km by 50 km) raster instead of defined polygons as in the earlier studies, and c) different simulation years.

With regard to the Mediterranean region only limited information about the magnitude of N_2O emissions from soils is available (see review by Butterbach-Bahl and Kiese, 2005). The few publications available show that forest soils in this area can even function as sinks for atmospheric N_2O (Rosenkranz et al., 2005). Except for parts of Eastern Spain also our model calculated low estimates of N_2O emissions in the Mediterranean region (<0.5 kg N m^{-2} yr^{-1}), which was largely due to a model-intrinsic limitation of microbial N turnover processes by water stress.

4.2 NO emissions

NO emissions from forest soils across Europe in the years 1990, 1995 and 2000 were calculated to be in the range of 84.9 to 98.4 kt N yr^{-1} and, thus, approx. 10% higher than for N_2O. The range of uncertainty as calculated with the MSF method is 38.3 to 254.0 kt N yr^{-1}. Using either a methodology based on Skiba et al. (1997) or Davidson and Kingerlee (1997), Simpson et al. (1999) estimated NO emissions from soils covered with natural or semi-natural vegetation in Europe. By applying the Skiba et al. (1997) methodology excluding Russia, Ukraine and the former Yugoslavia the authors came up with an estimated NO emission of approx. 20 kt N yr^{-1} from forest soils across Europe. The respective range for the Davidson and Kingerlee methodology was 10 to 300 kt N yr^{-1} (Simpson et al., 1999). The differences in the magnitude of estimated NO emissions from forest soils in Europe between the Skiba et al. (1997) methodology as applied in Simpson et al. (1999) and our approach is mainly due to the intended simplicity of the Skiba et al. (1997) methodology which mainly considers temperature and N input to natural systems as parameters for estimating soil bound NO emissions but neglects effects of texture, SOC or pH.

The simulated average NO emission per ha forest soil was 0.6 to 0.7 kg N ha^{-1} yr^{-1}, which is in agreement with estimated average NO emissions from forest soils in the Southeastern United States (Davidson et al., 1998). In our study the highest emissions of NO from forest soils (>3 kg N ha^{-1} yr^{-1}) were simulated for highly N-affected forest areas in the Benelux states and Northern Germany, which is in accordance with field observations by Van Dijk and Duyzer (1999), who reported average NO emissions >6 kg N ha^{-1} yr^{-1} for beech and Douglas fir forests exposed to an atmospheric N deposition of approx. 40 kg N ha^{-1} yr^{-1} at Speulderbos, Netherlands. The latter results were also confirmed by measurements within the NOFRETETE project. Also for the Höglwald region in Southern Germany, for which long-term measurements of NO emissions from beech and spruce forests are available (Gasche and Papen, 1999; Butterbach-Bahl et al., 2002b) simulated emissions are in accordance with field observations. For the respective grid cell we simulated an average emission of 1.5 to 3.0 kg N ha^{-1} yr^{-1}, which is lower than the observed NO emissions from the spruce site of the Höglwald Forest (>6 kg N ha^{-1} yr^{-1}), but in agreement with observed average NO emissions from the beech site (approx. 2.8 kg N ha^{-1} yr^{-1}) (Butterbach-Bahl et al., 2002b). The relatively minor discrepancies are only due to differences in scale, since in our approach generalized information for the 50 km by 50 km grid cell was used, e.g. with regard to soil properties or atmospheric N deposition. However, for large forest areas in Sweden also NO emissions in a range of 1.5 to 3.0 kg N were calculated (Fig. 9c), that are not confirmed by any measurements at present. Johansson (1984) who carried out measurements in forests close to Stockholm found that NO emissions from unfertilized forest soils were lower than 0.1 kg N ha^{-1} yr^{-1}. One still can argue that the differences in scales make it difficult to compare the results, but by studying the reasons why simulated NO emissions in large parts of Sweden were elevated we found that this was mainly due to increased NO production via chemo-denitrification. Since the mechanisms in the PnET-N-DNDC model which are dealing with NO production via chemo-denitrification are in accordance with results from laboratory and field studies (e.g. van Cleemput and Baert, 1984; Gasche and Papen, 1999; Kesik et al., 2005[1]), the main reason for such a discrepancy may be due to an underestimation of NO consumption in the model. At present the model only considers that NO can be consumed by denitrification, but in soil incubation studies it was shown that also oxidative NO consumption may significantly contribute to NO consumptions especially in soils rich in SOC (Dunfields and Knowles, 1997). However, since the mechanistic basis of oxidative NO consumption is not well described at present this process is still not included in the model. A further reason for the discrepancy between observed and simulated NO emissions for parts of Scandinavia may be due to differences in soil properties which were used in our model simulations as compared to those found in the individual studies. The soil pH at the sites where Johansson (1984) carried out his measurements was 4.0 or 4.5, respectively. But the soil information derived from the Soil Geographical Data Base of Europe and used in the present work revealed that the soil pH in most parts of Scandinavia is <4.0 (see Fig. 2d), i.e. at a level where chemo-denitrification in the PnET-N-DNDC model is active (Li et al., 2000). Furthermore, in our model approach we do not consider the microsite variability of soil pH. It has been shown in several studies that e.g. in acid forest soils the soil pH can vary on a microsite scale for up to 3 pH units (e.g. Häussling et al., 1985; Strong et al., 1997; Bruelheide and Udelhoven, 2005). If nitrite production by e.g. nitrification would mainly be associated with the higher pH microsites and if one would disregard transport and other consumption except chemo-denitrification, the model would certainly overestimate NO production by chemo-denitrification. For the given reasons, there is a need to develop algorithms which are addressing microsite variability of soil pH in future model versions.

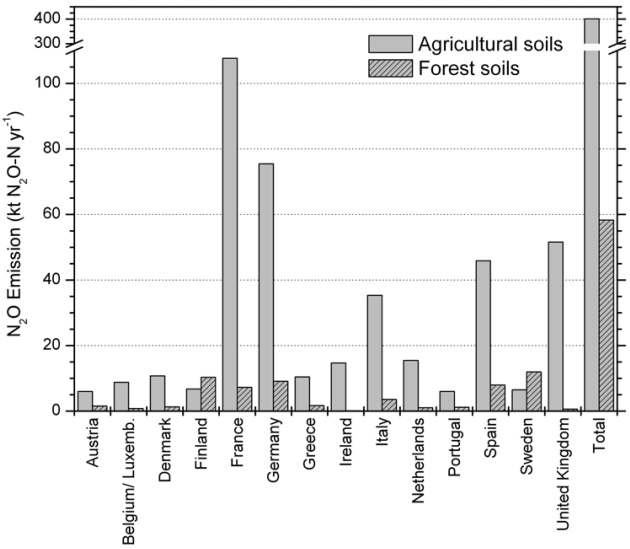

Fig. 11. Comparison of total direct N_2O emissions from agricultural soils (data for the year 1996 by Boeckx and van Cleemput, 2001) and of N_2O emissions from forest soils (data for the year 2000, this paper) for the EU15 countries.

4.3 Relevance of N trace gas emissions from forests soils in Europe as compared to other sources

Fertilized agricultural soils are assumed to be the predominant source for atmospheric N_2O. Boeckx and Van Cleemput (2001) evaluated the emission of N_2O from agricultural soils in Europe using the IPCC methodology (Mosier et al., 1998). They estimated that the average direct N_2O emission from agricultural soils across Europe is 5.6 kg N $ha^{-1}\,yr^{-1}$. Somewhat lower numbers where calculated by Freibauer and Kaltschmitt (2003) who considered besides the amount of N fertilization also climatic, soil and management factors for the calculation of their estimate. For agricultural, minerotrophic soils for the temperate and sub-boreal climate regions in Europe they came up with an average emission of 2.0 kg N $ha^{-1}\,yr^{-1}$. This means that agricultural soils are approx. a four to ten-fold stronger source for N_2O as compared to forest soils, where the average emission was calculated to be in a range of 0.55 to 0.62 kg N $ha^{-1}\,yr^{-1}$. However, if the total N_2O emissions for each land use type for different countries are calculated, the importance of forest soils as sources for atmospheric N_2O becomes evident. Using our approach and the estimates by Boeckx and Van Cleemput (2001) the emission strength of forest soils for N_2O is on average 14.5% of the emission strength of agricultural soils in Europe (Fig. 11).

Pyrogenic emissions are the dominating source for atmospheric NO_x in Europe. Using estimates of Vestreng et al. (2004), total pyrogenic emissions for our simulation area in the year 2000 would amount to approx. 10 000 kt N yr^{-1}. Our calculations for NO emissions from forest soils using meteorology for the years 1990, 1995 and 2000 was in the range of 85 to 99 kt N yr^{-1}, which is <1% of the pyrogenic NO_x emissions. Using a temperature function developed by Williams et al. (1992), Stohl et al. (1996) estimated that emissions of NO from all soils including forest, grassland and arable soils, to be only 1.6% of total pyrogenic emissions. Simpson et al. (1999) presented calculations of soil-NO emissions with a range of methods (a modified version of Skiba et al., 1997; BEIS-2 – from Novak and Pierce, 1993; Yienger and Levy, 1995; Davidson and Kingerly, 1997) accounting in some of these for N inputs from atmospheric N deposition or fertilizer. The range of estimates was very large. Using the Davidson and Kingerly methodology to derive an upper and lower boundary, with the extreme assumptions of no N affected forest, or 100% N-affected forest, yielded a range of 13 to 350 kt N yr^{-1} for forests. Compared to the approaches documented in Simpson et al. (1999), the PnET-N-DNDC approach as presented here is by far more complicate but with sounder physical basis, since it considers besides temperature and N availability also the effect of e.g. soil pH, moisture or texture on NO emissions from forest soils.

It needs to be noted that the NO once emitted from the soil underlies rapid oxidation to NO_2 in the presence of O_3. E.g. Rummel et al. (2002) showed, that during daytime up to 90% of the NO emitted from the soil may get converted to NO_2 within the trunk space and canopy of forests by the reaction of NO with O_3, which is transported from above the canopy by turbulence. NO_2 can be re-deposited to plant or soil surfaces or metabolised by plant tissues (Meixner, 1994; Ludwig et al., 2001; Sparks et al., 2001), so that the amount of NO and NO_2 (summarized as NO_x, i.e. NO+NO_2) finally emitted to the atmosphere at the canopy level may get strongly reduced. For tall vegetation covers such as forests, the so called canopy reduction factor for NO_x emissions is correspondingly estimated to be in the order of 50% (Jacob and Bakwin, 1991; Yienger and Levy, 1992; Ganzefeld et al., 2002).

5 Conclusions

The GIS-coupled process-oriented model PnET-N-DNDC, which was tested prior to its regional use on a large field data set, was used for the calculation of inventories of N_2O and NO emissions from forests soils in Europe. The results demonstrate that forest soils are a significant source of N_2O in Europe. With regard to NO forest soils only contribute <1.0% to total NO_x emissions in Europe. This number may even be lower if deposition processes of NO_x in the canopy are considered (canopy reduction). However, due to the seasonality of NO emissions the relative contribution of NO emissions from forest soils to total NO_x emissions can be larger during the vegetation period and can be of importance especially in rural areas. After a thorough validation of the

model against field observations and an extensive sensitivity analysis of the model performance, we conclude that the PnET-N-DNDC performs very well simulating NO and N_2O emissions from European forest soils. This work demonstrates that GIS-coupled process-oriented models are valuable tools to realistically estimate biogenic N trace gas emissions from soils. From our point of view this is the most comprehensive effort to date to simulate NO and N_2O emissions from European forest soils.

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