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Nitrous oxide emissions from five fertilizer treatments during one year – High-frequency measurements on a Swedish Cambisol

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ABSTRACT

Nitrous oxide (N_2O) is a strong greenhouse gas, and the emissions from managed soils are increasing. Emissions of N_2O are highly variable in time and space, and there are potential triggers for emission peaks both in crop season and no-crop season. The aim of this study was to compare how fertilizer treatments, differing in rate and source of nitrogen (N), influence direct N_2O emissions from soil, in crop season as well as in no-crop season, with the use of automated, high-frequency chamber measurements. Emissions were measured from cereal production on a Swedish clay-rich soil fertilized with biogas digestate, pig slurry and two levels of mineral N, as well as from control plots receiving no fertilizer N. The results showed that N_2O emissions per unit area were low in all treatments, compared to other studies. Emissions from the treatment with mineral fertilizers at recommended rates were similar to the emissions from the control (0.65 and 0.48 kg N_2O-N ha^{-1} yr^{-1}, respectively). One-year cumulative emissions from a mineral N input rate 50 % higher than recommended were about three times higher than the control. Emissions of N_2O from the pig slurry and biogas digestate treatments per unit area were of the same magnitude as from the high mineral N treatment. While the emissions from the high mineral N treatment were associated with elevated concentrations of nitrate in the drainage water, the high emissions from the organic fertilizer treatments were probably a result of large input of ammonium and degradable organic matter both in the year studied and in the preceding year. Most (approximately 75 %) of the N_2O emissions occurred between harvest in autumn and sowing in spring, mainly in periods of freeze-thaw cycles. The relative differences between treatments were roughly the same during crop season and no-crop season. This study concludes that it is possible to combine high yields with very low N_2O emissions - even on a clay soil in a semi-humid climate - when using mineral fertilizers at recommended rates.

1. Introduction

Nitrous oxide (N_2O) is a strong greenhouse gas, representing 5 % of the total radiative forcing caused by anthropogenic greenhouse gas emissions (Pachauri et al., 2014). Furthermore, among currently emitted substances, N_2O is the one with the most damaging effect on stratospheric ozone (Ravishankara et al., 2009). Global N_2O emissions from soils (natural and managed) are about 60 % higher than at the beginning of the industrial era (1861), and most of this increase stems from cropland (Tian et al., 2019). The ambition to reverse this trend has led to extensive efforts to understand the mechanisms behind the emissions and to find mitigating management practices.

The main N_2O producing processes in soil are nitrification, including nitrifier denitrification, and denitrification (Kool et al., 2010; Braker and Conrad, 2011). The globally most important factors influencing emissions from agricultural soils are a) rate and type of nitrogen (N) application, b) crop type, c) soil organic matter content, d) soil pH and e) soil texture, where higher availability of N and organic matter, lower soil pH and finer texture all enhance N_2O emissions (Stehfest and Bouwman, 2006). In addition to these globally relevant predictors, in cool climates freeze-thaw cycles of the soil are related to high emissions (Goodroad and Keeney, 1984; Wagner-Riddle et al., 2017).

Nitrogen input rate has a strong influence on the N_2O emissions. In some studies, N_2O emissions have been found to increase linearly with N

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input (e.g., Lebender et al., 2014), while in others, a non-linear response has been observed, attributed to crop uptake of N (Zebarth et al., 2008; Kim et al., 2013). Crop uptake influences the mineral N content of the soil and the risks of N losses (Delin and Stenberg, 2014). In cases where emissions close to or after harvest dominate the annual N_2O budget, crop uptake is likely to influence the emission rates, with smaller effect on emissions from fertilizer rates at or below crop demand as a consequence.

The type of fertilizer used may also influence the emissions. In contrast to mineral fertilizers, organic amendments contain organic matter, which serves as substrate for denitrifiers, leading to higher denitrification and, potentially, higher N_2O emissions (Rochette et al., 2000; Senbayram et al., 2009; Senbayram et al., 2012). Organic fertilizers are a heterogeneous group, containing a wide range of amendments from green manures to livestock slurries, differing in e.g. dry matter content, pH and content and availability of carbon (C) and N. These properties influence the amendments' capacity to enhance soil N_2O emissions. In a review on the effects of anaerobic digestion of different feedstocks, soil emissions of N_2O were in general lower for treatments with digestates compared to undigested feedstocks, and this difference was particularly clear for pig slurries (Möller, 2015). During the anaerobic digestion process, easily degradable C is consumed (Möller, 2015), and therefore biogas digestate should theoretically stimulate soil heterotrophic microorganisms, and thus denitrification, less than untreated slurry.

Several meta-studies have been made in order to model N_2O emission factors (EFs) for different climates and/or fertilizer types. Several of these studies have concluded that EFs for organic N sources are in general lower than EFs for mineral sources (Aguilera et al., 2013; van der Weerden et al., 2016; Albanito et al., 2017; Charles et al., 2017). Based on these results, the Intergovernmental Panel on Climate Change (IPCC) in 2019 presented disaggregated EFs for mineral and organic fertilizers in wet climates, in their recommendations for the national greenhouse gas inventories *tier 1*) Hergoualc'h et al., 2019). The EF for mineral ("synthetic" in the IPCC guidelines) fertilizers, and for mixed inputs of both mineral and organic fertilizers, was there set to 0.016 kg N_2O-N kg^{-1} N input, while the EF for organic ("other", including crop residues as well as manures) amendments was set to 0.006 kg N_2O-N kg^{-1} N input.

There is a need for full-year N_2O emission studies in cold climates in order to identify the global contribution of freeze-thaw induced N_2O from arable land to the atmosphere (Wagner-Riddle et al., 2017). Furthermore, nitrous oxide emissions are highly variable in time and space (Henault et al., 2012), but there are still few studies with continuous or near-continuous measurements of emissions over time periods of one full year or more. More specifically, there are only few studies with high-frequency measuring between harvest in the fall and sowing in spring (Wagner-Riddle et al., 2007; Risk et al., 2014). There is one long-term study with high-frequency chamber measurements in a forest (Luo et al., 2012), but to the best of our knowledge, there is no full-year study with high-frequency chamber measurements reported for arable land. Since emissions during the part of the year when the soil is without a crop can contribute substantially to the annual budget (Wagner-Riddle et al., 2017), and the uncertainty of measurements increases significantly when measuring frequency decreases (Lammirato et al., 2021), this is an area where more knowledge is desirable. As described, different studies point in different directions regarding N_2O emissions from organic compared to mineral N sources. With this background, we see the need for a comparison made in a cool, temperate region with ample precipitation, involving crops and mineral and organic N sources typical for the region. The aim of this study was to compare the effect of different N rates and N sources on direct N_2O emissions from a clay soil during one full year. In doing this, our objectives were to (1) provide an overview of annual flux dynamics, based on high temporal resolution data, and (2) determine whether there were differences in emissions among the alternative fertilizing practices, and if there were, describe how differences were distributed over the year. In order to improve the environmental standard of agriculture, minimizing emissions is not enough; while reducing emissions, it is also necessary to maintain (or increase) the yields. It is therefore reasonable to consider not only N_2O emissions per unit area, but also emissions in relation to the production (van Groenigen et al., 2010). In this paper, we present one-year emission results for the contrasting treatments using four scales: area, yield, N yield, and fertilizer N input.These were our hypotheses: .

1. Emissions of N_2O increase with higher N surplus, defined as N input with fertilizers minus N in yield.
2. Organic amendments cause higher N_2O emissions per unit area compared to mineral fertilizers with the same amount of plant available N added.
3. The effect of fertilizers on cumulative seasonal N_2O emissions is more pronounced during the crop season than when the soil is without a crop.

2. Materials and methods

Nitrous oxide measurements were performed from 19 May 2015–30 June 2016 at Lanna research station, in a plain area in the south-west of Sweden (58°20 N, 13°7 E). The soil is a silty clay (40–46 % clay) with 2.3–3.5 % organic matter and pH 6.6–7.2 in the top soil (0–20 cm). The soil type is classified as a Cambisol (Greve et al., 2000). Particle density is approximately 2.6 g cm^{-3} and dry bulk density in the top soil 1.25 g cm^{-3}. Annual precipitation (19 May 2015–18 May 2016) was 532 mm, close to the average for the years 1991–2020 of 583 mm. The distribution of precipitation over the year was also normal. Mean temperature (1 May 2015–30 April 2016) was 7.2 °C, close to the long-term (1991–2020) average of 6.9 °C. In winter 2015–2016, there were 49 days with mean temperature ≤ 0 °C; the mean corresponding figure for 1996–2020 was 52 days. Complete data with daily resolution were not available from Lanna, so the data on days with freezing mean temperatures were taken from Hällum, 5 km south-west of Lanna, in the same plain area (Meteorological and Hydrological Institute, 2021). Hällum air temperature data were available only for 1996–2020, hence the reference period in this case was reduced to 25 years. A comparison of monthly mean temperatures in Lanna and Hällum in 2015 showed that Hällum was mostly slightly colder (variation from 0.1 °C warmer to 0.4 °C colder, with a mean of 0.2 °C colder in Hällum).

The crop grown in the field was winter wheat in 2014, spring barley in 2015 and spring oats in 2016. Field operations during the experiment were typical for conventional farming in the area, with plowing in autumn and harrowing shortly before sowing in spring (in autumn if a winter crop was to be sown). A first dose of mineral fertilizers was applied in April, for spring crops at the time of sowing. All crop residues were returned to the field at harvest.

2.1. Fertilizer treatments

The N_2O measurements were part of a bigger project conducted on 20 plots with five fertilizer treatments in a randomized block design. Due to logistic reasons, emissions of N_2O could be measured only in ten plots, i.e. in two replicates per treatment. The plots belonged to a facility for the measuring of N leaching, described in detail by Wallman and Delin (2022). From 2014–2016, i.e., starting the year before the N_2O measurements, there were four fertilized treatments plus control: .

1. Control, no N fertilizer added (CL).
2. Normal mineral N rate (NM).
3. High mineral N rate (HM).
4. Biogas digestate (BD).
5. Pig slurry (PS).

The NM treatment represents fertilizer application with mineral N according to recommendations by the Swedish Board of Agriculture, which reflects what was the expected economic optimum for winter wheat production in 2014, spring barley in 2015 and spring oats in 2016 in this production zone (Table 1). The calculated break-even point in 2015 based on expected prices of N fertilizer and barley was at 10.5 kg extra yield per additional kg N (Albertsson, 2013; Albertsson et al., 2014, 2015). The HM treatment represents 50 % higher N application rates compared to expected economic optimum. For NM and HM, the mineral fertilizer dose was split in two, applied in April and May. The form of mineral fertilizer used in the April application in 2014 (in the winter wheat crop) was ammonium nitrate (NH_4NO_3), with 50 % of N as ammonium (NH_4^+) and 50 % as nitrate (NO_3^-), and the complementary N dose in May was given as calcium nitrate, including 7 % of N as NH_4^+ and 93 % as NO_3^-. In April 2015 and 2016 (at sowing), NPK-22–6–6 was applied, with 61 % of the N content as NH_4^+ and 39 % as NO_3^-. In May 2015 and 2016 additional N was given as NH_4NO_3. Surface broadcasting was used for the application of mineral fertilizers. For BD and PS, the same mineral fertilizer as in NM and HM, with the same N rates as for NM, was given in April all three years, and the organic amendments were applied in May, with trailing hose in growing crop (no incorporation). The control received no fertilizer N, but P and K. The N deposition in the area was approximately 6 kg N ha^{-1} yr^{-1} (Pihl Karlsson et al., 2010).

Plant available N in organic amendments was defined as the NH_4-N content at the time of application (Table 1). In both the digestate and the pig slurry, the plant available N content was 3.5–4.3 g NH_4-N kg^{-1} 2014–2016. pH in the digestate was about 8.0–8.1 and in the pig slurry 7.2–7.4 (measured only in 2014 and 2015). The pig slurry used originated from fattening pigs. The raw material for the biogas digestate was also mainly slurry from fattening pigs, but from another farm than in the PS treatment. When the digestate and pig slurry were applied to the field, samples were taken and sent to a laboratory for analysis (Agrilab AB in 2014 and Eurofins Agro Testing Sweden in 2015 and 2016). Before that, preliminary sampling had been made to guide the application rates. The preliminary sampling turned out to be misleading in some cases, which in 2014 resulted in N application rates 50 % higher than intended in the PS treatment. This means that the limit for N input of 170 kg ha^{-1} yr^{-1} from organic sources, set by the EU nitrate directive, was exceeded (Table 1) (European Council, 1991). In addition, the pig slurry used in 2014 had a high content of C. With the manure applied, 1.4 Mg C ha^{-1} was included, which was nearly 4 times the C content added with the biogas digestate in 2014.

Table 1
Fertilizers applied 2014–2016 and properties of organic amendments used. Split application was practiced in all fertilized treatments. Abbreviations: CL = control, NM = normal mineral N, HM = high mineral N, BD = biogas digestate, PS = pig slurry, org. fert. = organic fertilizers, pl. avail. N = plant available N (NH_4-N and NO_3-N), tot. N = total N (the sum of organic and plant available N), DM = dry matter, C/N = ratio of carbon to total nitrogen in organic amendments, n.a. = not available.

Year	Treatment	Mineral N kg N ha^{-1}	N in org.fert. kg NH_4-N ha^{-1} (kg tot-N ha^{-1})	Total kg pl.avail. N ha^{-1} (kg tot-N ha^{-1})	DM %	C/N
2014	CL	0	0	0		
	NM	160	0	160		
	HM	240	0	240		
	BD	80	91 (122)	171 (202)	4.0	3.0
	PS	80	160 (238)	240 (318)	8.6	6.1
2015	CL	0	0	0		
	NM	120	0	120		
	HM	180	0	180		
	BD	55	108 (127)	163 (182)	3.0	1.7
	PS	55	70 (86)	125 (141)	3.0	2.0
2016	CL	0	0	0		
	NM	120	0	120		
	HM	180	0	180		
	BD	55	70 (92)	125 (147)	2.9	n.a.
	PS	55	86 (112)	141 (167)	3.1	n.a.

2.2. Nitrous oxide flux measurements

The chambers for automated measurements were installed in the field in the beginning of May 2015, and the measurements started in the afternoon 19 May. Measurements went on until midnight 30 June 2016, i.e., in total for more than 13 months. In this paper, the first year of these measurements, i.e., 19 May 2015–19 May 2016, was used as a basis for the comparisons of emissions at different scales (in relation to area, yields, N offtake and N input), while the whole measurement series was included in the analysis of mechanisms of importance for the N_2O emissions.

Chambers, including the lids, were made of transparent plastic, without insulation. The bottom area of the chambers was 0.50 by 0.50 m and the height 1.05 m. Each chamber rested on a collar with the bottom area 0.44 by 0.44 m and height 0.03 m. This makes the total volume 0.27 m^3. Chambers were permanently placed in the field, but for each chamber, there were three collars in the same plot. The chambers were moved between these collars twice a week, weekly or once in two weeks, depending on season, with the highest frequency during summer. This was done to ensure that conditions for crop and soil in the chamber were similar to conditions in the ambient environment, e.g. regarding precipitation, temperature and wind. Each chamber had two fans: one for mixing the air within the closed chamber and one for ventilating the chamber while open. Chambers were removed from the plots before field operations and returned afterwards. Measurements from periods when a chamber was out of position were not included in the data set.

The chambers had their lids closed for 20 min every hour, 24 h per day. During closure, 8–9 gas samples were taken. Concentrations of N_2O (as ppmV) of the samples were measured with a trace gas analyser (TGA 100A, Campbell Scientific). The data logger used was CR3000 (Campbell Scientific). From the N_2O concentrations, fluxes were calculated via linear regression. This means that, under normal operation, the measuring system produced one flux estimate every hour for each of the 10 chambers.

2.3. Measurements of crop yield and N offtake

Crop yields were measured in four plots per treatment, i.e., all treatment plots in the experimental field were included, not only those with chambers for N_2O measurements. During harvest, three areas of 20 m^2 in each plot were harvested separately, and a grain sample from each such sub-plot was sent to analysis (The Rural Economy and Agricultural Society). Samples were weighed before and after removal of litter. The cleared sample was analysed for contents of water and N, using near infrared transmission (NIT) spectroscopy. Average values of yield, litter, water and N content from all three subplots were used for the calculation of mean values per plot, which were then used in the statistical analysis of this paper.

2.4. Weather and soil monitoring

Manual daily measurements of precipitation were made 500 m from the chamber plots. Other meteorological measurements were conducted 1.3 km away from the field at the ICOS station of Lanna, where also soil temperature and soil water content were measured (icos-sweden.se). The landscape structure (flat, arable land), soil type, crop and management were the same as in the field experiment, and the fertilizer input was identical to that in the NM treatment. Air temperature and air pressure for flux calculations were measured automatically every 10 min. Soil temperature and soil water content were measured automatically every 30 min with four replicates at five depths. Soil volumetric water content (VWC) was measured automatically with time domain reflectometry (TDR) devices.

2.5. Gap filling

In the data set of N_2O fluxes, there were gaps of different lengths, from a few hours up to several weeks, mostly for all chambers at a time. The gaps originated from technical problems or field operations. In order to estimate cumulative emissions for one year and the distribution per season, gap filling was applied.

The method used for the filling of gaps was a modified version of the lookup table approach described by Mishurov and Kiely (2011), i.e., a simple imputation with predictive mean matching. From measured data, two lookup tables were created for each chamber. One lookup table was created from data measured when soil temperature was above 0 ˚C. This lookup table consists of the means of fluxes grouped by soil temperature decile, soil water content decile and time elapsed since the last N input (6 classes). Another lookup table was created from data measured when soil temperature was below 0 ˚C. This lookup table consists of the means of fluxes grouped by soil temperature decile and rate of soil temperature change (deciles) in the hour prior to chamber closure. Mean soil temperature and mean soil water content at 5 cm depth were used. Gaps were filled with entries from one of the two tables, depending on the soil temperature during the gap. In the gap-filled data set for the full measuring period 19 May 2015–30 June 2016, measurements represented 83 % of all data points. For further information, see Supplementary material.

During the time period 2016-04-20 07:00–2016–05–14 07:30, measurements of volumetric water content appear not to have been working satisfactorily; all four replicates were stuck around specific values for long periods of time, in spite of sparse precipitation in combination with rising soil temperatures. However, since these soil moisture data were in no sense extreme, and we were lacking better options, we decided to keep these low-quality data in the data set used for the gap filling.

2.6. Seasons

Time was divided into two seasons based on the crop year. In order to have full months, the crop season was set to 1 April-31 August, and the no-crop season to 1 September-31 March. This division means that the no-crop season was a continuous period covering autumn and winter 2015–2016, while the crop season of the first year was the sum of two separate periods: 19 May-31 August 2015 and 1 April-19 May 2016. As a result, the first fertilizer input, with mineral N applied to all fertilized treatments was covered in 2016, while effects of the second fertilizer application, when the organic amendments were added to the BD and the PS treatments, were covered in 2015 (Table 1).

2.7. Statistical analysis

A statistical analysis of seasonal emissions per unit area was made using the gap-filled data. The statistical analysis aimed at investigating the differences between treatments, per season. The unit used in the analysis was the chamber (plot). Documentation on chamber movements was not detailed enough to allow for the use of chamber stations (collars) as units (Section 2.2). This means that three geographical spots were underlying each chamber unit.

As a way to handle repeated measures (two seasons), a linear mixed-effects model was used to describe the mean daily N_2O emissions with treatment and season, and the interaction between the two as fixed effects, and chamber as a random effect. The natural logarithm of the mean N_2O emissions was used in the model, in order to correct for skew and for differences in variability between treatments. Input data were cumulative measurements per chamber and season divided by the number of days per season, giving the seasonal mean of daily fluxes per chamber, i.e., one record per chamber and season. Treatments were compared pairwise per season and p-values for these comparisons were calculated. Due to the small number of replicates ($n = 2$), no adjustment for multiple comparisons was made. All statistical tests were made at a 5 % significance level.

In order to assess whether the gap filling altered the relations between treatments, the analysis described above was repeated for the measurement data set without gap filling. The result of the comparison of the data sets with and without gap filling is available in the Supplementary material.

Nitrogen offtake (N in yield) was determined from the N share of the yield and the yield per hectare (Section 2.3). The surplus of N was calculated as fertilizer total N input minus N offtake. Statistical analysis of differences between treatments regarding yields, N offtake and N surplus were made with a linear mixed-effects model with treatment, year and the interaction between treatment and year as fixed effects and plot as a random effect. Results were not adjusted for multiple comparisons. The number of replicates in the statistical analysis of yield, N offtake and N surplus was four.

2.8. Other calculations

The statistical analysis of annual cumulative fluxes was supplemented with a pairwise assessment of treatments with regard to the frequency with which both replicates of one treatment were higher or lower, respectively, relative to another treatment (i.e., no overlap). For this purpose, the non-gap-filled data set of fluxes was sub-sampled to extract only data for hours in which measurements from all chambers were available. In total, this data set comprised 7777 h with one measurement from each of the ten chambers each hour, covering the whole measuring period, 19 May 2015–30 June 2016.

Cumulative emissions per treatment were calculated from the gap-filled data set for the crop season, the no-crop season, the first year, the start of the second year (1.5 months) and for the whole measuring period. For each replicate, all hourly fluxes were summed up for the time period in question. Means per treatment were calculated as simple arithmetic means of the two replicates.

When emitted N_2O-N per mass of yield and per N offtake was calculated, mean cumulative, gap-filled N_2O-N emissions per treatment during one year were divided by mean yield and mean N offtake per treatment, respectively. No statistical analysis was carried out on these measures, and also not on emissions per unit N input, which were calculated for each of the fertilized treatments as the mean gap-filled N_2O emissions for one year divided by total N input.

2.9. Software used

For the cleaning and handling of data, calculation of fluxes, gap filling, statistical analysis and plotting, the statistical software R (Core Team, 2019) was used, with the addition of the packages tidyverse (Wickham et al., 2019), emmeans (Lenth, 2019), egg (Auguie, 2019) and lubridate (Grolemund and Wickham, 2011).

3. Results

3.1. Yields and N surplus

Barley yields in 2015 were 7–8 Mg ha^{-1} in all fertilized treatments, which was about 3 times higher than the CL yield (Table 2). For yield and N offtake, the ranking of treatments was in most cases as follows: HM had the highest levels, NM intermediate and CL very low, while BD and PS were between NM and HM, although in 2014 yields and N offtake in BD and PS were lower than in NM (significant only for BD). The HM treatment stood out in the N offtake comparison because of its combination of high yield and high N content of the yield. In both 2015 and

Table 2
Yield of winter wheat (2014), barley (2015) and oats (2016) (all with 14 % water content), N offtake and N surplus per hectare. Surplus of N was calculated as total fertilizer N input minus N offtake. Presence of the same letter (a, b, c, d, e) indicate no significant differences between treatments ($\alpha = 5\%$). Comparisons were made only within years. Abbreviations: CL = control, NM = normal mineral N, HM = high mineral N, BD = biogas digestate, PS = pig slurry.

Year	Treatment	Yield Mg ha^{-1}	N offtake kg N ha^{-1}	N surplus kg N ha^{-1}
2014	CL	3.5^{a}	43^{a}	-43^{a}
	NM	7.7^{bc}	136^{b}	24^{b}
	HM	8.0^{c}	168^{c}	72^{c}
	BD	7.0^{d}	110^{d}	92^{d}
	PS	7.4^{bd}	128^{b}	190^{e}
2015	CL	2.5^{a}	28^{a}	-28^{a}
	NM	7.0^{b}	88^{b}	32^{b}
	HM	7.9^{c}	120^{c}	60^{c}
	BD	7.3^{bc}	96^{b}	86^{d}
	PS	7.5^{bc}	95^{b}	46^{e}
2016	CL	2.5^{a}	28^{a}	-28^{a}
	NM	6.1^{b}	76^{b}	44^{b}
	HM	6.8^{c}	102^{c}	78^{c}
	BD	6.4^{bc}	83^{b}	64^{d}
	PS	6.7^{c}	83^{b}	84^{c}

2016, N offtake had an approximately linear increase in relation to N input from CL to HM. Among the fertilized treatments, NM had the lowest N surplus, followed by HM and then the organic amendment treatments.

3.2. *Cumulative emissions per hectare*

Full-year, cumulative, gap-filled emission means per treatment ranged from 0.48 kg N_2O-N ha^{-1} for CL to 1.6 kg N_2O-N ha^{-1} for PS (Fig. 1). The treatments form two groups, where the one-year emissions from CL and NM were half or less of the emissions per unit area compared to HM, BD and PS. These differences were significant ($p < 0.05$) in all cases during both seasons for CL, while for NM, quantitatively relevant, but not always statistically significant, differences were observed (p 0.04–0.11). The pattern of CL and NM being lower than HM, BD and PS was even clearer and more pronounced when considering not only the first year, but the whole measuring period.

In 31–42 % of all measurement hours with data from all ten chambers, the HM, BD and PS treatments had higher fluxes observed in both their respective chambers than in both of the CL chambers, while the CL treatment had higher fluxes in both chambers only in 5–6 % of the time (Table 3). The HM, BD ans PS treatments had higher fluxes in both chambers than observed in any of the NM chambers in 22–30 % of the measurement hours, while both NM chambers had higher fluxes in 6–8 % of the hours. The outcome of this analysis is largely consistent with that of the statistical analysis of cumulative fluxes, i.e., HM, BD and PS tended to cause higher emissions relative to CL and NM. However, the analysis also suggests a difference between the CL and the NM treatment, with fluxes in both NM chambers being higher than fluxes in both CL chambers more than twice as often than the other way round.

Table 3
The frequency when both chambers of a given treatment have higher fluxes than both chambers of another treatment in the same hour of measurements. Data from the whole measuring period (19 May 2015–30 June 2016) were included, but only hours with measurements from all ten chambers. Abbreviations: CL = control, NM = normal mineral N, HM = high mineral N, BD = biogas digestate, PS = pig slurry, repl. = replicate, comp. =compared.

Comparison	A > B	B < A
Both CL repl. (A) comp. with both NM repl. (B)	8 %	19 %
Both CL repl. (A) comp. with both HM repl. (B)	5 %	35 %
Both CL repl. (A) comp. with both BD repl. (B)	5 %	31 %
Both CL repl. (A) comp. with both PS repl. (B)	6 %	42 %
Both NM repl. (A) comp. with both HM repl. (B)	6 %	25 %
Both NM repl. (A) comp. with both BD repl. (B)	8 %	22 %
Both NM repl. (A) comp. with both PS repl. (B)	7 %	23 %
Both HM repl. (A) comp. with both BD repl. (B)	15 %	14 %
Both HM repl. (A) comp. with both PS repl. (B)	15 %	13 %
Both BD repl. (A) comp. with both PS repl. (B)	14 %	17 %

During the first year studied, no-crop season emissions represented about 75 % of the annual emissions in all treatments, including CL (Fig. 1). The no-crop season was 7 months, while the crop season was 5 months, but even in a comparison of seasonal mean daily fluxes, the emissions during the no-crop season were about twice as high as during the crop season. Most of the no-crop season emissions (65–80 %) appeared in periods of freeze-thaw cycles (Fig. 3).

3.3. *Comparison of treatments using different scales*

Per unit area, CL and NM had similar cumulative N_2O emissions in the first year of measurements, which were low compared to the other treatments (Fig. 2). The NM treatment remained low also in the comparisons of emissions in relation to yield, N offtake and N input. In contrast, PS had among the highest emissions at all scales, at least twice as high as NM. The CL treatment had the lowest emissions per unit area, but high N_2O emissions per unit yield and per unit N offtake because of its low yield and low N content. For the HM, BD and PS treatments it was their high emissions per unit area that were reflected also in the yield-related emission measures.

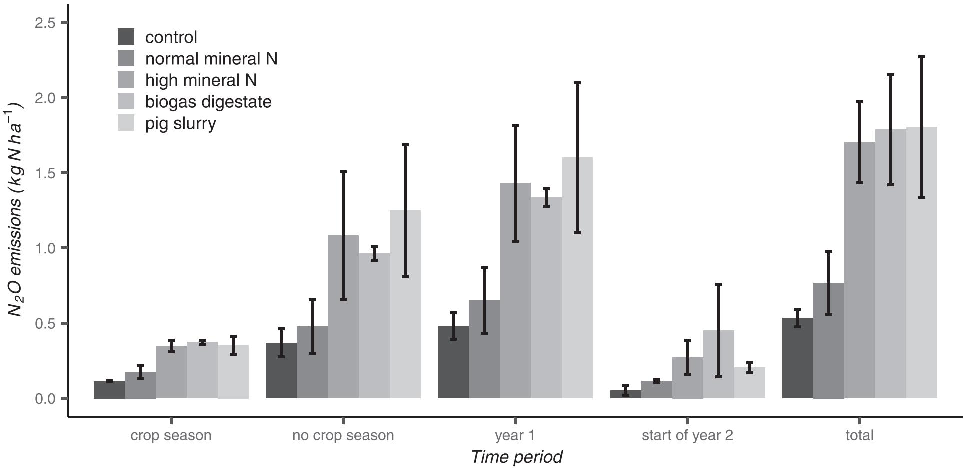

Fig. 1. Cumulative gap-filled N_2O emissions per fertilizer treatment, per time period, average per treatment. Time periods include: Crop season = 19 May - 31 August 2015 plus 1 April - 19 May 2016, no-crop season = 1 September 2015–31 March 2016, first year = 19 May 2015–19 May 2016, start of year 2 = 19 May - 30 June 2016, whole measuring period (total) = 19 May 2015–30 June 2016. Error bars represent standard error, which in this case, with two replicates, is identical to the range.

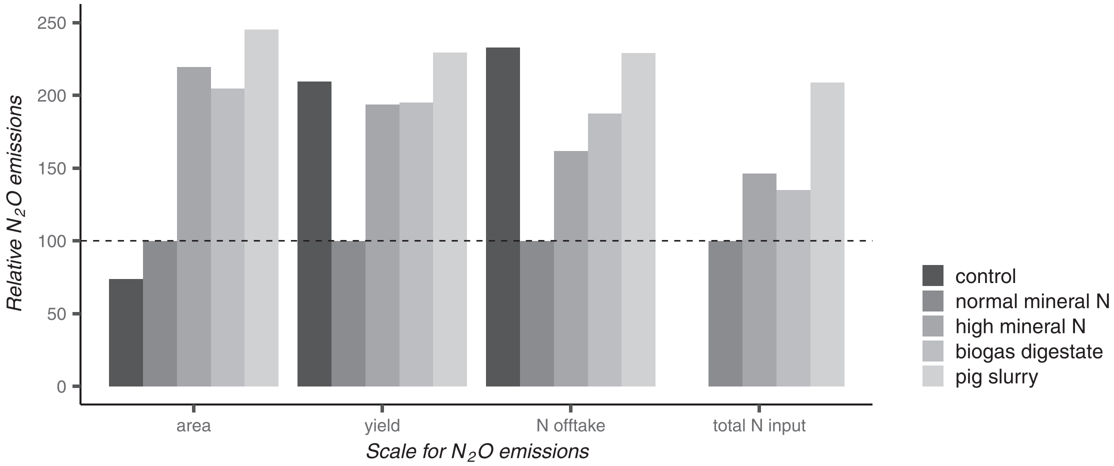

Fig. 2. One-year N_2O emissions per treatment in relation to area, yield, N offtake and fertilizer total N input. All emissions are set relative to the normal mineral N treatment (100). Barley yields in 2015, fertilizer total N input in 2015 and cumulative N_2O emissions 19 May 2015–19 May 2016 were used for the calculations. The control did not have any fertilizer N input, and was therefore not included in the comparison per total N input.

3.4. Nitrous oxide fluxes related to soil conditions

The largest peaks in daily N_2O fluxes occurred in connection to either freeze-thaw cycles or fertilizer application in combination with high soil water content (Fig. 3). Beside these peaks, elevated fluxes appeared at high soil water content levels, mostly in the no-crop season. Periods with soil temperatures below 0 ˚C were short, and snow cover, if any, was thin.

The highest fluxes for time periods more than 15 days after fertilizer application were related to freeze-thaw cycles and appeared at soil temperatures around 0 ˚C in all treatments (Fig. 4). At soil temperatures below − 5 ˚C, N_2O fluxes were mostly close to 0 or negative. Between + 4 and + 8 ˚C, fluxes were low (0–5 g N_2O-N ha^{-1} d^{-1}), even though some elevated fluxes could be observed within this temperature interval at soil VWC above 30 % (58–67 % water filled pore space, WFPS). At soil temperatures above + 8 ˚C, fluxes tended to be higher, particularly when VWC exceeded 22 %.

3.5. Fluxes following fertilizer application

Fertilizer application was followed by peaks in N_2O fluxes, particularly in the first five days after the second fertilizer input in 2016. Looking at that period of time, N_2O fluxes were highest in the first day, except in the PS treatment, where maximum flux appeared after two days (Fig. 5). In 2015, flux measurements started about 24–30 h after fertilizer application, which means that the time closest to the fertilizer application event was covered only in 2016, when measurements started 10 min to 6 h after N application. Measured fluxes of N_2O after application of fertilizer were both higher and sustained at a high level for a longer time in 2016 compared to 2015, in spite of soil moisture being lower (from day two) in 2016 (Fig. 5). Mean soil temperature in the first five days after the second fertilizer application was lower in 2015 (11.5 ˚C) than in 2016 (14.0 ˚C).

The time after the first fertilizer application in 2016 (when mineral N was applied to NM, HM, BD and PS treatments) could not be covered by this analysis because soil water content data for that time period were of low quality (Section 2.5). However, the flux measurements that are available from this time period tell us that emissions were low, probably because of the cold and dry weather.

4. Discussion

Peaks of N_2O fluxes mostly appeared at times of high soil moisture (> 30 % VWC), but elevated fluxes were not restricted to those moments (Fig. 3 and 4). We therefore reason that denitrification (including nitrifier denitrification) made the largest contribution to N_2O emissions from our soil during the year studied, but that nitrification was also important (Webster and Hopkins, 1996; Bateman and Baggs, 2005; Kool et al., 2011).

4.1. It is possible to combine good yields with very low nitrous oxide emissions

Our observation that N_2O emissions per unit area were higher or tended to be higher for treatments with higher N surplus (HM, BD and PS) than for treatments with lower N surplus (CL and NM) supports our first hypothesis, however the large difference in N surplus (Table 2) between CL and NM did not lead to any clear difference in N_2O emissions. The higher emissions in the HM treatment may be explained by increased denitrification at higher NO_3^- availability and by higher denitrification product ratio (N_2O-N:(N_2O-N + N_2-N)) following higher soil NO_3^- concentrations (Weier et al., 1993; Gillam et al., 2008). While Gillam et al. (2008) saw no increase in total denitrification rates at higher NO_3^- concentrations, Chantigny et al. (1998) had generally both higher denitrification product ratios and higher denitrification rates in soil cores receiving more N. The difference in N_2O emissions (a factor of 2) between NM and HM, was greater than the difference in N input (a factor of 1.5) between those two treatments, suggesting that N surplus (HM had double the surplus of NM) was more important than N application rates for N_2O emissions; Chantigny et al. (1998) drew a similar conclusion.

The lower emissions in relation to N offtake in the NM treatment compared with the HM treatment supports the findings of the meta-study by van Groenigen et al. (2010), which concluded that the lowest N_2O emissions in relation to above-ground N uptake are found at intermediate N application rates. Similar findings have been made widely (Zebarth et al., 2008; Lebender et al., 2014; Russenes et al., 2019).

The average increase of 15 kg yield per additional kg N between NM and HM was higher than the price ratio at the economic break-even point. This means that the actual economic optimum for barley in 2015 was higher than the expected 120 kg N ha^{-1}, probably between the NM and HM N input rates. The small, non-significant difference between CL and NM in emissions per unit area indicates a limited potential for reduction of N_2O emissions by cutting N input below recommended rates. Furthermore, reduced N input would probably also mean lower yields and thus increased yield-scaled emissions.

Annual N_2O emissions from all fertilized treatments reported here are in the lower range of other studies (Flessa et al., 1995; Kaiser and

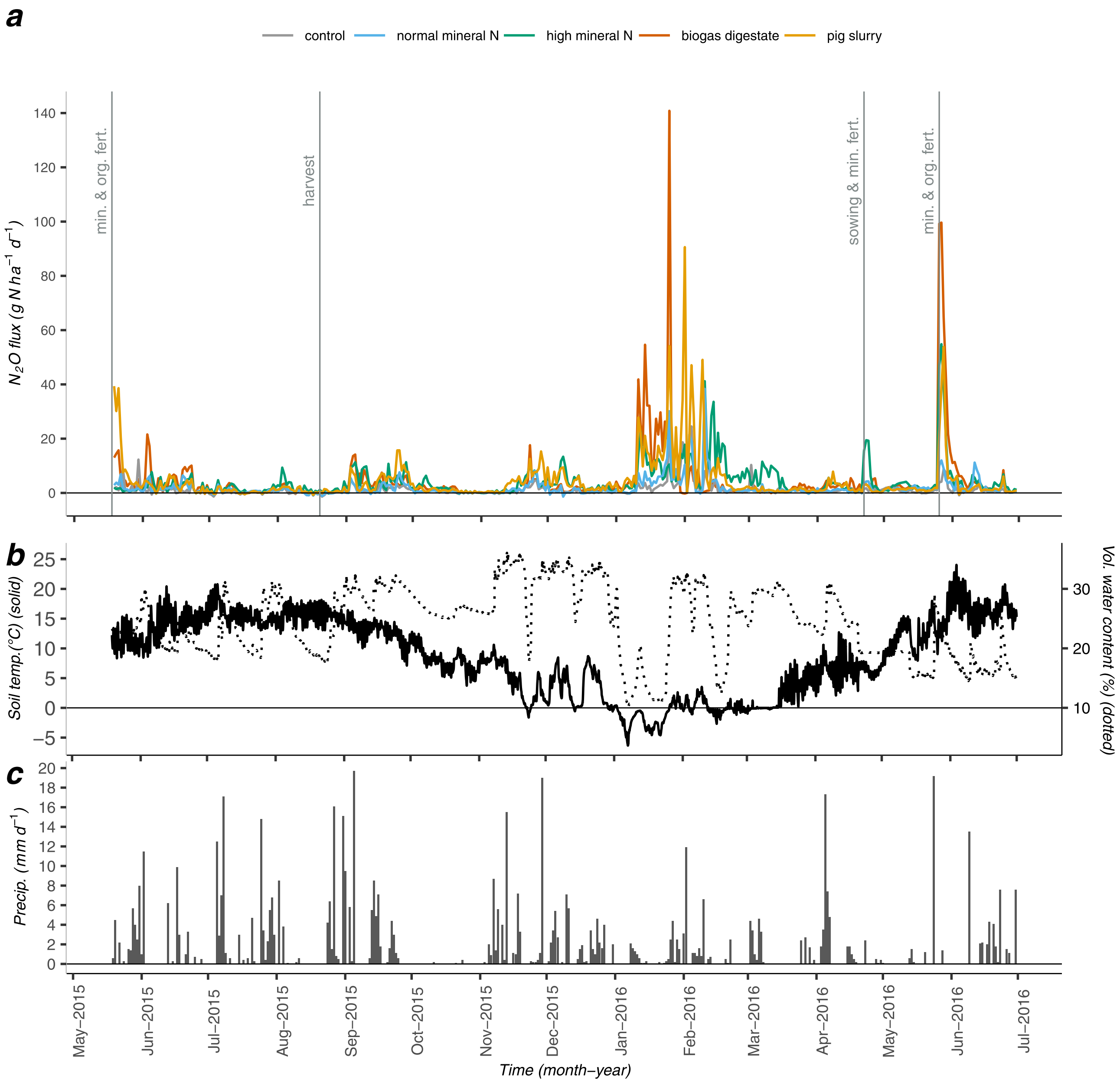

Fig. 3. Nitrous oxide fluxes and environmental factors 19 May 2015–30 June 2016. (a) Gap-filled daily mean N_2O flux per treatment (in color in the online version of the article), with timing of main field operations indicated. (b) Soil temperature and volumetric water content, both at 5 cm depth and with 30 min resolution. (c) Daily precipitation. Abbreviations: min = mineral, org. = organic, fert. = fertilizer application, vol. = volumetric, precip. = precipitation.

Ruser, 2000; Wagner-Riddle et al., 2007; Pelster et al., 2012; Lebender et al., 2014; Cardenas et al., 2019), although emission rates for the control appear to be more in line with what was previously reported (Chang et al., 1998; Pelster et al., 2012; Lebender et al., 2014; Cardenas et al., 2019). Initially, we intended to use micrometeorological methods for the measuring of N_2O emissions (in separate fields, one per treatment, at Lanna research station), but emissions turned out to be below detection limit most of the time. The generally low fluxes were also confirmed by eddy-covariance measurements at the ICOS station (NM treatment), where only emission peaks could be detected (unpublished). So, even on a fine-textured soil, with an annual crop, in a climate where precipitation is larger than evapotranspiration, and where freeze-thaw cycles contribute to temporarily very high N_2O fluxes, N_2O emissions per unit area were generally low, and yield-scaled emissions from the treatment most similar to common practice (NM) were particularly low.

4.2. High nitrous oxide emissions from organic fertilizer treatments were not associated with high nitrate concentrations in drainage water

Attribution of N_2O emissions to a single year's N input was not as straight-forward in the organic fertilizer treatments as it was for the mineral treatments. We therefore here take a wider perspective, including measurements of N yields and N losses other than N_2O in the same field to sketch a probable picture of the N dynamics.

In 2014, there were large N surpluses in the organic fertilizer treatments, particularly in PS (Table 2), but this did not result in high N leaching (Wallman and Delin, 2022). Losses of N as NH_3 after application of digestate and slurry were considerable (9 and 26 kg N ha^{-1}, respectively), but still not high enough to explain the low N offtake and leaching. The clay at Lanna Research Station is dominated by illite, which is known to have high NH_4^+ fixing capacity. In addition to being fixed between mineral layers, NH_4^+ can also bind to to mineral surfaces,

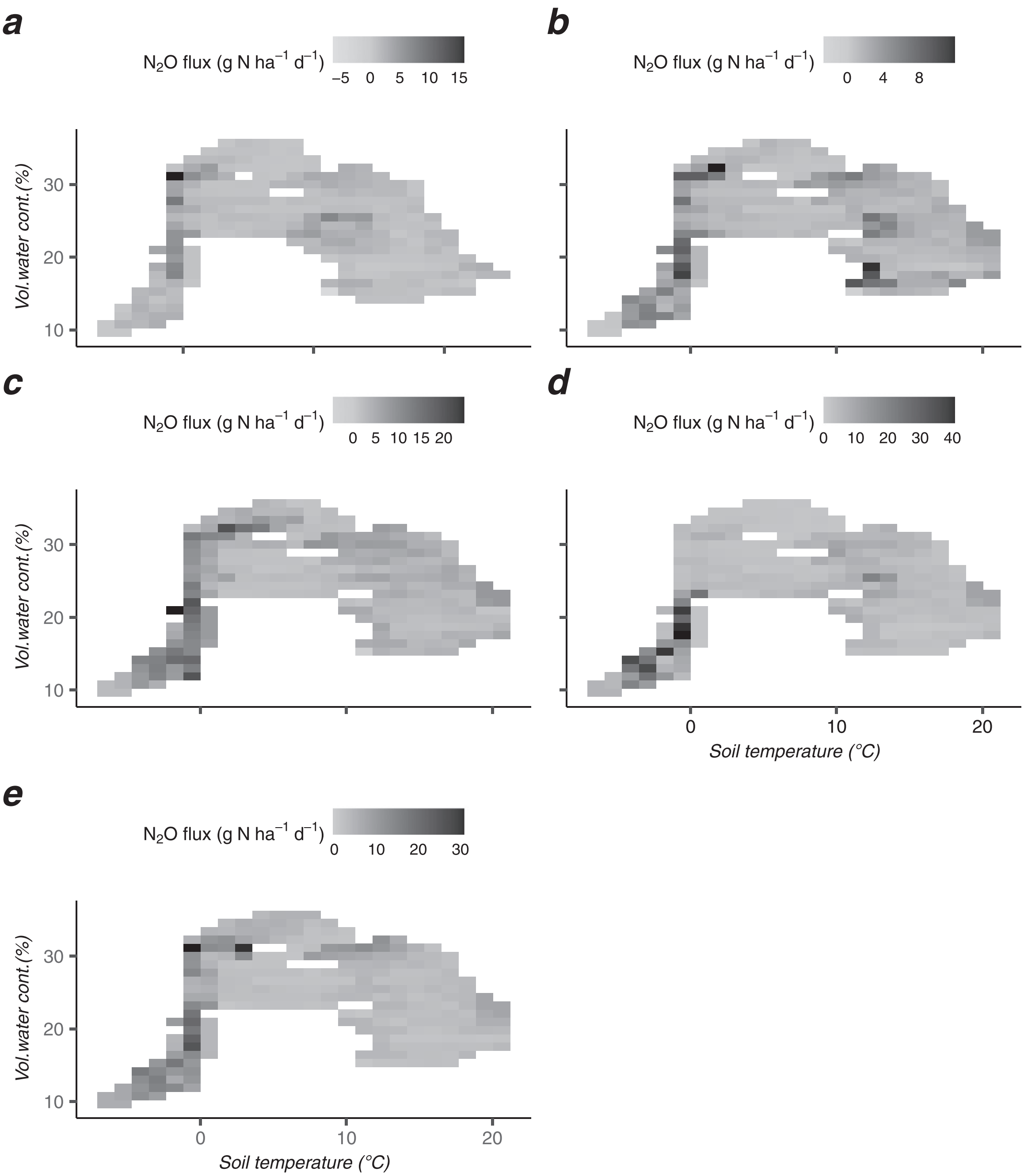

Fig. 4. Co-variation of N_2O fluxes with soil temperature and soil moisture. (a) Control. (b) Normal mineral N treatment. (c) High mineral N treatment. (d) Biogas digestate treatment. (e) Pig slurry treatment. Measured N_2O fluxes, represented in gray-scale, at different combinations of volumetric soil water content and soil temperature at 5 cm depth, per treatment (treatment means). Only fluxes measured more than 15 days after fertilizer application are included. Note the different flux scales.

i.e. exchange sites, where it is available to plants, but protected from leaching (Dittert et al., 1998; Scherer and Weimar, 1994). We reason that considerable shares of the NH_4^+ added in the BD and PS treatments in 2014 were fixed or bound to exchangeable sites, which means that it could later be nitrified or taken up by plants. In addition, some of the organic N added in the PS and BD treatments in 2014 and 2015 may also have become mineralized during the period of N_2O measurements, adding to the amount of N available to crops and microbes (Gutser et al., 2005; Bhogal et al., 2016).

In studies on denitrification in different soil types, fine-textured soils have been found to contain enough organic matter for denitrification to accelerate after addition of mineral N, while in more coarse-grained soils, denitrification was limited by availability of organic C (Gillam et al., 2008; Rochette et al., 2008; Pelster et al., 2012). In our study, soil was fine textured (silty clay), but rather low in organic matter; the content of organic C was in the same range as the lighter soils in the studies by Rochette et al. (2008) and Pelster et al. (2012). We therefore could expect addition of organic C to have an effect on denitrification rates. Mean N concentration (mainly NO_3^-) in the drainage water 2014–2017 was significantly higher in the HM treatment than in the other treatments (Wallman and Delin, 2022). This indicates that denitrification in the HM treatment was not limited by NO_3^-, but rather by C availability. In contrast, BD and PS, with N_2O emissions in the same range as HM, had significantly lower drainage water N concentrations than HM, between those of CL and NM. This probably means that much of NO_3^- produced through nitrification was immediately denitrified, a

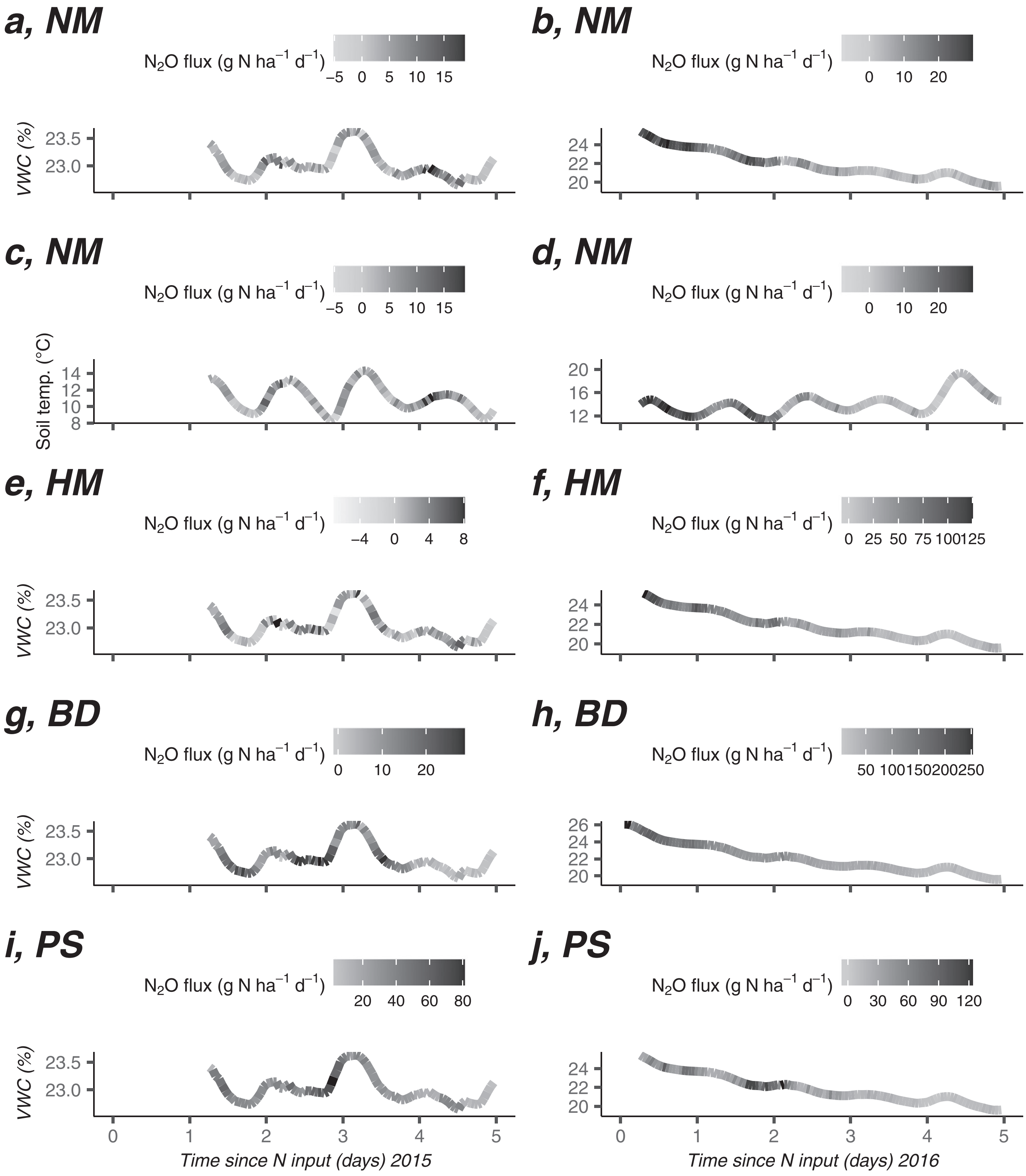

Fig. 5. Co-variation of N_2O fluxes with time since N input and soil temperature or soil moisture. (a and c) Normal mineral N treatment 2015. (b and d) Normal mineral N treatment 2016. (e) High mineral N treatment 2015. (f) High mineral N treatment 2016. g) Biogas digestate treatment 2015. (h) Biogas digestate treatment 2016. (i) Pig slurry treatment 2015. (j) Pig slurry treatment 2016. Measured N_2O fluxes, represented in gray-scale, at different combinations of soil water content or soil temperature and time since fertilizer N application, per treatment (treatment means). Included are fluxes measured up to 5 days after N application in May both years. Soil temperature is displayed only for the NM treatment; the temperatures were not measured individually per treatment and fluxes are already displayed in sub-plots (e-j). Note the different flux scales. Abbreviations: VWC = volumetric water content, temp. = temperature.

process fueled by organic C. Organic matter both provide denitrifiers with energy and carbon, and contribute to lower O_2 supply in soil during its decomposition (Robertson and Groffmann, 2007). Considering the lower NO_3^- input with fertilizers in BD and PS compared with the mineral treatments, combined with the high N_2O emissions in BD and PS, nitrification rates appear to have been higher in the organic fertilizer treatments than in NM to make possible the higher emissions via nitrification and/or denitrification. This could be an effect of the higher NH_4^+ input in the organic fertilizer treatments. Higher nitrification rates from organic compared to mineral N sources have been observed in a study where mineral and organic fertilizer treatments received the same input of NH_4^+-N, likely an effect of nitrification being stimulated by the rise of pH caused by the application of organic amendments (Müller et al., 2003).

The BD treatment had a plant available N input close to HM in 2015, and considering the very high N surplus of the PS treatment from 2014, of which a large amount was probably left in soil in spring 2015 thanks to NH_4^+ fixation, it is reasonable to compare also PS with HM rather than with NM regarding effects of N inputs. With this perspective, we cannot state that N_2O emissions from the organic fertilizer treatments were higher than from mineral fertilizer treatments per unit area, thus our second hypothesis was not supported. However, we can see that the organic fertilizers were less efficient than the mineral ones, resulting in significantly lower N offtake in BD and PS and tendencies of higher N_2O emissions in relation to N offtake for BD and PS compared to HM (Fig. 2).

4.3. *Sustained effect of fertilizer treatments during the no-crop season*

Nitrous oxide emitted when soil was without a crop, i.e. between harvest in late summer and sowing in spring, dominated in the year studied. Differences between treatments were similar in the crop season and the no-crop season (Fig. 1), refuting our third hypothesis. This suggests that even when emissions were triggered by freeze-thaw cycles, fertilizer treatments, or rather the N surplus resulting from the treatments, influenced the amount of N_2O emitted. Due to unrecorded losses and other soil processes, this surplus did not reflect the actual amount of mineral N left in soil after harvest, but may still give an indication of the relative N status of the soil in the different treatments.

Contrary to our findings, Russenes et al. (2019) found no differences between contrasting N input rates in emissions during winter and spring thaw in a study of N_2O emissions in spring wheat production in southeastern Norway, where snow covered the soil for at least three months during winter. The lack of treatment effects on freeze-thaw related emissions in that study was attributed to very low soil mineral N concentrations in all treatments in the beginning of winter. We did not have data on soil mineral N contents in the different treatments after harvest in 2015. There are examples in literature where fertilizer N input in the preceding year is reflected in spring-thaw N_2O emissions (Nyborg et al., 1997; Wagner-Riddle and Thurtell, 1998; Lebender et al., 2014), although two of these studies (Nyborg et al., 1997; Wagner-Riddle and Thurtell, 1998) refer to fertilizing in autumn, meaning that there was not, as in our case, a season of crop uptake in between N input and no-crop season emissions.

The high N_2O fluxes generated during freeze-thaw cycles were probably mainly a result of denitrification (Müller et al., 2002; Mørkved et al., 2006; Wagner-Riddle et al., 2008; Risk et al., 2013; Wertz et al., 2018). At temperatures below 0 ˚C, both frozen and liquid soil water can be present, particularly in fine-textured soils (Congreves et al., 2018). Our results show examples of high N_2O fluxes in combination with low soil water content induced by freezing (as low as 12 % VWC) (fig 4). Even in those cases, N_2O may be produced via denitrification, as liquid water can be present in water films surrounding soil particles and denitrifiers active in spite of the soil being partly frozen (Teepe et al., 2001).

4.4. *Fluxes after N input*

Initial measured N_2O fluxes after the May fertilizer application were much higher in 2016 than in 2015 (Fig. 5), and the observed flux peaks appeared earlier. It is possible that there were higher fluxes in the first 24–30 h after application in 2015, particularly as both soil moisture and soil temperature were higher on the first day after application than in the following days, but unfortunately this time period was not included in the measurements (neither was it gap-filled, being before the start of the measuring period). For the cumulative emissions figures, we estimate that the delay in the onset of measurements have minimal influence. Even if the initial fluxes after fertilizer application would have been of the same magnitude as in 2016, the missing time period was short, and therefore they would have made just a small contribution to the total emissions per season or year. The one-year emissions of the treatment with the highest post-application emissions in 2016, BD, would have increased by approximately 10% if the first day after application in 2015 would have had the same fluxes as in 2016.

In 2016, NM, HM and BD all had an immediate response to N input, which was surprising in the case of BD, as the digestate contained plant available N only as NH_4^+ (Fig. 5). It is possible that there was NO_3^--N left in soil from the April input of mineral N, and that the input of organic matter in May triggered reduction of that NO_3^- to N_2O (Stevens and Laughlin, 2001). However, in that case the BD and PS treatments should have had a similar response to organic fertilizer input in May, but the PS treatment showed a discrepant pattern; it had its highest flux measured two days after fertilizer application, when soil moisture had decreased to 22.5 % VWC and temperature was 12 ˚C, i.e., environmental conditions less supportive of denitrification than they were closer to the application event. For applied NH_4^+-N to be emitted as N_2O in the BD and PS treatments, it was either transformed to N_2O and released during nitrification (ammonia oxidation), or had to undergo both nitrification and denitrification either as separate processes, or as nitrifier denitrification. Considering the soil moisture of 20–26% VWC, it is likely that nitrifier denitrification was responsible for a considerable share of the N_2O emissions (Kool et al., 2010; Kool et al., 2011). We speculate that microbial respiration could have been more stimulated in PS than in BD due to more easily degradable C, and that this activity created a higher degree of anaerobiosis in the PS treatment, reducing nitrification and possibly also enhancing N_2O reduction to N_2 (Weier et al., 1993; Miller et al., 2009).

Fluxes of N_2O reached high rates after fertilizer application in HM, BD and PS in 2016, but remained moderate in NM (Fig. 5). A study of N_2O emissions from a loamy soil after application of either pig slurry or mineral fertilizers (N as NH_4NO_3), as part of a long-term management, showed similar results; the mineral N treatment in that study had barely any effect on N_2O fluxes, while the pig slurry treatments had high peaks of N_2O in connection to rainfall after application of N (Rochette et al., 2000). This was explained by higher denitrifying enzyme activity and higher denitrification rates in the pig slurry treatments than in the mineral N treatment closely after N input.

4.5. *Representativity*

In the present study, N_2O fluxes were measured for 13 months, with high frequency, and thus the short-term temporal variability in fluxes was well covered. Several N_2O emission studies have stressed the importance of having enough replicates, because of the high spatial variability of the fluxes (McDaniel et al., 2017; Jungkunst et al., 2018). In this study, there were only two replicates, due to logistic issues. However, the importance of spatial hotspots was probably lessened by the high frequency of the measurements, as hotspots are mostly not long-lasting (Röver et al., 1999). Furthermore, the inclusion of three spatially different stations within each replicate was likely to smoothen potential spatial extremes in cumulative figures or means over time (Section 2.2). In spite of the few replicates and high temporal variation, there were significant differences ($p < 0.05$) or clear tendencies for differences ($p < 0.10$) between the CL and NM treatments on the one hand and HM, BD and PS treatments on the other in ten out of twelve within-season comparisons (and $p < 0.11$ in the remaining two cases). Based on the emission dynamics observed (Fig. 1 and 3), we believe that these differences would have been even clearer had there been more replicates. The analysis of measurement hours where there was no overlap between observed fluxes across two treatments gave a similar picture as the statistical analysis of cumulative fluxes (although they also point at a possible third position of NM, as higher than CL but still lower than HM, BD and PS) (Table 3). This indicates that the observed significant differences and tendencies of differences were a result of frequent and fairly consistent differences in fluxes between the treatments. That two separate analyses, mainly based on the same data, but with independent approaches, give similar pictures of the relative magnitude of the emissions from the contrasting treatments strengthens our confidence in the results.

Whether our results are representative for other years depends to a high degree on weather conditions during the year studied compared to normal, particularly precipitation pattern and freezing periods. Precipitation was lower than normal in June and October 2015 as well as in January, May and June 2016, but this was mostly compensated in the adjacent months. The number of days with mean temperature $\leq 0^{\circ}$C in winter 2015–2016 was nearly identical to the reference period (49 and 52 days, respectively), although the distribution differed slightly. We do not expect these modest deviations from the patterns of the reference period to have had a pronounced effect on the fluxes. On the whole, we estimate that the measured one-year emissions are representative for this soil under cereal production.

In the production area where Lanna research farm is located (plains in northern Götaland), yields of spring barley were normal in 2015 (5120 kg ha^{-1}) (Statistics Sweden, 2014, 2015, 2016). This was substantially lower than the NM yield in our experiment (Table 2), which may have influenced the N_2O emissions. N input in the district was in general lower than the NM treatment, 108 kg N ha^{-1} for fields receiving mineral N only (Statistics Sweden, 2017). Considering both the lower N input and the lower yield, we estimate that the soil N surplus in the production district in general was about 10 kg ha^{-1} higher than in our NM fields, which may have induced slightly higher N_2O emissions.

Similar conditions to those in our study can be found worldwide, regarding barley yields, mineral N input, mean annual temperature, annual precipitation, soil organic matter content, soil texture and soil type, e.g., parts of Europe and northern Asia, eastern North America, southern South America and New Zealand (ArcGIS, 2014, 2017; FAOSTAT, 2020a,b.; Land Data Assimilation System, LDAS, at NASA, 2021; Food and Agriculture Organization of the United Nations, FAO, 2021). We therefore consider the results of this study relevant for large parts of temparate-cool, semi-humid regions of the world.

5. Conclusions

- It is possible to combine the benefits of fertilizer use in terms of good yields with very low N_2O emissions. Lowering N input below recommended rates does not seem to be an efficient strategy to reduce N_2O emissions.
- Emissions of N_2O from recommended mineral N input were low per unit area, yield, N offtake as well as in relation to N input, compared to other fertilizer treatments.
- In the mineral fertilizer treatments, high N_2O emissions were associated with high NO_3^- concentrations in drainage water, but this was not the case for organic fertilizer treatments. Instead, high emissions from those treatments were associated with high input of NH_4^+ and organic matter.
- There is a need for a perspective of multiple years when investigating the effect of organic N sources on N_2O emissions, and the effects of NH_4^+-based fertilizers (mineral or organic) on clay soil.
- Fertilizer effects on N_2O emissions can remain in the no-crop season.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.agee.2022.108062.

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