



Spatial and temporal variability of nitrous oxide emissions in a mixed farming landscape of Denmark

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Received: 2 December 2011 – Published in Biogeosciences Discuss.: 14 December 2011

Revised: 29 June 2012 – Accepted: 11 July 2012 – Published: 7 August 2012

Abstract. Nitrous oxide (N₂O) emissions from agricultural land are variable at the landscape scale due to variability in land use, management, soil type, and topography. A field experiment was carried out in a typical mixed farming landscape in Denmark, to investigate the main drivers of variations in N₂O emissions, measured using static chambers. Measurements were made over a period of 20 months, and sampling was intensified during two weeks in spring 2009 when chambers were installed at ten locations or fields to cover different crops and topography and slurry was applied to three of the fields. N₂O emissions during spring 2009 were relatively low, with maximum values below 20 ng N m⁻² s⁻¹. This applied to all land use types including winter grain crops, grasslands, meadows, and wetlands. Slurry application to wheat fields resulted in short-lived two-fold increases in emissions. The moderate N₂O fluxes and their moderate response to slurry application were attributed to dry soil conditions due to the absence of rain during the four previous weeks. Cumulative annual emissions from two arable fields that were both fertilized with mineral fertilizer and manure were large (17 kg N₂O-N ha⁻¹ yr⁻¹ and 5.5 kg N₂O-N ha⁻¹ yr⁻¹) during the previous year when soil water conditions were favourable for N₂O production during the first month following fertilizer application. Our findings confirm the importance of weather conditions as well as nitrogen management on N₂O fluxes.

1 Introduction

Atmospheric nitrous oxide (N₂O) concentrations have increased during the industrial era due to increased anthropogenic emissions (Smith, 2004). Nitrous oxide acts as a potent greenhouse gas and is also involved in the destruction of stratospheric ozone. There has been an increasing effort towards quantifying and identifying the sources and sinks of N₂O, in order to better predict and possibly mitigate future emissions by improved management of land and resources.

Approximately 65 % of atmospheric emissions of N₂O originate from soils (Smith, 2004). N₂O is produced in soils as an intermediate in the two contrasting microbial processes: autotrophic nitrification and heterotrophic denitrification. The contributions of these two processes to N₂O emissions vary with climate, soil conditions and soil management (Skiba and Smith, 2000). Land use type and changes in the crop rotation are also particularly important in determining the rate of N₂O emissions from agricultural land. For a specific crop type and soil type, it is typically found that fertilization rate, fertilizer type, timing, and cultivation play an important role in the processes controlling N₂O emission from soils (Kavdir et al., 2008). N₂O fluxes can be sporadic and short-lived, depending on the environmental conditions of the soil, particularly the temperature and moisture content (Smith et al., 1998; Frohling et al., 1998). Schaufler et al. (2010) observed a positive correlation of N₂O fluxes with

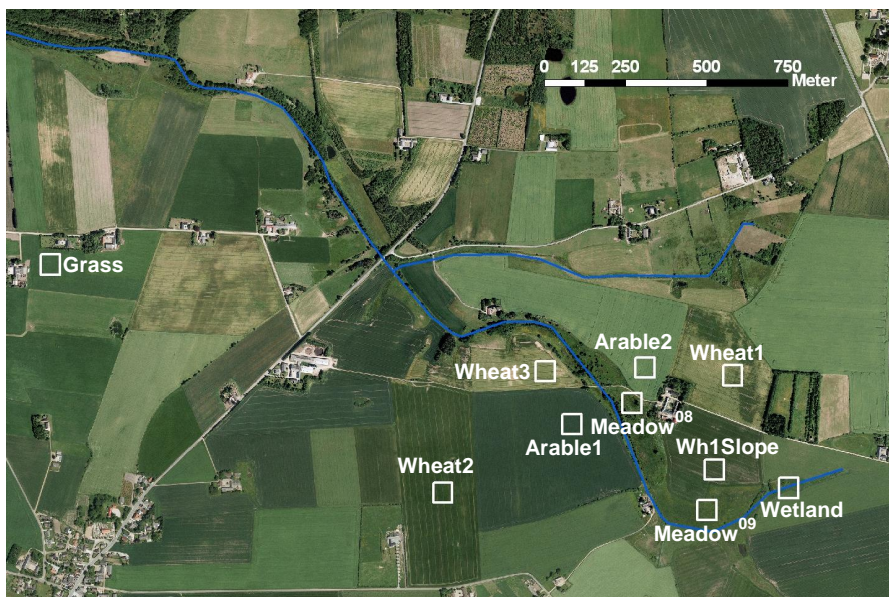


Fig. 1. Aerial photograph showing the ten locations for manual chamber measurements within the Bjerringbro landscape. Arable1 and Arable2 were studied throughout the period 2007–2008 and the 2009 campaign. Meadow⁰⁸ was studied during 2007–2008, and remaining sites were only monitored during the intensive campaign in 2009. The aerial photograph was taken in early summer of 2010.

soil moisture but found no significant relationship between emissions and N fertilization or N deposition level when analyzed over all soil moisture conditions. In short, several factors influence N₂O flux patterns, and the interactions between factors make it difficult to predict emissions on a short or long time scale (Machefert et al., 2002).

Many previous inventories or measurement studies on N₂O flux dynamics have focused on ecosystems or land-use types such as forests (Pihlatie et al., 2005; Kesik et al., 2005), wetland/organic soils (Maljanen et al., 2003), grasslands (Flechar et al., 2007; Chatskikh et al., 2005), or arable crop rotations in experimental plots (Petersen et al., 2006; Chirinda et al., 2010). However, there have been few attempts (Dunmola et al., 2010) to cover the mosaic of land use types encountered at the landscape scale within a single study. Patey et al. (2006) demonstrated how micrometeorological measurement techniques can be used for the up-scaling from field to landscape. While these techniques provide valuable integrated information at the landscape scale, they do not easily distinguish between contributions from individual areas. Emissions of N₂O most often form a heterogeneous signal with local hotspots contributing significantly to the total flux (Matthews et al., 2010). Laville et al. (2011) concluded that the uncertainty in cumulated N₂O emissions due to infrequent sampling was less than the uncertainty due to spatial variability of the sampling sites.

The current study was designed as a component of a measurement programme to estimate nitrogen fluxes and land management at the landscape level (Theobald et al., 2011) and to support modelling of the landscape-scale N fluxes

(Duret et al., 2011). The objective of the study was to examine the major drivers of N₂O emissions in a diverse agricultural landscape. Chamber measurements were made over a 20-month period at 10 sites in a Danish landscape to identify variations in emissions due to topography, land use or crop type, and management. During a limited period, more intensive measurements were made using additional chambers to study the effect of slurry application on N₂O emissions from three wheat fields. Based on the experiences during the previous growing season, we expected to find significant fluxes of N₂O following the targeted springtime manure application, which was the focus period of the intensive measurements.

2 Materials and methods

2.1 Landscape and duration of experiment

The study landscape covers an area of 6 km² and is located near Bjerringbro, Denmark (56.3° N, 9.7° E), in a mixed farming area with a mosaic of dairy, pig, and arable farms (Dalgaard et al., 2002). A stream surfaces in the area, forming a small valley in the otherwise generally flat landscape (Fig. 1). The soil type of the agricultural fields is a sandy loam with clay and organic matter contents of 12 % and 3–4 % in the topsoil, respectively, and 12–18 % and 1–2 % in the 25–50 cm depth interval, respectively. Narrow areas along the stream are managed or unmanaged meadow (15 % and 25 % organic carbon in the 0–25 cm and 25–50 cm depth intervals, respectively) while the remaining area is used for intensive agricultural production with fields that are fertilized

Table 1. Field names, crops and fertilization schemes at sites studied during 2007–2009. The duration of measurements (either long-term (L) or intensive campaign (C)) is also given.

Field	Duration	Area (ha)	Crop in 2008/2009	Fertilizer type	2008		2009	
					kg N ha ^{-1a}	Dates	kg N ha ^{-1a}	Dates
Meadow ⁰⁸	L	6.5	Permanent grass	Synthetic	50 ^b	5/5/2008	44 ^b	1/5/2009
Arable1	L	8.5	Spring barley/ winter oilseed rape	Synthetic Manure (cattle)	43 185.5	3/5/2008 15/3/2008	55+78 53	1/2/2009; 1/4/2009 1/9/2008
Arable2	L	12.7	Winter wheat/ winter barley	Synthetic Slurry (pig)	43+43 136	17/3/2008; 22/4/2008 20/3-2008	55+88 0	1/2/2009; 1/4/2009 –
Wheat1	C	9.1	Oats/ winter wheat	Synthetic Slurry (pig)	76 0	1/4/2008 –	50+50 87	1/3/2009; 1/4/2009 23/4/2009
Wheat2	C	11.6	Winter wheat/ winter wheat	Synthetic Slurry (pig)	72 135	5/5/2008 1/4/2008	72 112	5/5/2009 22/4/2009
Wheat3	C	4.1	Oats/ winter wheat	Synthetic Slurry (pig)	76 0	1/4/2008 –	50+50 87	1/3/2009; 1/4/2009 27/4/2009
Grass	C	3.5	Grass-clover in rotation	Synthetic Slurry (cattle)	100+150 164 ^b	10/4/2008; 15/6/2008 1/3/2008	100+150 164 ^b	10/4/2009; 15/6/2009 15/2/2009
Wetland	C	1.0	Mixture of herbs	None	0	–	0	–

^a kg total N.^b Excluding manure excreted by grazing cattle.

with a combination of synthetic fertilizer and animal manure (Wohlfart et al., 2012). The climate of the region, Central Jutland, is coastal temperate with a mean annual precipitation (1961–1990) of 722 mm and annual mean temperature of 7.7 °C. The mean temperatures of the coldest and warmest months of the year (February and July) are 0.1 °C and 15.4 °C, respectively. The experiment took place during September 2007–April 2009. During the first full year, measurements focused on three long-term sites (Arable1, Arable2 and Meadow⁰⁸; Table 1 and Fig. 1). In spring 2009, a coordinated effort was made to study more sites during an intensive period (21–28 April 2009). Ahead of the intensive campaign, the long-term sites were pre-monitored, beginning on 30 March 2009. The study took place at sites and fields that were conventionally managed by the farmers of the area. Three different farmers own and managed the fields studied. Agricultural management was recorded through farmer interviews, by consulting agronomic advisory services, and by observations made by the researchers. During the intensive measurement campaign of April 2009, pig slurry was applied to three wheat fields (Wheat1, Wheat2, Wheat3) using trailing hoses, and the days of application were agreed to accommodate the measurement activities. An overview of fields and fertilization rates during 2007–2009 is given in Table 1.

2.2 Chamber measurements

Four chamber types, CH1–CH4, were applied for measuring fluxes of N₂O within the landscape (Table 2). All were two-part static chambers located at the sites shown in Fig. 1. A total of 75 chambers were installed in groups of 5–6 chambers at ten different locations or fields to cover different topography, land use, and crop types. CH2 chambers were deployed throughout 2007–2009 at monthly or bi-monthly intervals, with a higher frequency in periods of increased management activity, especially fertilization. All chamber types were used during the April 2009 campaign when some chamber types were installed in groups in the same fields (Table 2). For the 2009 campaign, the chamber frames were installed prior to and remained in place during and after slurry application. For the long-term installations in Arable1 and Arable2, the CH2 steel frames were installed and only removed for harvest and seedbed preparation. On each sampling day and for each chamber, 30-ml (CH1, CH3) or 10-ml (CH2) gas samples were taken at four regular time intervals between 0 and 70 min (CH1, CH3) or 0 and 90 min (CH2). Gas sampling from CH4 was 3 samples made at 15-min intervals. Samples were stored in pre-evacuated 12.5-ml or 6-ml glass vials in preparation for analysis. N₂O concentrations in the gas samples were subsequently analysed using gas chromatography at three different laboratories. Samples from CH2 chambers were analysed according to Petersen et al. (2008) in 2007–2008 and according to Petersen et al. (2011) in 2009.

Table 2. Chamber types and fields where they were used in the Bjerringbro landscape. C2009 indicates the fields where the chambers were used during the 2009 intensive campaign.

Chamber type ID	Chamber area (m ²)	Effective volume (L)	Number of chambers	Materials and other information	Operated at fields
CH1	0.24	50	39	Al-frame & plastic cover clamped to frame. No vent, no mixing.	C2009: Wheat1, Wh1Slope, Wheat2, Wheat3, Grass, Wetland, Meadow ⁰⁹
CH2	0.21	45 or 100	16	Steel frame with plastic cover inserted into water-filled rim of frame. Vented, no mixing.	2007-8: Arable1, Arable2, Meadow ⁰⁸ C2009: Arable1, Arable2, Wheat3
CH3	0.24	65	10	Plastic frame and plastic cover inserted into water-filled rim of frame. No vent, no mixing.	C2009: Wheat1, Wheat2
CH4	0.007	0.5	10	Plastic cylinder and plastic cover. No vent, no mixing.	C2009: Wheat2

Samples taken from CH1 and CH3 chambers were analysed according to Vilain et al. (2010). Samples taken from CH4 were analysed according to Machon et al. (2010).

N₂O fluxes were calculated using the HMR procedure (Pedersen et al., 2010), available as an add-on package for the free programming software R (<http://www.r-project.org>). The HMR tool analyses non-linear concentration time series based on the model by Hutchinson and Mosier (1981). If the concentration data develop linearly with time, a linear regression is applied. HMR suggests the optimal analysis method and offers a graphical interface where the user makes the final decision on analysis method for each concentration time series. HMR calculates statistics for each regression, enabling the rejection of fluxes that are not significantly different from zero. In this study, a concentration dataset of four samples over time was accepted only if it gave a statistically significant regression ($p \leq 0.15$).

2.3 Meteorology and soil

Meteorological records (hourly and daily time resolution) of air temperature, global radiation, and precipitation were obtained for the entire measurement period from a meteorological station operated by the Danish Meteorological Institute and located in a similar agricultural landscape 25 km east of the area. Daily reference evapotranspiration (E_{ref}) was derived from the meteorological data and the Makkink equation (de Bruin and Lablans, 1998). The Makkink equation has been shown to provide adequate evapotranspiration estimates for an unstressed grass crop for Danish conditions (Kjaersgaard et al., 2008), and E_{ref} was assumed to be close to the potential evapotranspiration of the range of crops and land-use types studied. Soil water content was measured using a portable TDR and a 0.25 m probe near the chamber lo-

cations. Measurement frequency was the same as that of the N₂O flux measurements, except for some occasions when the TDR probes could not be inserted because the soil was too dry. In addition, during the intensive campaign in 2009, meteorological variables (radiation, air and soil state variables including soil water content in the topsoil) were measured at field Wheat2 with a high time resolution (15 min) using an instrument setup similar to that described in Horvath et al. (2005) and Meszaros et al. (2009). During the campaign period, a mast equipped with instruments for Bowen ratio measurements (Campbell Scientific, Loughborough, UK) of heat fluxes was also installed at field Wheat2. Throughout the 20-month period of measurements, soil was sampled near chamber positions on a subset of the dates when N₂O fluxes were measured. On each sampling date, five soil cores were taken from random locations at each site and the samples from the 0–25 cm and 25–50 cm soil layers were pooled to obtain representative composite subsamples for the two depths. Samples were kept cool (< 10 °C) during transport to the laboratory. Samples were frozen until analysed for NO₃⁻ and NH₄⁺ content, according to Mutegi (2010) for samples taken near chambers CH2 and according to Vilain et al. (2010) for samples taken near chambers CH1 and CH3.

3 Results

3.1 Meteorology

Meteorological conditions during the 20-month measurement period (2007–2009) were not unusual. The two winters were mild with occasional frosts. Springs of 2008 and 2009 were both characterised by relatively dry conditions during mid to late April–May. In this rain-fed agricultural landscape

with sandy loam soils, these conditions caused the soils to dry out. Accordingly, measurements of soil water content at 5 cm depth of field Wheat2 during the intensive measurement period (19–29 April 2009) showed a steady decrease in the volumetric water content from 15 % to 10 %. Another indicator of soil water depletion and crop water stress was that during 19–28 April 2009 actual evapotranspiration, measured in the same wheat field using the Bowen Ratio method, was 64 % of E_{ref} that represented a grass surface without water stress. This ratio was calculated as the accumulated latent heat fluxes divided by accumulated daily reference evapotranspiration.

3.2 Chamber type and flux analysis method

When considering all significant fluxes recorded during 2007–2009, application of the flexible HMR procedure for analysing chamber N_2O concentrations resulted in calculated fluxes that were on average 32 % higher than fluxes calculated using a linear regression only (Fig. 2). For the limited period of intensive measurements during 2009, the HMR-based fluxes were 16 % higher than fluxes derived solely from a linear regression. During the intensive period of 2009, more chamber types were applied and some types were located in the same fields (Table 2). There was however no attempt to duplicate chamber measurements, so a day-to-day comparison between chambers types was not possible due to the spatial variability and different sampling schemes. Table 3 presents mean values of measured emissions during the 2009 campaign with the four types of chambers. Fluxes measured at Wheat3 using two chamber types (CH1, CH2) were in good agreement before the slurry application. At Wheat2, using another combination of chambers (CH1, CH3), fluxes were in good agreement both before and after the slurry application. However at Wheat2, chambers CH4 generally estimated small fluxes that were unaffected by the slurry application, in contrast to the two other chamber types. In field Wheat1, chambers CH3 generally measured smaller fluxes than chambers CH1.

3.3 Spatial variability

The variability of N_2O flux measurements made at a specific site using 5–6 chambers was quantified via the coefficient of variation (CV). The CV of flux estimates was 30–150 % depending on time of year and site, with the only obvious trend indicating higher values for the meadow site in 2007–2008, where CVs were 100–200 %. CVs were typically high during periods of relatively low fluxes when some chambers estimated a zero flux, and CVs were typically low when fluxes were high. During the 2009 intensive campaign, CVs, calculated per chamber type and site, ranged between 60 % and 140 %. Practically all measured fluxes were positive.

The effect of land use and land management on N_2O fluxes was studied using the full year of measurements at

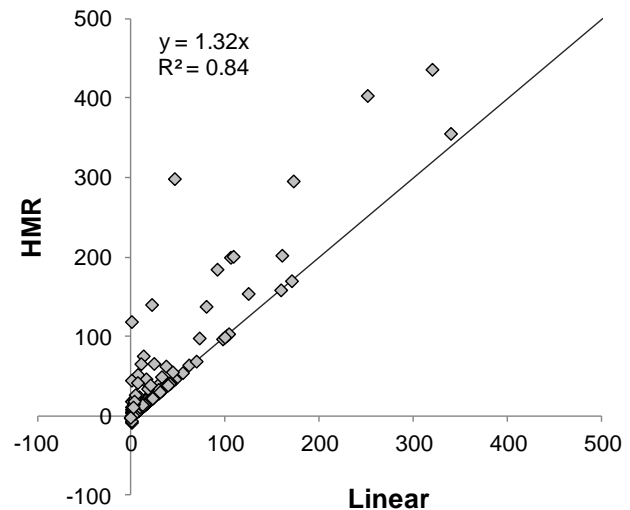


Fig. 2. Calculated N_2O fluxes ($\text{ng N m}^{-2} \text{s}^{-1}$), using the HMR tool, versus calculated fluxes using a linear regression for the analysis of chamber concentrations. The figure shows all significant individual chamber fluxes below $500 \text{ ng N m}^{-2} \text{s}^{-1}$ (2007–2009; $n = 364$). Four individual chamber fluxes were larger than $500 \text{ ng N m}^{-2} \text{s}^{-1}$. The line is the 1:1 line; the R^2 value is for a linear regression forced through the origin: $\text{HMR flux} = 1.32 \times \text{Linear flux}$. For the limited period of intensive measurements during 2009, the equivalent regression was $\text{HMR flux} = 1.16 \times \text{Linear flux}$.

sites Arable1, Arable2, and Meadow⁰⁸. Differences in management (Table 1) were mainly due to fertilization since the arable fields received both intensive organic and inorganic fertilizers, while the meadow was occasionally grazed by heifers and lightly fertilized with inorganic fertilizer. Fluxes from the meadow were low at all times during the year 2007–2008 ($< 20 \text{ ng N m}^{-2} \text{s}^{-1}$), while fluxes from the arable fields followed an annual pattern with emission peaks during spring and early autumn of up to $600 \text{ ng N m}^{-2} \text{s}^{-1}$ (Fig. 3).

During the intensive campaign of 2009, more types of arable fields were studied in addition to areas with more extensive land use. Table 3 shows that mean N_2O fluxes were all below $12 \text{ ng N m}^{-2} \text{s}^{-1}$. Table 3 allows a comparison of campaign emissions from different sites, with a range of fertilization intensities from unmanaged wetland to wheat and other arable fields that had been intensively fertilized for years. Measured wetland emissions were among the lowest recorded fluxes, but emissions of the fertilized fields were also found to be modest during the period.

The effect of topography and associated soil wetness variability on N_2O fluxes was investigated during the intensive period of 2009 by installing chambers along a transect (Fig. 1) from a plateau or shoulder (Wheat1) down a gentle slope (Wh1Slope) of the wheat field to the footslope in the small stream valley (Meadow⁰⁹). The first two transect points had a similar soil texture with a clay content of 7–8 % and a

Table 3. Measured N_2O fluxes ($\text{ng N m}^{-2} \text{s}^{-1}$) for chambers used during the intensive campaign in April 2009. Values are means (standard error of the mean in parentheses) for the entire intensive measurement period, or for the periods before and after slurry application to the wheat fields. Fields are defined in Table 1, and chamber types (CH1–CH4) are defined in Table 2.

Site	Period	CH1	CH2	CH3	CH4
Wheat3	Entire	5.8 (1.8)	6.4 (1.8)		
	Before	5.8 (1.8)	5.8 (2.2)		
	After		9.1 (2.6)		
Wheat2	Entire	10.2 (1.7)		8.6 (2.0)	3.0 (0.4)
	Before	0.1 (0.1)		0.3 (0.3)	3.4 (0.9)
	After	11.7 (1.8)		11.4 (2.3)	2.9 (0.5)
Wheat1 ^{TR}	Entire	8.3 (0.8)		2.5 (0.6)	
	Before	4.6 (1.2)		0.6 (0.6)	
	After	9.8 (0.8)		3.6 (0.8)	
Wh1Slope ^{TR}	Entire	5.5 (0.6)			
	Before	4.0 (1.1)			
	After	6.3 (0.7)			
Meadow ^{09-TR}	Entire	9.7 (2.3)			
Wetland	Entire	2.9 (0.7)			
Grass	Entire	2.3 (0.6)			
Arable1	Entire		8.7 (1.3)		
Arable2	Entire		3.3 (0.7)		

^{TR} Site also located on the sloping transect.

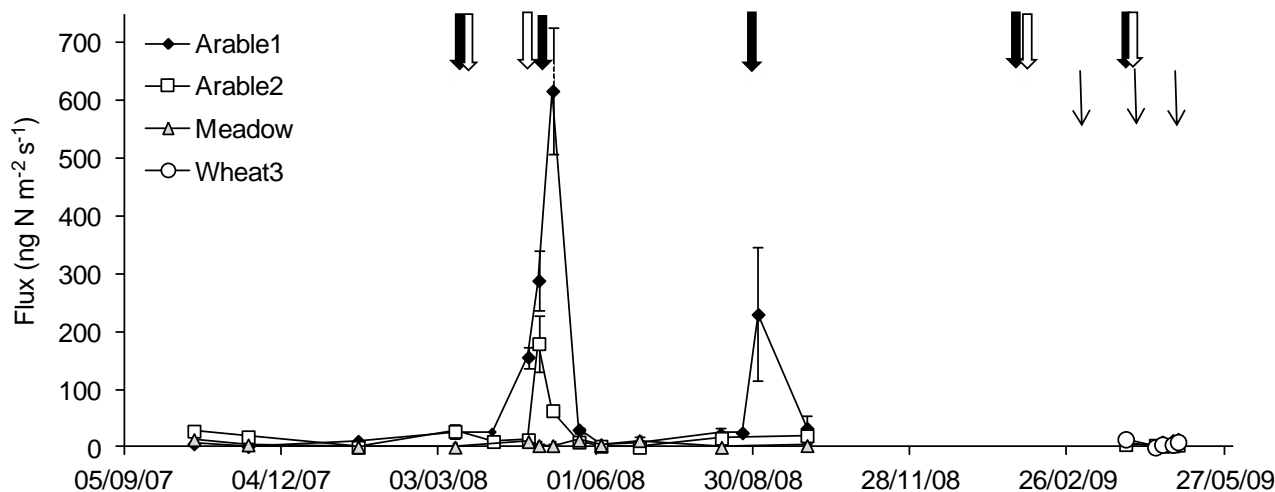


Fig. 3. Measured N_2O fluxes for the fields with CH2 type chambers. Error bars show the standard error of the mean. Arrows indicate the timing of fertilizer or manure application: Arable1 (black), Arable2 (white), and Wheat3 (thin arrows). Chambers located at Meadow⁰⁸ during 2007–2008 were operated at Wheat3 during the 2009 campaign. Fewer measurements were made at Arable1 due to late sowing of the crop in spring and autumn of 2008; hence, emissions at Arable1 were estimated for a few missing dates.

soil organic matter content of 3 % in 15–30 cm soil layer. The humus soil in the valley meadow had a larger clay and organic matter content at the 15–30 cm depth (16 % and 15 % respectively). The meadow soil was abundant in organic N (31 and 50 g N kg^{-1} DM (dry matter) for the 0–25 and 25–50 cm soil depths, respectively). In this soil the C/N ratio was around 12 for both depths. Soil water content by weight at depth of 5–15 cm was considerably higher in the valley

(48 %) than in the wheat field (9 %) on 21 April 2009. On the same date, mineral N contents were higher in the valley than in the wheat field: $\text{NO}_3\text{-N}$ and $\text{NH}_4\text{-N}$ concentrations at the wheat field were 4.2 and 4.6 mg N kg^{-1} DM, respectively, while $\text{NO}_3\text{-N}$ and $\text{NH}_4\text{-N}$ concentrations in the valley were 25 and 38 mg N kg^{-1} DM, respectively. The recorded N_2O fluxes along the transect were within the range observed at the other fields during the intensive period of April 2009,

and, in spite of the different soil conditions, no clear trend in fluxes with respect to position within the transect could be detected, neither in the individual fluxes (data not shown) nor in the mean fluxes (TR; Table 3).

3.4 Temporal variability

During the 2009 campaign, fluxes increased when wheat fields were fertilized with pig slurry, with about a doubling of the low emission rates (Table 3) although the effect was short-lived (2–3 days; Fig. 4). Since there was no precipitation during the campaign, irrigation (approx. 14 mm) was applied at the soil surface within chamber type CH1 on 26th April 2009. We did not see any clear effect of irrigation on the emissions at any of the irrigated sites (Fig. 4).

Temporal variability was most prominent for the two sites that were continued throughout the 2007–2009 period (Arable1 and Arable2). During April and early May of 2008, both fields experienced high N_2O emissions with fluxes at Arable1 exceeding $600 \text{ ng N m}^{-2} \text{ s}^{-1}$ and fluxes at Arable2 exceeding $150 \text{ ng N m}^{-2} \text{ s}^{-1}$ (Fig. 3). Emissions were low over the summer and increased again in September 2008 at Arable1 when a winter oilseed rape crop had been established. Compared with the fluxes in 2008, all fluxes recorded during spring 2009 were small. Spring peak emissions in 2008 were observed after sowing and fertilization of the crops (Table 1). At Arable2, the highest fluxes were observed after the application of two mineral fertilizer doses and a slurry application. At Arable1, fluxes began to increase significantly after the application of farmyard manure in spring. The autumn emission peak at Arable1 was observed after cultivation and sowing of the winter oilseed rape in late August followed by slurry application on 1 September 2008 (Fig. 3).

4 Discussion

4.1 Chamber methods

The static chamber method used for investigating the landscape scale variability of nitrous oxide fluxes is a reliable technique that can be deployed in remote areas. Fluxes can also be measured using micrometeorological techniques (Pihlatie et al., 2005) or automated chambers to allow measurements at a much higher temporal resolution (Ambus and Robertson, 1998; Laville et al., 2011), but for the present experiment the static chamber method was applied in order to increase spatial coverage (Theobald et al., 2011). The application of manual static chambers requires great care and is subject to several sources of uncertainty (Livingston and Hutchinson, 1995). The simple static chambers applied in the present experiment would receive a relatively high score according to e.g. dimensions and design, deployment time, number of samples and sample vial type in a systematic evaluation of chambers (Rochette and Eriksen-Hamel, 2008). Only one of the chamber types (CH2) was vented, but

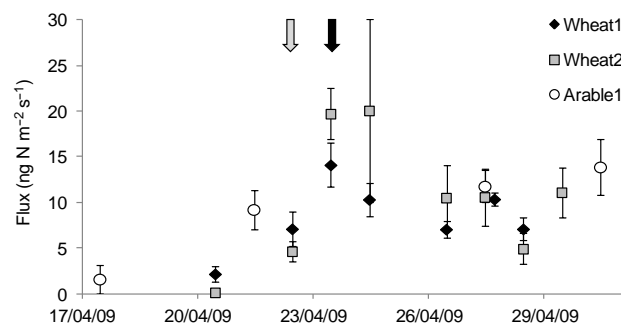


Fig. 4. Measured N_2O fluxes at two wheat fields and a winter oilseed rape field during the 2009 campaign. Error bars quantify the standard error of the mean. Arrows indicate the timing of slurry application at Wheat2 followed by Wheat1. Flux measurements at the treated fields were made some hours after the slurry application. The oilseed rape (Arable1) was not fertilized during the period. Chambers at both wheat fields were irrigated (14 mm) on 26 April 2009.

fluxes measured with CH2 chambers did not appear to differ from other chambers (statistical tests not appropriate due to spatial variability effects). There was an indication that the small CH4 chambers did not resolve the temporal variability as well as the larger chambers: their average flux did not show any response to slurry application (Table 3). One reason could be that inspection at the Wheat2 field revealed how slurry was applied, using trailed hoses, into only four of the ten pre-installed 10-cm diameter collars. Hence, these chambers were too small to capture management effects at field scale. Evaluations and protocols on static chamber designs and deployment are emerging (Smith and Conen, 2004; Rochette and Eriksen-Hamel, 2008; Christiansen et al., 2011; Rochette, 2011), and these guidelines should be consulted for future applications of the static chamber method. Also the choice of analysis of the concentration data (linear regression versus non-linear regression types) is important for the resulting fluxes (Anthony et al., 1995; Kutzbach et al., 2007; Venterea 2010). Our findings show that emission estimates were increased by 16–32 % (depending on period taken into account) when using the flexible HMR analysis tool. Kroon et al. (2008) and Thomsen et al. (2010) similarly reported how accounting for non-linearity increased N_2O flux estimates by approximately 100 % and 25–65 %, respectively. Unlike Venterea (2010), who suggested that the theoretical underestimation of linear regression-based fluxes would increase with decreasing soil wetness, our results indicated a smaller underestimation during the dry soil period of 2009 compared with the mean underestimation over the total measurement period (Fig. 2).

4.2 Spatial and temporal variability

Heterogeneity in N_2O fluxes is expected in natural and managed systems, and the coefficients of variation found in this landscape study (30–200 %) were much in line with the range of variability found in other studies at a similar scale (Ambus and Christensen, 1995; Dunmola et al., 2010; Vilain et al., 2010). Also, site-specific mean emissions may be categorised as being low or high emissions. In the following discussion, we will refer to emissions at a given site and day as being “low” when the measured flux did not exceed $20 \text{ ng N}_2\text{O-N m}^{-2}\text{s}^{-1}$. This threshold is close to the one applied by Kavdir et al. (2008).

4.2.1 Riparian areas

The lack of topographic effects on N_2O fluxes, or rather, the finding that emissions were low at all times in the meadow, was somewhat unexpected. Many previous studies have suggested that topography influences N_2O dynamics, with higher fluxes often found at the footslope compared to fluxes measured on slopes or shoulder positions along a topographical transect (Ambus and Christensen, 1995; Ambus, 1998; Velthof et al., 2000; Vilain et al., 2010). A likely mechanism would be the significant potential for denitrification in the wetter areas where nitrate-rich waters from adjacent agricultural fields converge (Wohlfart et al., 2012). The higher fluxes at the footslope are often associated with the relatively higher water-filled pore space in the soil at the bottom of the slope (Vilain et al., 2010; Gu et al., 2011). By contrast, Dunmola et al. (2010) investigated N_2O fluxes at upper, middle, lower, and riparian positions of an agricultural landscape and found that the lowest N_2O emissions were consistently from the uncropped riparian sites. Similarly, Vilain et al. (2010) observed lower fluxes in an unfertilized riparian area compared to a nearby fertilized plot. Within our landscape, gravimetric water content was high in the meadow compared to the slope and shoulder positions of the measurement transect in April 2009. Taking the different soil bulk densities into account, estimated water-filled pore space (WFPS) averaged 50 % in the meadow and 30 % at the slope and shoulder positions under these specific (dry) springtime conditions. Considering soil wetness during the entire period (2007–2009), the groundwater table in the meadow was located just below the soil surface during winter and gradually lowered to approximately 40 cm below ground during summer. Hence, the relatively wet soil moisture conditions should favour denitrification processes. With respect to substrate availability, NH_4 was abundant in the organic meadow soil with respect to NO_3 ; NH_4 concentrations were $0.020\text{--}0.045 \text{ g NH}_4\text{-N kg}^{-1} \text{ DM}$ during 2007–2008 with no clear annual trend, and the NH_4/NO_3 ratio ranged between 2 and 65 for the 0–25 cm depth and was equally high for the deeper soil layer. Soil sampled from the top 30 cm soil layers in April 2009 also confirmed a NH_4/NO_3 ratio above 1.0 in the meadow and

particularly at the wetland site. In a much more detailed study at a riparian wetland, Hedin et al. (1998) found a pattern in subsurface water chemistry and redox conditions with a dominance of NO_3 and N_2O in the near-stream environment and dominance of NH_4 and DOC in the more inland environments. They suggested that NO_3 near the stream was due to upwelling of nitrate-rich groundwater, while NH_4 abundance further away from the stream was mainly due to mineralisation of soil organic matter. In the inland region, denitrification (and N_2O production) would be limited by NO_3 availability. At our site, some of the adjacent fields and the meadow had tile-drainage (Wohlfart et al., 2012). Therefore, runoff, possibly rich in nitrate, from the adjacent fields may have been primarily directed to the stream via the drains, thus by-passing the meadow to a large extent. If our meadow was dominated by redox conditions similar to those observed by Hedin et al. (1998), this could help explaining why we always saw small N_2O fluxes from the meadow. However, the measurement setup at the meadow (five flux chambers grouped at Meadow⁰⁸ location during 2007–2008 and five other chambers at Meadow⁰⁹ location during 2009, supplemented by soil sampling for N content analyses) was insufficient to clarify the water and N dynamics. A more thorough understanding would require an analysis of the shallow groundwater system, including water sampling in transects along flow lines (Hedin, 1998; Burt, 2005). We conclude, in agreement with Vilain et al. (2010), that N_2O emissions at riparian buffer areas are highly site-specific and depend on the local hydrology and N input.

4.2.2 Arable land

With the lowest emissions encountered at the meadow and wetland sites, variability in N_2O emissions was mainly governed by land-use, i.e. fertilized arable fields emitted more N_2O than the wetland or meadow, which were not, or only marginally, fertilized (Table 1). Within-season management at the field scale was equally important since fluxes were highest after crop establishment and fertilization in spring and autumn (Fig. 3). The importance of land use and management is in agreement with findings of other researchers at field or plot scale. The review by Macheferet et al. (2002) revealed that high annual fluxes were predominantly encountered in agricultural ecosystems. Ambus and Christensen (1995) reported that mean annual N_2O emissions were lowest at five uncropped and unflooded sites, including forest and riparian land, compared with high annual emissions recorded at two arable sites. Chirinda et al. (2010) found that N_2O emissions were stimulated after spring fertilization in organic and conventional cropping systems. Kavdir et al. (2008) found that the response of N_2O emission to fertilization and crop type varied differently, calling for a separate interpretation of the results for each crop and fertilization level. In their study, N fertilization together with annual cropping doubled the N_2O emissions compared with perennial

crops. Some of these effects are likely to be linked to crop-specific effects of tillage and (cover) crop residue management (Chatskikh et al., 2008; Petersen et al., 2011). As discussed in Sect. 4.3, the prevailing soil moisture conditions are also important drivers of the temporal variability of N_2O emissions.

The highest emissions during the experimental period were found at Arable1 and Arable2. The peak emissions recorded at Arable2 in 2008 (up to $180 \text{ ng N m}^{-2} \text{ s}^{-1}$; Fig. 3) were comparable to other high values measured in arable systems (Hénault et al., 1998; Laville et al., 2011; Petersen et al., 2011) but were higher than maximum fluxes measured by Chirinda et al. (2010, $21 \text{ ng N m}^{-2} \text{ s}^{-1}$) and Mutegi et al. (2010, $14 \text{ ng N m}^{-2} \text{ s}^{-1}$) during the 2008 spring season at cereal crop sites located close (20 km) to the Bjerringbro landscape. Also during the 2008 spring season, Thomsen et al. (2010) observed maximum emissions of $50 \text{ ng N m}^{-2} \text{ s}^{-1}$ for cereal field plots on a loamy sand, treated with digested pig slurry, at a site located 55 km from the Bjerringbro landscape. By contrast, maximum measured emissions at Arable1 during spring and autumn of 2008 ($156\text{--}617 \text{ ng N m}^{-2} \text{ s}^{-1}$, Fig. 3) seemed to be extraordinarily high, although Kroon et al. (2008) observed large fluxes after fertilizer application at an intensively managed grassland and Denmead et al. (2010) found large and sustained emissions ($460 \text{ ng N m}^{-2} \text{ s}^{-1}$) at a fertilized acid sulphate soil producing sugarcane. The spring barley at Arable1 was fertilized with cattle farmyard manure (FYM) and mineral fertilizer in spring 2008, and manure was applied again before sowing the following oilseed rape crop in September 2008 (Table 1). This field had been exposed to many years of manure application, leading to an accumulation of labile organic matter pools. When applying FYM and incorporating it into the soil by ploughing, typically within a few hours to minimize NH_3 emissions, the manure can be unevenly distributed on the soil surface (Hansen, 2004), and later in the topsoil. Lumps of farmyard manure, rich in nitrogen and carbon, will exist that have a relatively high local water retention capacity, a high soluble carbon supply and a high O_2 demand, leading to the creation of anaerobic microsites that favour denitrification (Petersen and Sommer, 2011) and, therefore, have a high potential for N_2O emissions. Due to the high water holding capacity of the manure lumps, these conditions may prevail even when the soil is only moderately moist. In contrast, dissolved C and N in organic slurries with a much lower DM content are more homogeneously dispersed into a larger soil volume, thus changing the balance between aerobic and anaerobic decomposition. We hypothesise that heterogeneously incorporated FYM at Arable1 caused the unusually large N_2O emissions and suggest that the effects of FYM pockets acting as hotspots need further study. Thorman et al. (2007) applied pig and cattle FYM to cereal stubble and bare soil, respectively, with and without incorporation by ploughing. They found peak emissions from the ploughed pig FYM treatments to be up to 20 times larger than those from the equivalent

surface applied treatments, while there was no discernible difference in peak emissions between treatments with cattle slurry. On the other hand, Webb et al. (2004) did not find significant losses of N_2O when cattle or pig FYM was incorporated into the soil. Both Thorman et al. (2007) and Webb et al. (2004) used small chambers with a surface area smaller than 0.03 m^2 . There seems to be a lack of studies, at field level, of N_2O emissions following incorporation of FYM or deep litter into soil, using larger chambers or more integrative techniques such as eddy covariance that may take into account the effects of the spatial heterogeneity in FYM incorporation.

4.3 Fluxes and soil environmental conditions

As found in several other studies, N_2O fluxes were influenced by soil inorganic N content that varied throughout the growing season. N_2O emission rates, which peaked in spring and autumn 2008 after fertilization, were positively correlated with soil mineral N content when disregarding times of the year when soil conditions were unfavourable for N_2O production. In Fig. 5, dry soil conditions (as defined in Fig. 6) are highlighted as conditions when no clear response to mineral N content would be expected in the recorded N_2O flux. The relations shown in Fig. 5 for NO_3 content in the topsoil, and for total N content in the topsoil and at a depth of 25–50 cm, exhibit considerable scatter, but still indicate that the lowest emissions occurred when mineral N concentrations were below approximately $0.01 \text{ g N kg}^{-1} \text{ DM}$. As indicated in Fig. 5, soil moisture conditions influenced N_2O fluxes. Low soil moisture conditions that prevailed during spring of 2008 and 2009 were poorly represented by the TDR-based recordings of soil moisture (data not shown), since installation of the portable TDR probe into the dry soils was increasingly difficult and eventually impossible. Therefore, we illustrate the soil water conditions in the landscape using a running water budget (Fig. 6), calculated for each day as the accumulated precipitation during the previous 7 days minus the accumulated E_{ref} during the same period. A water budget of 0 mm on a given day would indicate balanced precipitation and evapotranspiration during the last 7 days. A threshold of -16 mm in the running water budget, equivalent to about 4 days of evapotranspiration without any precipitation, was assumed to be a limit defining the onset of dry soil conditions, primarily in the topsoil from where water would be evaporated first. Figure 6 (top) shows that springtime accumulated E_{ref} exceeded incoming precipitation from mid-April during both 2008 and 2009. During the intensive campaign of late April 2009, dry soil conditions prevailed. This was also temporarily the case during the equivalent period of 2008, whereas soil water content was higher in early April and much of May 2008. The timing of the dry and wet periods coincided closely with observed N_2O fluxes during the spring seasons of the two years. Hence, the unexpectedly low fluxes that were observed in 2009 were presumably caused by the

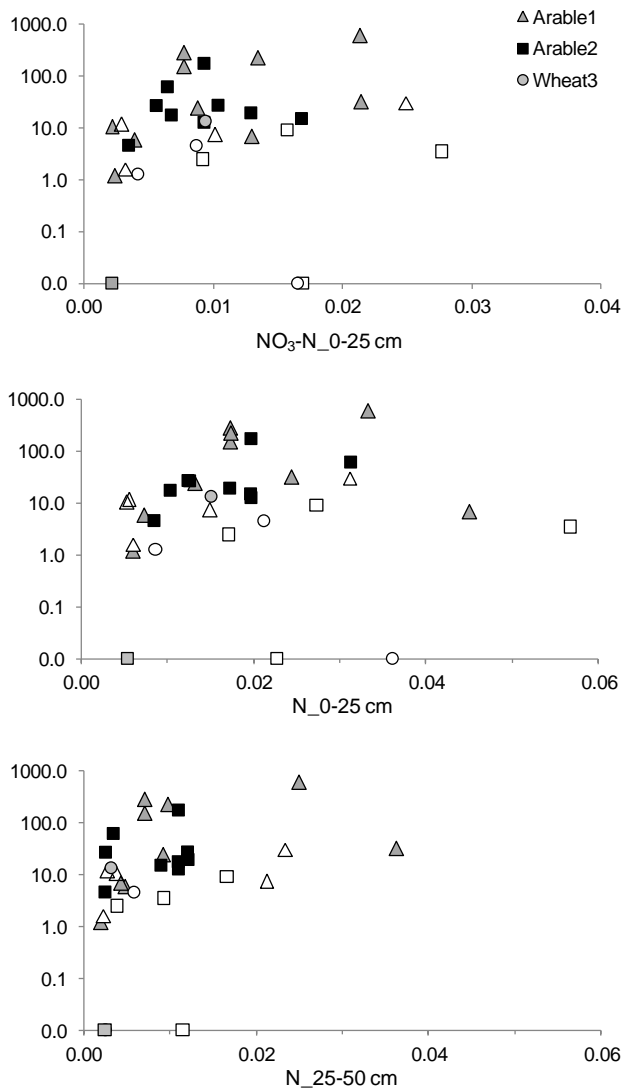


Fig. 5. N_2O fluxes ($\text{ng N m}^{-2} \text{s}^{-1}$) versus soil $\text{NO}_3\text{-N}$ or total mineral N content ($\text{g N kg}^{-1} \text{DM}$) for the 0–25 cm and 25–50 cm soil layers. Data are from 2008–2009 for three arable sites with CH2 type chambers. Open symbols represent measurements when soil conditions were “dry” (running water balance < -16 mm; see Fig. 6).

dry soil conditions. The amount of liquid supplied with the slurry was no more than 2.0–2.5 mm, hence barely contributing to re-wet the topsoil. Nor was the small-scale irrigation of CH1 chambers (14 mm) enough to counteract the soil water deficit of 20 mm on 26 April 2009 (Fig. 6). By contrast, high fluxes in late April and May 2008 were observed after re-wetting of the soil by 32 mm of rainfall to reach a positive soil water balance. When soil is re-wetted after a dry period, a pulse of N_2O has been observed by several authors due to increased nutrient availability for microbes (Mummey et al., 1994; Zebarth et al., 2008; Chirinda et al., 2010). Soil water

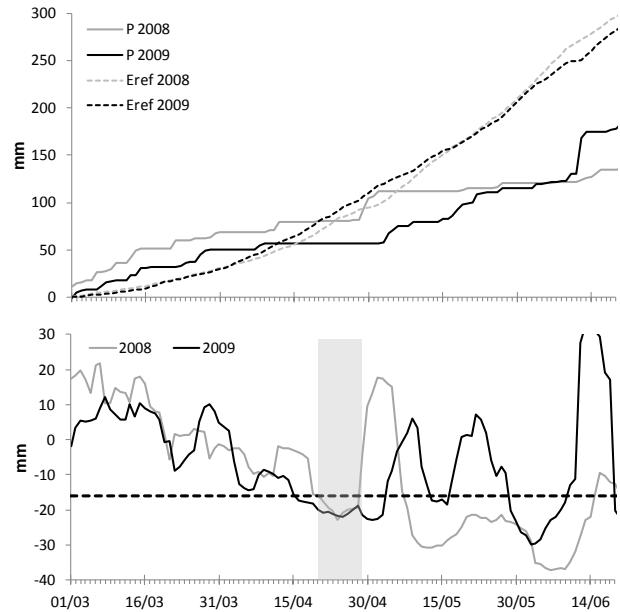


Fig. 6. Top: accumulated precipitation (P) and reference evapotranspiration (E_{ref}) during March–June of 2008 and 2009. Bottom: running 7-d water budget ($\Sigma P - \Sigma E_{\text{ref}}$) during March–June of 2008 and 2009. The timing of the 2009 intensive campaign is indicated by the shaded area. A water budget threshold has been indicated by the dashed line at -16 mm to identify periods of “dry soil conditions” when the running budget was below the threshold.

content influences N_2O emission from all types of soil. In general, aerobic microbial activity peaks at an intermediate water content and it has been reported that nitrification and associated N_2O production peak at around 60 % water-filled pore space or higher (Schjønnning et al., 2011), while optimum conditions for denitrification may occur at up to 60–90 % water-filled pore space (Linn and Doran, 1984; Hénault et al., 2005).

4.4 Annual nitrous oxide emissions

The emissions recorded at Arable1, Arable2, and Meadow⁰⁸ during the year from 15 October 2007 to 15 October 2008 were accumulated to give annual emissions (Table 4). The accumulation was done by linear interpolation of fluxes during periods of frequent measurements. For periods with more infrequent sampling, the environmental and management conditions prevailing at the time of a gap were considered: Fewer measurements were made at Arable1 due to late sowing of the crop in spring and autumn of 2008, leading to delayed re-installation of chamber frames. For the calculation of annual emissions, flux values for three missing dates in March and late August 2008 at Arable1 were estimated to remain at a low value (measured before the time gap) until the time of sowing and fertilization (Fig. 3). The estimated annual emission at the meadow ($1.2 \text{ kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$)

Table 4. Accumulated N₂O fluxes during the year from 15 October 2007 to 15 October 2008 at three sites equipped with CH₂ chambers. For the calculation of annual emissions, emission values for three missing dates at Arable1 were estimated to remain low until the time of sowing and fertilization. N-input is the direct fertilizer and manure N addition during the one-year period.

N ₂ O flux unit	Arable1	Arable2	Meadow ⁰⁸
kg N ₂ O-N ha ⁻¹ yr ⁻¹	17.5	5.5	1.2
Emission/N-input (%)	7.7	2.5	2.4*

* Excluding manure excreted by grazing heifers.

was comparable to fluxes measured in equivalent systems (0.66 kg N ha⁻¹ yr⁻¹, Ambus and Christensen, 1995; and 2–4 kg N ha⁻¹ yr⁻¹, Hefting et al., 2003). The estimated annual fluxes at Arable1 (17.5 kg N₂O-N ha⁻¹ yr⁻¹) and Arable2 (5.5 kg N₂O-N ha⁻¹ yr⁻¹) were larger than annual emission estimates in conventional arable cropping systems (0.9 kg N ha⁻¹ yr⁻¹, Chirinda et al., 2010; 1.7–2.9 kg N ha⁻¹ yr⁻¹, Laville et al., 2011; 1–3 kg N ha⁻¹ yr⁻¹, Kavdir et al., 2008). The reported annual estimates from the literature were generally obtained under controlled research grade experimental conditions, while the present results reflect the management, in terms of timing and historical fertilizer application rates, of a commercial Danish farmer. Kroon et al. (2008) also found large cumulative N₂O-N fluxes on a commercial dairy farm, and Denmead et al. (2010) measured large cumulative emissions at a commercial sugar cane farm. The differences between our results and the majority of results from the literature continue to be evident when comparing annual N₂O-N emissions as fractions of the recent N input to the crop growing cycle (Table 4). Our losses of 2.5–7 % of annual fertilizer and manure N input were high compared to many systems examined in the literature (0.5 %, Chirinda et al., 2010; 1.1–1.9 %, Laville et al., 2011; 0.7–2.4 %, Kavdir et al., 2008), and higher than the IPCC default N₂O emission factor of 1 % of N applied (IPCC, 2006). However, the emissions observed at our sites were gross fluxes influenced by historical N inputs and residue management and therefore not only attributable to the most recent N additions. Within agricultural systems, there can be substantial background N₂O emissions as demonstrated when N-application is omitted (Kavdir et al., 2008; Petersen et al., 2006).

5 Conclusions and implications for modelling

Our calculations of N₂O emissions from an agricultural landscape confirmed the influence of using non-linear regression vs. linear regression on the inherent flux underestimation with static chamber techniques. Rather than simple spreadsheet solutions for flux calculations, we recommend applying a non-linear regression tool that is neither complicated nor ambiguous, such as the freely available HMR software.

Estimated annual emissions of nitrous oxide from two arable fields (Arable1 and Arable2) that had been fertilized with mineral fertilizer and manure were large (17 kg N ha⁻¹ yr⁻¹ and 5.5 kg N ha⁻¹ yr⁻¹, respectively) during a year when soil water conditions were favourable for N₂O production during the first month following fertilizer application in spring. Peak emissions during that first year reached 200–600 ng N m⁻² s⁻¹, with the largest emissions occurring after spring fertilization. By contrast, emissions were found to be consistently and relatively low (< 20 ng N m⁻² s⁻¹) at a range of land use types when monitored during two weeks of the spring/fertilization period of the following year. The latter period coincided with dry soil conditions, and the modest fluxes recorded, even after slurry application, were attributed to these dry environmental conditions. Consequently, the intensive monitoring period failed to demonstrate clear trends in N₂O emissions as a function of position in the landscape, either topographical or according to land use type.

Models of organic matter turnover and microbial activity in arable soils use different empirical relationships for describing the effects of soil temperature and soil moisture conditions (Rodrigo et al., 1997; Bauer et al., 2008). The findings in our landscape that the environmental conditions and soil dryness controlled or limited N₂O emissions confirm the importance of the water response function for modelling.

Generally, models may have difficulties in capturing responses or effects of different fertilizer inputs on N₂O emissions (Frolking et al., 1998; Chatskikh et al., 2008). Modelling emissions of N₂O from field applied with manure, especially farmyard manure, is a particular challenge due to the heterogeneity in the distribution of O₂ supply and demand within the soil. Within the Bjerringbro landscape, we found large N₂O emissions from an arable field after the application and incorporation of farmyard manure and attributed the N₂O losses to lumps of manure acting as hotspots within the field soil. These emissions may be difficult to predict using even the best available models (Chen et al., 2008; Farquharson and Baldock, 2008).

Acknowledgements. This research was funded by the European Commission (NitroEurope Integrated Project, Contract 017841.00 of the EU Sixth Framework Programme for Research and Technological Development) and the Danish Ministry for Food and Agriculture. The authors would also like to thank COST Action 729 and the ESF-NinE programme for co-funding the joint measurement campaign and a workshop at the Bjerringbro landscape during April 2009. Thanks are also due to COST Action ES 0804 and the TÁMOP 4.2.1./B-09/1/KMR-2010-0003. We thank J. Bienkowski, K. Janku, A. Pogány, A. Bordás, B. Durand, and A. Cobena for assisting in the field during the intensive campaign. Finally, we thank the three local farmers at Bjerringbro for letting us access and make measurements on their fields and for allowing us to decide the exact days of slurry application during April 2009.

Edited by: S. Reis

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