Biogeosciences Discuss., 8, 11941–11979, 2011
www.biogeosciences-discuss.net/8/11941/2011/
doi:10.5194/bgd-8-11941-2011

Biogeosciences Discussions

Nitrous oxide emissions at the landscape scale: spatial and temporal variability

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Received: 2 December 2011 – Accepted: 5 December 2011 – Published: 14 December 2011

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

Abstract

Nitrous oxide (N_2O) 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 N_2O emissions, measured using static chambers. Measurements were done 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. N_2O emissions during the spring 2009 period 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, grassland, meadow, and wetland. Slurry application to wheat fields resulted in short-lived two-fold increases in emissions. The moderate N_2O fluxes and their moderate response to slurry application were attributed to dry soil moisture conditions due to the absence of rain during the four previous weeks. Measured cumulated annual emissions from two arable fields that were both fertilized with mineral fertilizer and manure were large (17 kg N_2O-N ha^{-1} yr^{-1} and 5.5 kg N_2O-N ha^{-1} yr^{-1}, respectively) during the previous year when soil water conditions were favourable for N_2O production during the first month following fertilizer application, confirming the importance of the climatic regime on N_2O fluxes.

1 Introduction

Atmospheric nitrous oxide (N_2O) 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 N_2O, in order to better predict and possibly mitigate future emissions by improved management of land and resources.

Approximately 65 % of atmospheric emissions of N_2O originate from soils (Smith 2004). N_2O is produced in soils as an intermediate in the two contrasting microbial

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processes, autotrophic nitrification and heterotrophic denitrification. The contribution of these two processes to N_2O emissions vary with climate, soil conditions and soil management (Skiba and Smith, 2000). Land use type, and changes in the crop rotation are particularly important in determining the rate of N_2O 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 N_2O emission from soils (Kavdir et al., 2008). N_2O fluxes can be sporadic and short-lived, depending on the environmental conditions of the soil, particularly temperature and moisture content (Smith et al., 1998; Frolking et al., 1998). Schaufler et al. (2010) observed a positive correlation of N_2O fluxes with soil moisture but found no significant relationship between emissions and N fertilization or N deposition level when lumping together all soil moisture conditions. In short, several factors influence N_2O 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 N_2O flux dynamics focused on ecosystems or landuse types such as forest (Pihlatie et al., 2005; Kesik et al., 2005), wetland/organic soils (Maljanen et al., 2003), grasslands (Flechard 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. Pattey 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 N_2O 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 N_2O emissions due to infrequent sampling was less than the uncertainty due to spatial variability of the sampling sites.

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The objective of the current study was to examine the major drivers for N_2O emissions in a real agricultural landscape. Chamber measurements made over a range of time (20 months) and space (10 sites) at a Danish landscape focused on identifying 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 to three wheat fields on N_2O emissions. Based on the experiences during the first growing season, we expected to find significant fluxes of N_2O following the targeted springtime manure application. The 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 (Duretz et al., 2011).

2 Materials and methods

2.1 Landscape and duration of experiment

The study landscape 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 utilized for intensive agricultural production with fields that are fertilized with a combination of synthetic fertilizer and animal manure (Wohlfart et al., 2011). The climate of the region, Central Jutland, is coastal temperate with an 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,

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respectively. The experiment took place during September 2007–April 2009. During the first full year, measurements focused on three permanent sites (Arable1, Arable2 and Meadow[a]; 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 permanent 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 the individual fields studied. Agricultural management was recorded through farmer interviews, by consulting agronomic advisory services, and by observations made by the investigators. 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 experimental and measurement activities. An overview of fields and fertilization intensities during 2007–2009 is given in Table 1.

2.2 Chamber measurements

Four chamber types, CH1-CH4, were applied for measuring fluxes of N_2O within the Bjerringbro landscape (Table 2). All were two-part static chambers, and chamber installation sites are 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 bimonthly intervals at all times of the year, 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 located in groups in the same fields (Table 2). For the long-term installations in Arable1 and Arable2, the CH2 steel frames were semi-permanently installed and only removed for harvest and seedbed preparation. On each sampling day, four 10 mL gas samples were taken per chamber over the period of 50–70 min (CH1, CH3) or 80–90 min (CH2). Gas sampling from CH4 was 3 samples at 15 min intervals. Samples were stored in pre-evacuated 6 mL glass vials for later analysis. N_2O concentrations in gas samples were subsequently

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analysed using gas chromatographs 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. 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).

N_2O 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). HMR analyses, when appropriate, 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 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 samplings over time was accepted only when yielding a statistically significant regression according to its p-value being smaller than 0.15.

2.3 Meteorology and soil

Meteorological records (hourly and daily time resolution) were available over the entire period from a meteorological station located in a similar agricultural landscape 25 km east of the area. During the intensive campaign in 2009, meteorological variables (radiation, air, and soil state variables incl. soil water content in the topsoil) were measured with a high time resolution (15 min) using a similar instrument setup to that described in Horvath et al. (2005) and Meszaros et al. (2009). During the campaign a mast equipped with instruments for Bowen ratio measurements (Campbell Scientific, Loughborough, UK) of heat fluxes was also installed. Soil water content was measured using a portable TDR and a 0.25 m probe near chamber locations. Measurement frequency was the same as that of the N_2O flux measurements, however at times the portable TDR probes could only be inserted with difficulty, or could not be inserted at all, due to dry soil conditions. Soil for inorganic N content analyses was sampled near chamber positions and on a subset of the dates when N_2O fluxes were measured. On each sampling date, five soil cores were randomly sampled to 25 cm depth at each site and pooled to enable a representative composite subsample. The same procedure was carried out for soil sampled from the soil depth interval 25–50 cm. Samples were kept cool (<10 °C) and transported to the laboratory. Samples were frozen until analysis for NO_3 and NH_4 content which was made according to Mutegi (2010) for samples extracted near chambers CH2 and using the same procedure as Vilain et al. (2010) for samples extracted near chambers CH1.

3 Results

3.1 Meteorology

Meteorological conditions during the 20-month period (2007–2009) were not unusual. The two winters were mild with only a few frosts. Springs of 2008 and 2009 were both characterised by relatively dry conditions during mid to late April–May. In our rain-fed agricultural landscape this caused soils to dry out. Accordingly, continuous measurements of soil water content at 5 cm depth of field Wheat2 showed a steadily decreasing volumetric water content from 15 % to 10 % during the intensive measurement period (19–29 April 2009). Actual evapotranspiration, measured in the same field using the Bowen Ratio method, was 60–70 % of reference evapotranspiration during 19–29 April 2009 (ratio calculated as daily accumulated gap-filled latent heat flux data divided by daily reference evapotranspiration derived from meteorological station data and the Makkink equation (de Bruin and Lablans, 1998)).

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

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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 were co-located in fields (Table 2). There was however no attempt to duplicate chamber measurements, so a direct comparison between chambers and fluxes was not possible, taking spatial variability into account. Figure 3 shows measured fluxes in field Wheat2, using three types of chambers. N_2O fluxes were within the same low range, and fluxes of chambers CH1 and CH3 increased slightly after slurry application. At the same time chambers CH4 generally estimated smaller fluxes, which were unaffected by the slurry application. Chambers CH1 and CH2 were co-located at field Wheat3 and generally agreed on the magnitude of N_2O fluxes (data not shown).

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 no-flux, and CVs were typically low when fluxes were generally high. During the 2009 intensive campaign, campaign CVs per chamber type and site ranged between 60 % and 140 %. Practically all measured fluxes were positive.

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) over a gentle slope (Wh1Slp) of the wheat field to the footslope in the small stream valley (Meadow[b]). The first two transect points had a similar soil texture with a clay content of 7–8 % and a soil organic matter content of 3 % in the depth interval 15–30 cm. The humus type soil in the valley meadow had a larger clay and organic matter content at the 15–30 cm depth (16 % and 15 %

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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). The C/N ratio was around 12 for both depths. Soil water content by weight at 5–15 cm was considerably higher in the valley (48 %) than in the wheat field (9 %) on 21st April 2009. At the same date, mineral N contents were higher in the valley than in the wheat field: NO_3-N and NH_4-N concentrations at the wheat field were 4.2 and 4.6 mg N kg^{-1} DM, respectively, while NO_3-N and NH_4-N concentrations in the valley were 25 and 38 mg N kg^{-1} DM, respectively. The recorded N_2O fluxes in 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 (Fig. 4).

The effect of land use and land management on N_2O fluxes was studied using the full year of measurements at sites Arable1, Arable2, and Meadow[a]. Differences in management (Table 1) were mainly due to fertilization since the arable fields were managed with intensive organic and inorganic fertilizer while the meadow was occasionally grazed by heifers and lightly fertilized with inorganic fertilizer. Fluxes at the meadow were low at all times during the year 2007–2008 (<20 ng N m^{-2} s^{-1}) while fluxes at the arable fields had an annual pattern with high emissions, up to 600 ng N m^{-2} s^{-1}, during spring and early autumn.

During the intensive campaign of 2009, more types of arable fields were studied in addition to areas with more extensive land use. Figure 5 shows observed fluxes at four sites, ranging in fertilization intensity from unmanaged wetland to winter oilseed rape grown in a field (Arable1) that had been intensively fertilized for years. All observed fluxes were low, below 20 ng N m^{-2} s^{-1}. There was a trend that unmanaged areas or areas not recently cultivated emitted the lowest fluxes. Fertilized fields emitted somewhat higher fluxes (wheat2 and Arable1), however the fertilized winter barley (Arable2) had low emissions.

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3.4 Temporal variability

During the 2009 campaign, fluxes increased when wheat fields were fertilized with pig slurry (Figs. 3 and 6) with about a doubling of the low emissions although the effect was short-lived (2–3 days). Since there was no precipitation during the campaign, irrigation (approx. 14 mm) at the soil surface within chamber type CH1 was performed on 26th April 2009. There was no clear effect of irrigation in terms of changed emissions at any of the irrigated sites (Figs. 3 and 6) except a possible small emission peak following the irrigation.

Temporal variability was most obvious when considering 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 fluxes of N_2O with fluxes at Arable1 exceeding 600 ng N m^{-2} s^{-1} and fluxes at Arable2 exceeding 150 ng N m^{-2} s^{-1} (Fig. 7). Emissions were low over the summer and increased again in September 2008 at Arable1 when a winter oilseed rape crop had been established. It was notable that compared to fluxes in 2008, all fluxes recorded during spring 2009 were small.

Spring time high emissions in 2008 were recorded after sowing and fertilization of the crops (Table 1). At Arable2 the highest flux was observed after the completion of two split mineral fertilizer and a slurry application, and at Arable1 fluxes began to increase significantly after the application of farm yard manure in spring. The peak in emissions in autumn at Arable1 appeared after cultivation and sowing of the winter oilseed rape in late August followed by slurry application on 1st September 2008.

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

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may 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 more low-technology methods were 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 fluxes measured with CH2 chambers did not appear to differ from other chambers (statistical test not relevant due to spatial variability effects). There was a trend that the small CH4 chambers did not resolve the temporal variability as well as the larger chambers: Their average flux did not indicate a response to slurry application (Fig. 3). One reason is that observations at the field (Wheat2) revealed how slurry was applied, using trailed hoses, into only four of the ten 10 cm diameter chamber 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). Our findings show that emission estimates were increased by 16–32 % (depending on period taken into account) when using the HMR analysis tool, which was in accordance with recent experience. Kroon et al. (2008) and Thomsen et al. (2010) reported how accounting for non-linearity increased N_2O flux estimates by approximately 100 % and 25–65 %, respectively.

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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. In the following discussion we will refer to emissions at a given site and day as being "low" when the average measured flux did not exceed 20 ng N_2O-N m^{-2} 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. Previous studies suggested that topography influences N_2O dynamics, with the 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., 2011). The higher fluxes at the footslope were often associated with the relatively higher water-filled pore space in the soil at the bottom of the slope (Vilain et al., 2010). 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 at 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

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filled pore space (WFPS) averaged 50 % in the meadow and 30 % at the slope and shoulder positions under these specific (dry) spring time 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 conditions and gradually lowered to approximately 40 cm below ground during summer conditions. 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 compared with NO_3; NH_4 concentrations were 0.020–0.045 g NH_4-N kg^{-1} 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 confirmed a NH_4/NO_3 ratio above 1.0 at 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 were drained by tiles (Wohlfart et al., 2011), and 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 at the meadow measurement positions. However, the measurement setup at the meadow (five flux chambers grouped at Meadow[a] location during 2007–2008 and five other chambers at Meadow[b] 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

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conclude, in accordance with Vilain et al. (2010), that N_2O emissions at riparian buffer areas are highly site-specific and depending on the local hydrology and N input.

4.2.2 Arable land

With the low emissions encountered at the meadow and wetland sites, N_2O emissions were mainly governed by land-use, i.e. fertilized arable fields versus wetland or meadow that were not or only marginally fertilized (Table 1 and Fig. 5). Within-season management at the field scale was equally important since fluxes were highest after crop establishment spring and autumn and fertilization in spring and autumn (Fig. 7). The importance of land use and management is in accordance with findings of other investigators at field or plot scale. The review by Machefert 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. The response to spring fertilization was similar in the organic and conventional systems despite a lower N application (>100 kg N ha^{-1}) rate in the organic systems than in the conventional system (165 kg N ha^{-1}). Kavdir et al. (2008) found that the response of N_2O emission to fertilization and crop types 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.

The highest emissions during the experimental period were found at Arable1 and Arable2. The maximum levels recorded at Arable2 (up to 180 ng N m^{-2} s^{-1}; Fig. 7) were comparable to other high emissions measured in arable systems (Laville et al., 2011; Petersen et al., 2011) but were higher than maximum fluxes measured by Chirinda et al. (2010, 21 ng N m^{-2} s^{-1}) and Mutegi et al. (2010, 14 ng N m^{-2} s^{-1}) during the 2008 spring season at cereal crop sites located close (20 km) to the Bjerringbro

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landscape. Also during the 2008 spring season, Thomsen et al. (2010) observed maximum emissions of 50 ng N m^{-2} 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 (156–617 ng N m^{-2} s^{-1}, Fig. 7) seemed to be extraordinary, although Kroon et al. (2008) observed large fluxes after manure and fertilizer application at an intensively managed grassland site. The spring barley at Arable1 was fertilized with cattle farmyard manure (FYM) and mineral fertilizer in spring 2008 and the field was treated again with manure before sowing of 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 fertilizing with farm yard manure or deep litter that is incorporated into the soil by ploughing, typically within few hours after spreading to minimize NH3 emissions, the manure can be unevenly distributed on the soil surface (Hansen, 2004) and later in the topsoil. Lumps of farm yard manure, high in nutrient concentration, 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 potential for high 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. On the contrary, dissolved C and N in organic slurries with a much lower DM content are more homogenously 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 examination. Thorman et al. (2007) applied pig and cattle FYM to cereal stubble and bare ground, 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 that 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 by incorporation of cattle or pig FYM into 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 conditions, 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 over the growing season. N_2O emission rates, peaking in spring and autumn 2008 after the time of fertilization, were positively correlated with soil mineral N content when disregarding times of the year when conditions were unfavourable for N_2O production. In Fig. 8, dry soil conditions (as defined in Fig. 9) 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. 8 for NO_3 content in the topsoil, and for total N content in the topsoil and the 25–50 cm intervals, are scattered, but still indicate that the largest emissions were obtained at the highest mineral N contents. As indicated in Fig. 8, 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 at the landscape using a running water budget (Fig. 9), calculated for each day as the accumulated precipitation within the previous 7 days subtracted by the accumulated reference evapotranspiration, E_{ref}, during the same period. E_{ref} was calculated based on the Makkink equation (de Bruin and Lablanc, 1998) that represents well the reference evapotranspiration under Danish conditions (Kjaersgaard et al., 2008). A water budget of 0 mm on a given day would indicate a balance between recent precipitation and evapotranspiration. A threshold of −16 mm in the running water budget, equivalent to 4–7 days of evapotranspiration

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without any precipitation, was assumed a limit defining the onset of dry soil conditions, primarily in the topsoil from where water would be evaporated first. Figure 9 (above) shows that spring-time accumulated reference evapotranspiration 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, while early April and much of May 2008 were more abundant in soil water. The timing of the dry and wetter periods coincided closely with observed N_2O fluxes (low versus high, respectively) during the spring seasons of the two years. Hence, the unexpectedly low fluxes that we found in 2009 were presumably restricted by the 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 soil. Neither 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. 9). 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; Chirinda et al., 2010). Franzluebbers et al. (2000) reported that re-wetting of dry soil caused increased carbon mineralization in soils after 3 days, causing N immobilization to meet the demands of the active soil microbial population. On the other hand further incubation showed that after 3 days N mineralization increased with increasing C mineralization. Therefore, in their study N_2O emission in August 2004 was, not were greater than in previous years due to mineralization of N and/or denitrification caused by rainfall.

Soil water 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 (WFPS) or higher (Schjønning et al., 2011), while optimum conditions for denitrification may occur at up to 60–90 % water-filled pore space (Linn and Doran, 1984). However, WFPS is really a proxy for the prevailing soil water and aeration conditions.

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Mutegi et al. (2010) and Petersen et al. (2008) suggested that the relative soil gas diffusivity (R_{diff}) is a better determinant for N_2O fluxes since R_{diff} considers the soil air phase and the pore continuity. Schjønning et al. (2003) found net nitrification in three texturally contrasting soils to correlate with gas diffusivity in bulk soil under wet conditions. They found that net nitrification peaked at a lower value of R_{diff} for a clayey soil than for a sandy soil, so R_{diff} was not a universal predictor. Microbial aerobic and anaerobic activities rely on diffusion in air, and the relative gas diffusivity influences the potential for N_2O production and transport in the soil. Aerobic conditions are dominant for longer periods in sandy soils and O_2 limitation may not occur, favouring N_2O production via denitrification. Sandy soils also dry out sooner than loamy soils and their coarse texture and aeration promote nitrification and associated N_2O production. In a soil incubation study, van den Heufel et al. (2009) found that nitrate addition to the soil failed to increase total denitrification or net N_2O production. N_2O production was similar in all soils samples, independent of their origin from high- or low-emission soils, indicating that environmental conditions (including physical factors like gas diffusion) rather than the local microbial community composition governed N_2O emission rates.

4.4 Annual nitrous oxide emissions

The emissions recorded at Arable1, Arable2, and Meadow[a] during the year from 15th October 2007 to 15th October 2008 were accumulated to give annual emissions (Table 3). 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 the missing dates at Arable1 were estimated to remain low until the time of sowing and fertilization (Fig. 7). The estimated annual emission at the meadow (1.2 kg N_2O-N ha^{-1} yr^{-1}) was comparable to fluxes measured in equivalent systems (0.66 kg N ha^{-1}y^{-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 N_2O-N ha^{-1} yr^{-1}) and Arable2 (5.5 kg N_2O-N ha^{-1} yr^{-1}) were larger than annual emission estimates in conventional arable cropping systems

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($0.9\,kg\,N\,ha^{-1}\,yr^{-1}$, 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 cumulated N_2O-N fluxes at a commercial dairy farm. The differences continue to be evident when comparing annual N_2O-N emissions as fractions of the N input to the crop growing cycle (Table 3). Our losses of 2.5–7 % of fertilizer and manure N input are high compared to the 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 significantly higher than the IPCC default N_2O emission factor of 1 % of N applied (IPCC, 2006). It should be noted that within agricultural systems there can be substantial N_2O emissions even when N-application is omitted (Kavdir et al., 2008; Petersen et al., 2006).

5 Conclusions and implications for modelling

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 N_2O production during the first month following fertilizer application in spring. 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 environmental conditions. Consequently, the intensive monitoring period failed to demonstrate clear trends in N_2O 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

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conditions (Rodrigo et al., 1997; Bauer et al., 2008). The findings in our landscape that the environmental conditions and soil dryness controlled or limited N_2O emissions, confirmed the importance of the water response function for modelling. Recent studies point to the applicability of the soil relative gas diffusivity (R_{diff}) as a descriptor of conditions that are optimal for N_2O production; however R_{diff} does not seem to be a universal descriptor (Schjønning et al., 2011).

Generally models may have difficulties in capturing responses or effects of different fertilizer inputs on N_2O emissions (Frolking et al., 1998; Chatskikh et al., 2008). Modelling emissions of N_2O from field applied manure, especially farm yard manure, is a particular challenge due to the heterogeneity in the distribution of O_2 supply and demand within the soil. Within the Bjerringbro landscape we found large N_2O emissions from an arable field after the application and incorporation of farm yard manure and attributed the N_2O 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 (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 further wish 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. Finally, we thank J. Bienkowski, K. Janku, A. Pogány, A. Bordás, B. Durand, and A. Cobena for assisting in the field during the 2009 intensive campaign.

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Table 1. Field names, crops and fertilization schemes at sites studied during 2007–2009.

				2008		2009	
Field name	Area (ha)	Crop 2008/2009	Fertilizer type	kg N ha^{-1}*	Date	Kg N ha^{-1}*	Date
Arable1	8.5	Spring barley/winter oilseed rape	Synthetic	43	3/5/2008	55+78	1/2/2009; 1/4/2009
			Manure (cattle)	186	15/3/2008	53	1/9/2008
Arable2	12.7	Winter wheat/winter barley	Synthetic	43+43	17/3/2008; 22/4/2008	55+88	1/2/2009; 1/4/2009
			Slurry (pig)	136	20/3-2008	0	–
Wheat1	9.1	Oats/winter wheat	Synthetic	76	1/4/2008	50+50	1/3/2009; 1/4/2009
			Slurry (pig)	–	–	87	23/4/2009
Wheat2	11.6	Winter wheat/winter wheat	Synthetic	72	5/5/2008	72	5/5/2009
			Slurry (pig)	135	1/4/2008	112	22/4/2009
Wheat3	4.1	Oats/winter wheat	Synthetic	76	1/4/2008	50+50	1/3/2009; 1/4/2009
			Slurry (pig)	–	–	87	27/4/2009
Grass	3.5	Grass-clover in rotation	Synthetic	100+150	10/4/2008; 15/6/2008	100+150	10/4/2009; 15/6/2009
			Slurry (cattle)	164**	01/03/2008	164**	15/02/2009
Wetland	1.0	Mixture of herbs	None	0	–	0	–
Meadow	6.5	Permanent grass	Synthetic	50**	5/5/2008	44**	1/5/2009

*kg total N

**Excluding manure deposited by grazing cattle

Table 2. Chamber types and sites or fields where they were applied in the Bjerringbro landscape.

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.	2009: Wheat1, Wheat2, Wheat3, Grass, Wetland, Meadow[b]
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[a] 2009: Arable1, Arable2, Wheat3
CH3	0.24	65	10	Plastic frame and plastic cover inserted into water-filled rim of frame. No vent, no mixing.	2009: Wheat1, Wheat2
CH4	0.007	0.5	10	Plastic cylinder and plastic cover. No vent, no mixing.	2009: Wheat2

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Table 3. Accumulated N_2O fluxes during the year from 15 October 2007 to 15 October 2008 at three sites with CH2 type chambers. For the calculation of annual emissions, emission values for missing dates at Arable1 were estimated to remain low until the time of sowing and fertilization.

N_2O flux unit	Arable1	Arable2	Meadow[a]
kg N_2O-N ha^{-1} yr^{-1}	17.6	5.5	1.2
Emission/N-input (%)	7.7	2.5	2.4*

*Excluding input from grazing heifers

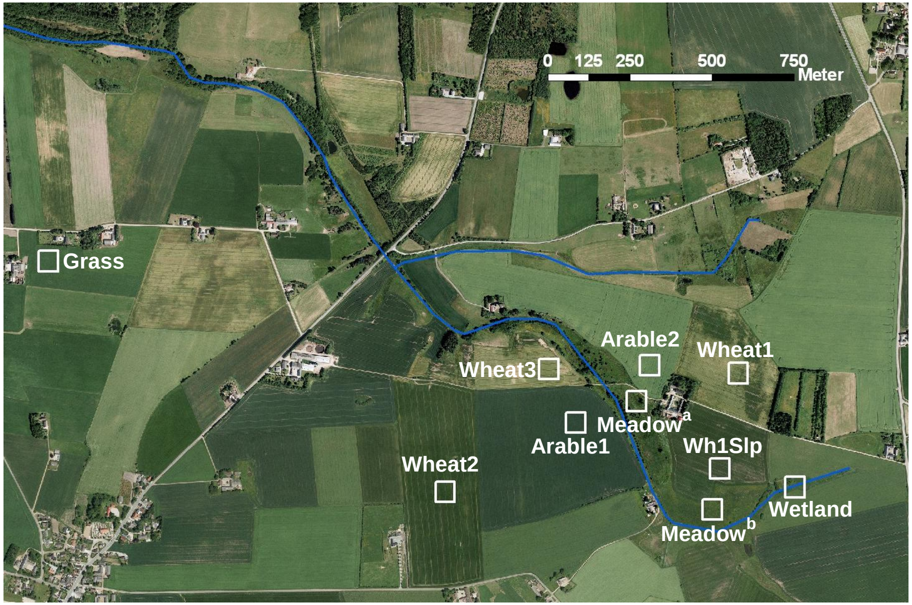

Fig. 1. Aerial photograph showing the ten locations for manual chamber measurements within the Bjerringbro landscape. Arable1 and Arable2 were studied throughout the period 2007–2008 and the 2009 campaign, Meadow[a] was studied during 2007–2008, and remaining sites were only monitored during the intensive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

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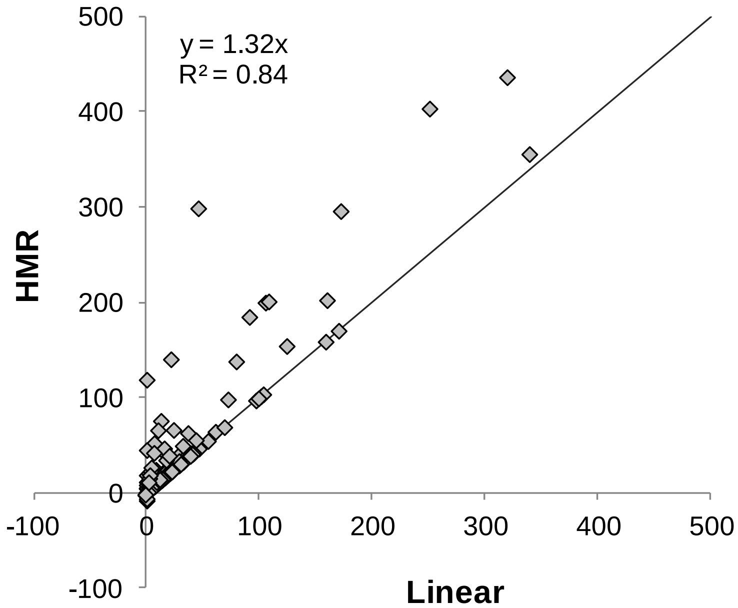

Fig. 2. Calculated N_2O fluxes (ng N m^{-2} s^{-1}) using the HMR tool versus calculated fluxes using a linear regression for the analysis of chamber concentrations. Figure shows all significant individual chamber fluxes below 500 ng N m^{-2} s^{-1} ($n = 364$). Four individual chamber fluxes were larger than 500 ng N m^{-2} s^{-1}. The line is the 1:1 line; the R2 value is for a linear regression: HMR flux = 1.32 × Linear flux.

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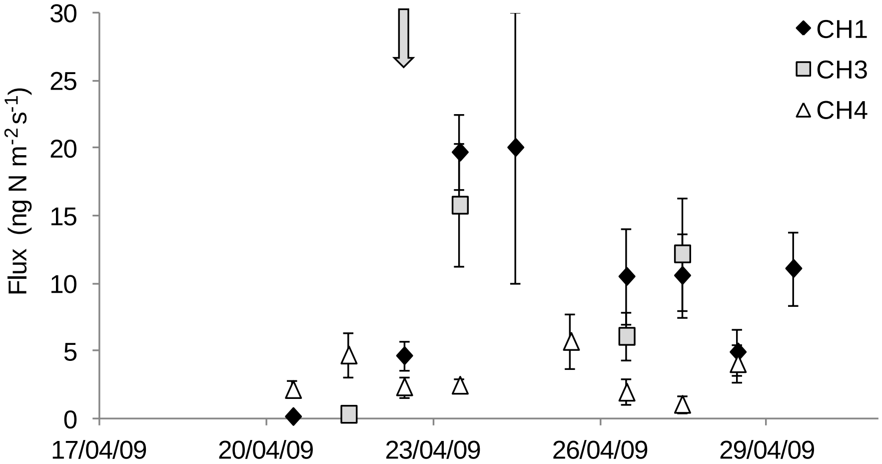

Fig. 3. Measured N_2O fluxes in field Wheat2 using three types of chambers. Error bars quantify the standard errors. The arrow indicates the timing of slurry application at the field.

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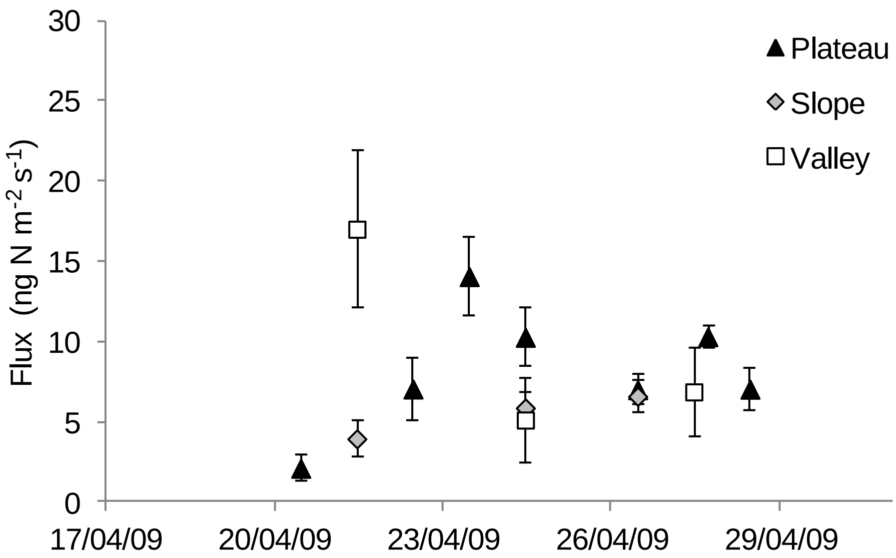

Fig. 4. Measured N_2O fluxes in a sloping transect beginning in field Wheat1 (plateau), descending to Wh1Slp (slope) and ending at Meadow[b] (valley). For locations refer to Fig. 1. Error bars quantify the standard errors. Slurry was applied to the wheat (plateau and slope) on 23 April 2009.

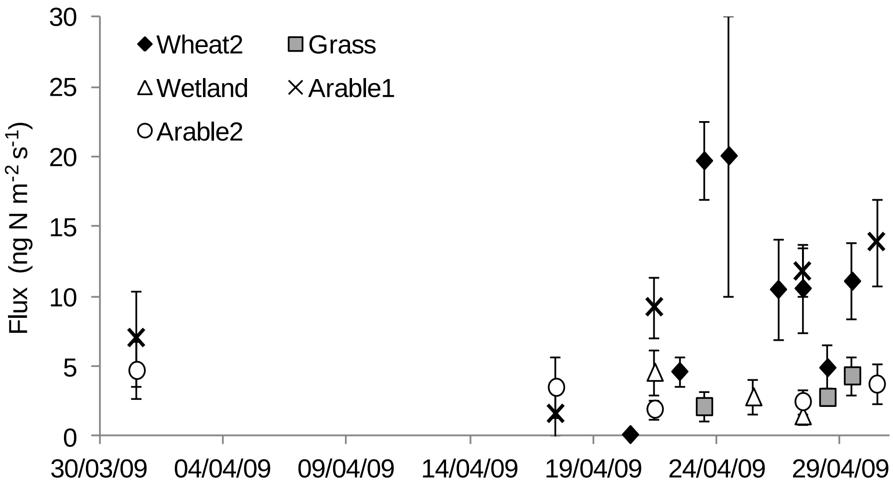

Fig. 5. Measured N_2O fluxes for different land use types. Error bars quantify the standard errors. Slurry was applied to the wheat field on 22 April 2009 and chambers in the wheat field were irrigated (14 mm) on 26 April 2009.

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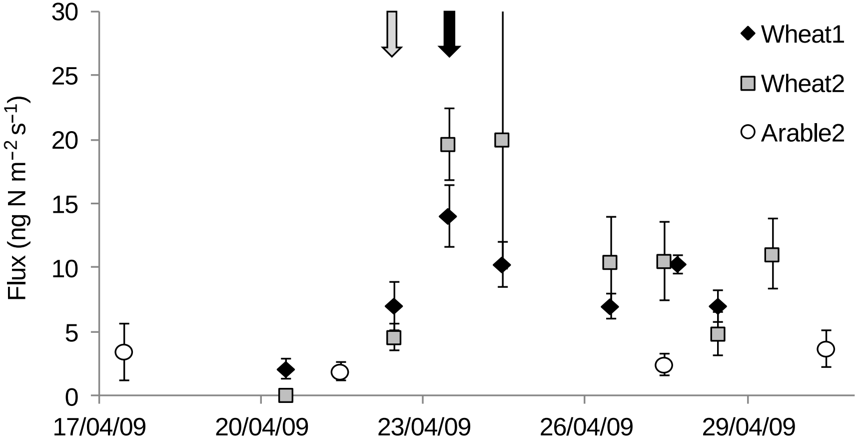

Fig. 6. Measured N_2O fluxes at two wheat fields and a winter barley field (Arable2) during the 2009 campaign. Error bars quantify the standard errors. Arrows indicate the timing of slurry application at Wheat2 followed by Wheat1. The barley field (Arable2) was not fertilized with slurry during the period. Chambers at both wheat fields were irrigated (14 mm) on 26 April 2009.

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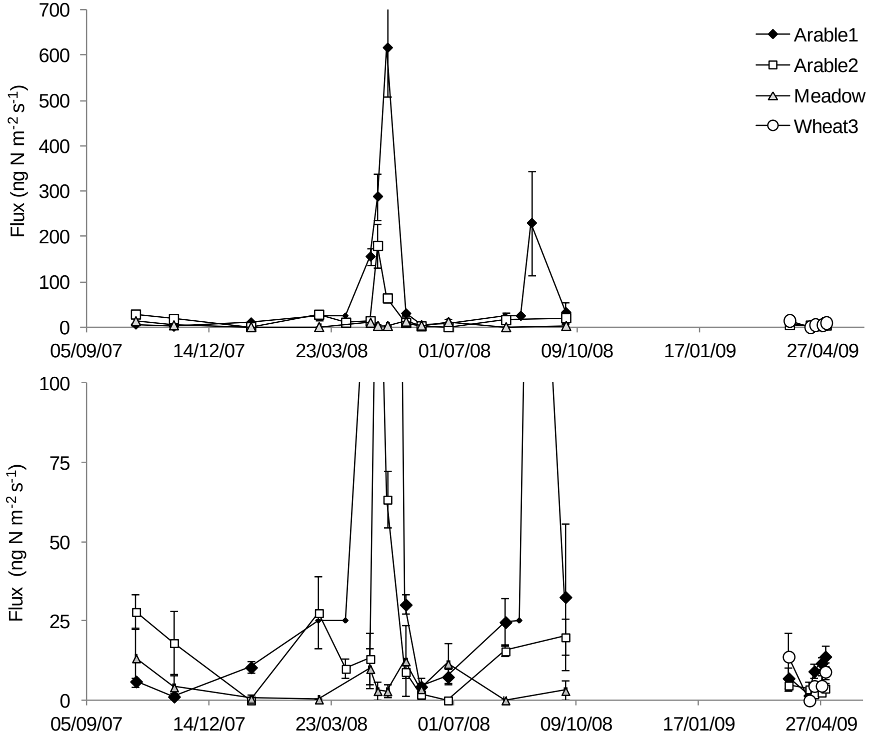

Fig. 7. Measured N_2O fluxes for the fields monitored by CH2 chambers, displayed at two different flux axes. Error bars quantify the standard errors. Chambers located at Meadow[a] during 2007–2008 were operated at Wheat3 during the 2009 campaign. Fewer measurements were made at Arable1 due to late sowing of the crop in spring and autumn of 2008, leading to delayed re-installation of chamber frames; hence small symbols at Arable1 in March 2008 indicate estimated emission values for the missing dates at Arable1.

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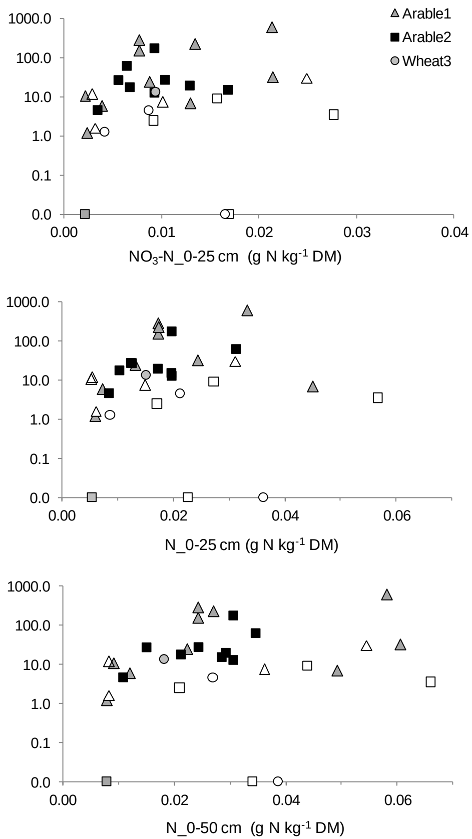

Fig. 8. N_2O fluxes (ng N m^{-2} s^{-1}) versus soil NO_3-N or total mineral N content. Data are from 2008–2009 for three arable sites operated with CH2 chambers. Open symbols represent measurements when soil conditions were "dry" (running water balance < −16 mm, see Fig. 9).

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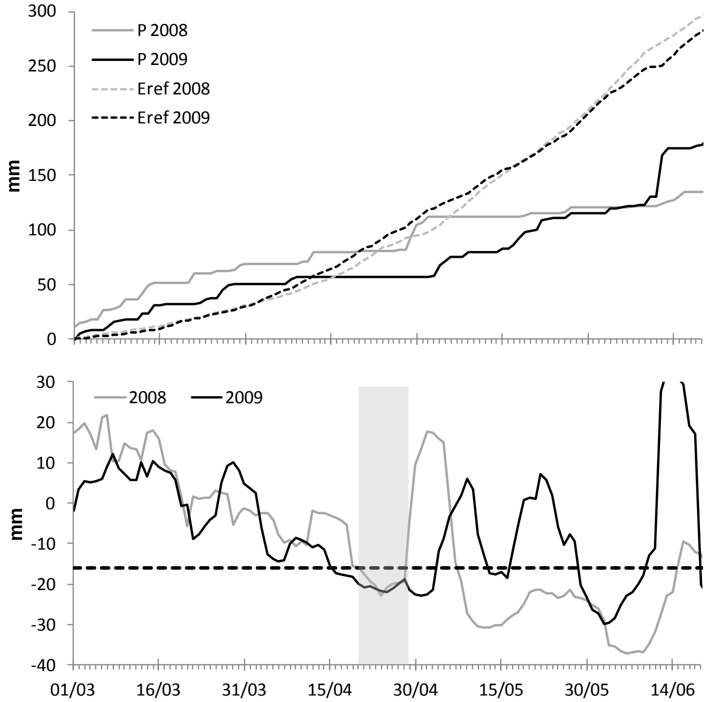

Fig. 9. Above: Accumulated precipitation (P) and reference evapotranspiration (E_{ref}) during March–June of 2008 and 2009. Below: Running 7-d water budget ($\Sigma P - \Sigma E_{ref}$) during March–June of 2008 and 2009. The timing of the 2009 intensive campaign has been indicated by a shaded area. A water budget threshold is indicated at −16 mm to identify periods of "dry soil conditions" when the running budget is below the threshold.

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