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Dynamics of nitrous oxide emissions from two cropping systems in southwestern France over 5 years: Cross impact analysis of heterogeneous agricultural practices and local climate variability

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ABSTRACT

Nitrous oxide (N_2O) emissions were measured and compared on 2 typical crop rotations of a grain farm and a dairy farm with feed cropping, over 5 years (from 2012 to 2016) in southwestern France. The annual N_2O emissions of the 5 typical rotational crops of the region (summer crops: irrigated maize and sunflower; winter crops: winter wheat, rapeseed and barley) varied from 0.95 ± 0.88 to 7.96 ± 1.73 kgN ha^{-1}, with the highest values observed on the dairy farm plot and for summer crops. N_2O emissions were analysed on a daily, monthly, seasonal and annual basis, and correlated with their main direct or indirect drivers, i. e. water and nitrogen (mineral or organic) supply amount, rotational crops, vegetation covering and tillage. We observed a marked seasonal pattern of N_2O emission peaks. On average, more than 50% of N_2O emissions occurred during spring for summer crops, and more than 40% occurred in winter for winter crops. We have identified agricultural practices that increase N_2O emissions. In particular, our results show that when the soil is left bare or with limited crop development, spring mineralization of organic N residues (from previous crop or winter cover crop) results in N losses, partly as emissions of N_2O, which are detrimental to agronomic performance (low NUE).

We also conducted an agronomic assessment of annual N_2O emissions versus nitrogen surplus and nitrogen use efficiency (NUE), which lead us to discuss agricultural practices that may mitigate N_2O emissions while optimizing agronomic and economic performance of crops. Indeed, we point out that N surplus and N fate may be controlled through the right timing of sowing, cover crop, irrigation and fertilization.

1. Introduction

At the global scale, atmospheric N_2O molar fraction has constantly increased since the pre-industrial period due to additional anthropogenic sources (Tian et al., 2020). N_2O is a powerful and long-lived trace GHG with a high global warming potential around 300-fold higher than that of carbon dioxide (CO_2), and with an approximate residence time of 116±9 years in the atmosphere (Prather et al., 2015), which makes it the third most important GHG. Between 2009 and 2019, despite the urgent need to decrease GHG emissions (Conference of the Parties on its thirteenth session, 2007), atmospheric N_2O molar fraction has increased at a rate of 0.96 ppb per year at the global scale (Tian et al. 2020; WMO Greenhouse gas bulletin, 2020) meaning that N_2O emissions (production and transport from the soil to the atmosphere) still increase. Tian et al. (2020) estimate that respective natural and anthropogenic N_2O sources contribute on average 57% and 43% of the global N_2O emissions and that the atmospheric chemical sink could offset nearly 80% of these emissions, impending a shortfall of 20%. While it seems difficult to reduce N_2O emissions from natural sources, a major effort must be made to reduce anthropogenic N_2O emissions. Direct emissions from agricultural soil due to nitrogen additions are currently estimated to account for 35% of total anthropogenic emissions. Notably, in developed or developing regions such as Europe, N_2O emissions from agriculture are 2.5 times greater than all other emission sources. This is related to the massive and increasing use of synthetic nitrogen (N) fertilizers in agricultural practices in order to increase agro-ecosystem yields (Davidson,

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2009).

N_2O emissions result from 2 processes, production and transport, from the soil where N_2O is produced to the atmosphere we breathe. Soil N_2O production involves interacting microbiological, physical and chemical drivers. The microbial processes of denitrification in anoxic conditions and nitrification in the presence of dioxygen are generally admitted to be the main sources of N_2O production in both managed and natural soils (Butterbach-Bahl et al., 2013). N_2O production levels directly depend on soil nitrogen content, carbon substrate and dioxygen availability (Robertson, 1989). Ammonium (NH_4^+), nitrate (NO_3^-), organic matter along with water content, which modulates dioxygen and nutrient diffusion, were identified as the major physical-chemical factors modulating the production of N_2O in agricultural soils (Hénault et al., 2005; Li et al., 2000; Parsons et al., 1993; Wijler and Delwiche, 1954). Recently, Hénault et al. (2019) and Žurovec et al. (2021) demonstrated from laboratory and in situ experiments that soil pH favoring the reduction of N_2O into N_2 could act as an important mitigator of N_2O production. Then, diffusion is the major process allowing transport of N_2O from the bottom to the top soil and afterwards to the atmosphere (Heincke and Kaupenjohann, 1999; Yoh et al., 1997; Ball, 2013). Diffusion intensity depends on soil properties, i.e. porosity and texture (in particular clay content) which modulate water circulation and amount in soil (the well-known water filled pore space). Agricultural practices (N-fertilizer application modalities, irrigation, crop residues management, cover crop, tillage depth, etc.) and meteorological conditions constitute a set of interacting direct and indirect abiotic and biotic drivers that modulate N_2O emissions (Skiba and Smith, 2000; Mutegi et al., 2010; Buchkina et al., 2010; Hénault et al., 2012; Tian et al., 2012; Lognoul et al., 2017; Giweta et al., 2017).

Considering agricultural practices, numerous short-term experimental studies highlight that annual N_2O emissions generally increase with the amount of synthetic N applied on a field (McSwiney and Robertson, 2005; Hoben et al., 2011; Rosas et al., 2015; Cardenas et al., 2019; Yao et al., 2019). Interestingly, the level and dynamics of N_2O emission markedly depend on the cultivated species (Yang et al., 2019a), their irrigation needs (Battude et al., 2017) and their inherent fertilization modalities i.e. mineral or organic, liquid or solid, split application, etc. (Misselbrook et al., 2014; Deng et al., 2015; Aita et al., 2015; Harty et al., 2016; Zimmermann et al., 2018). Van Groenigen et al. (2010)Van Groenigen et al. (2010) showed in a meta-analysis that agricultural practices optimizing the plants N-fertilizer use efficiency can significantly mitigate N_2O emissions despite high amounts of applied N. Organic N sources, such as crop residues and/or cover crops incorporated into the soil, have been reported to favor spring N_2O emissions (Pugesgaard et al., 2017) due to decomposer activity which increases the soil mineral N pool and depletes O_2 content (Carpenter--Boggs et al., 2000; Dalias et al., 2002; Guntiñas et al., 2012; Xu et al., 2014; Lacey and Armstrong, 2014; Mitchell et al., 2000). The conclusions of several meta-analyses studying N_2O emissions following crop residues incorporation, based on short-term field and laboratory experiments, are controversial. Chen et al. (2013) and Lehtinen et al. (2014) report a positive relationship between the amount of residue N or C inputs and subsequent soil N_2O emissions. Notably, they found the magnitude of the crop residue N impact to be comparable with the effect of synthetic N-fertilizer on soil N_2O emissions. However, in a meta-analysis integrating 112 scientific assessments at the regional scale, Shan and Yan (2013) found no statistically significant effect of crop residues on N_2O emissions compared with controls. Nevertheless, they conclude that, because of divergent results, more field data are required to reduce uncertainty. Moreover, none of the authors considered a possible time lag between residues incorporation and related N_2O emissions, which requires long-term monitoring of fluxes.

Irrigation strategies also significantly affect N_2O emissions by modifying soil water content and N cycle (transport, mineralization, leaching) (Yang et al., 2019b; Franco-Luesma et al., 2019; Franco--Luesma et al., 2020). In southwestern France, irrigation is widely used to satisfy water requirement of maize crop (Battude et al., 2017) especially early in the growing season when the soil is quite dry and mineral nitrogen easily accessible. The irrigation strategy may then favor N losses, not only *via* nitrate leaching but also *via* N_2O production and emission, occurring to the detriment of the crop growth (Quemada et al., 2013). Tillage depth also influences N_2O emissions by modifying several direct and indirect key drivers of production and diffusion, such as soil density, water infiltration rate, aggregation and aggregates distribution, organic and mineral C and N content along with microbial activity and diversity (Logan et al., 1991). Compared with no or superficial till-management (less than 20 cm deep), deep tillage (more than 20 cm deep) was reported to globally trigger higher N_2O emissions (Ball et al., 1999; Chatskikh and Olesen, 2007; Gregorich et al., 2008; Franco-Luesma et al., 2020). On the contrary, in the case of soil with poor drainage capacity, Rochette et al. (2008) reported that no-till, which is recognized to improve water retention capacity (Copec et al., 2015), increases N_2O emissions compared to tillage. Interactions between the multiple drivers can lead to non-linearity in N_2O emissions (Franco-Luesma et al., 2020). The effect of one management on N_2O emissions can be inhibited or enhanced by another one.

In this context and in light of some example of long-term direct N_2O measurements, the objectives of our study are: (1) firstly, to compare the N_2O emission dynamics of 2 typical crop rotations of a dairy farm (FR-Lam) and a grain farm (FR-Aur) in southwestern France; (2) secondly, to explore and analyze the difference in annual and seasonal emissions of 2 summer crops (sunflower and irrigated maize) and 3 winter crops (wheat, rapeseed and barley); (3) thirdly, to assess the effects of long-term rotation, heterogeneous agricultural practices (N supply, tillage, crop development stage, irrigation) and meteorological variability (rain, soil water content) on N_2O emission dynamics and, (4) lastly, to correlate annual N_2O emissions with agronomic indices. To address these objectives, we benefited from a unique long-term series of daily N_2O emissions (from 2012 to 2016) measured in south-western France.

The marked seasonal and inter-annual local climate variability during these 5 years triggered a wide range of soil water content conditions. This, combined with heterogeneous agricultural practices between the 2 sites (crop rotation choice, tillage depth, cover crop incorporation, irrigation, etc.) allowed us to test the following hypotheses:

- Spring emissions are higher during a summer cropping year than during a winter cropping year;
- Spring emissions intensity depends on vegetation coverage together with the amount of nitrogen returned by crop residues from the previous cropping year and autumn cover crop;
- Cover crop incorporation into the soil by deep tillage enhances N_2O emissions during mild and wet autumn and winter;
- Late spring and early summer irrigation operations on poorly developed crops, combined with high nitrogen availability in the soil, increase N_2O emissions;
- Cultivating summer crops triggers more N_2O emissions than cultivating winter crops
- Crop years with good agronomic performance (low N surplus and high Nitrogen Use Efficiency) are associated with low annual N_2O emissions.

2. Material and methods

2.1. Study sites

In this study, we refer to a set of data collected at Lamasquère (FR-Lam) and Auradé (FR-Aur) sites in south-western France near Toulouse (43°29′47′′N, 1°14′16′′E, 180 m in elevation; 43°32′59′′N, 1°6′22′′E, 250 m in elevation respectively) (Fig. 1). The distance as the crow flies between the two sites is 12 km. Both experimental plots are part of the Regional Spatial Observatory South West (OSR SW), the regional Zone Atelier Pyrénées-Garonne (ZA PYGAR, Ouin et al., 2021), the national

Fig. 1. Location of devices on FR-Lam and FR-Aur sites. Two sets of 3 chambers were installed on either side of the flux station in the prevailing wind direction (west-northwest and east-southeast).

research infrastructure Critical Zone Observatories: Research and Applications (OZCAR; Gaillardet et al., 2018) and the Integrated Carbon Observation System (ICOS; Franz et al., 2018) European network. Since 2005, both sites are fully equipped for the measurement of carbon dioxide, water vapor and energy fluxes (eddy covariance flux tower), meteorological, radiation and soil variables (see Béziat et al., 2009 and Tallec et al., 2013 for details). The FR-Lam crop site is part of an experimental dairy farm (Domaine de Lamothe, Ecole d'ingénieurs de Purpan) located on a plain where a winter wheat – irrigated maize crop rotation is performed. The crops are grown for feed and fertilized with dairy cow excreta or slurry. The FR-Aur crop site is part of a grain farm located in a hilly area where a winter wheat – rapeseed – barley – sunflower crop rotation is performed.

2.2. *Meteorological measurement*

Both sites are equipped with meteorological and flux stations. Matter (CO_2, H_2O) and energy fluxes, meteorological data (temperature, rain, pressure, etc.) radiation and soil variables (temperature, water content and heat fluxes) were measured at a half-hourly time step. The water filled pore space (WFPS) was calculated by multiplying the measured soil water content at 30 cm soil depth by the minimum bulk density measured at 30–60 cm soil depth (1.52 and 1.6 at FR-Lam and FR-Aur, respectively). The measurement methodology for each variable is described in details in Béziat et al. (2009) and Tallec et al. (2013). These variables were used to set up the data set gap-filling procedure as described in Bigaignon et al. (2020).

2.3. *N_2O emissions measurements and computation*

2.3.1. *Automated chambers set up*

To measure N_2O emissions, a set of 6 stainless steel automated chambers (covering an area of 1610 cm^2) were installed late 2011 on each site according to a closed dynamic set up. The chambers were distributed within the footprint of the eddy covariance flux tower (15–20 m (see Fig. 1) to enable gap-filling and GHG budget calculation. The chambers have an elongated shape (70 cm length x 23 cm width x 12.5 cm height above soil) to be easily installed in the crop inter-rows and avoid crop growing inside them. They are designed to measure GHG emitted from soil only (low disturbance from aboveground vegetation and acceptable integration of the flux heterogeneity at fine scale (Bessou et al., 2010)). The lowest 10 cm of the chambers are buried, allowing roots exploring the soil layer underneath. An air pump circulates air at approximately 1 $L.min^{-1}$ between each chamber and two low frequency infrared gas analysers, for the measurement of N_2O and CO_2 molar fractions (N_2O: Thermofisher 46i, Megatec, France, detection limit 0.02 ppm, $\pm$ 1%; CO_2: LI820, LiCor, Lincoln, NE, USA, detection limit 0.5 ppm, $\pm$ 3%). CO_2 molar fraction measurements were used to detect any leakage problems (visual and statistical quality check) and then to filter N_2O data. The chambers were only removed during tillage or harvest. The chambers were left open most of the time and automatically closed alternatively 17.5 min every 6 h, i.e. four cycles a day (00h20–02h20, 06h20–08h20, 12h20–14h20, 18h20–20h20), in order to detect a potential diurnal cycle in N_2O emission and also to avoid any microclimate effect by maximizing rainfall intercept. When the chamber was closed, an internal fan was triggered to ensure air homogeneity. The molar fraction of N_2O was measured every 10 s, leading to a total of 105 measurements over 17.5 min. For a more comprehensive description of the set up see Peyrard et al. (2016) and Tallec et al. (2019).

2.3.2. *N_2O flux calculation*

N_2O fluxes (F_{N_2O}) were calculated for each cycle and each chamber from the slope of concentration vs. time. Data were recorded after a delay of 50 s to account for the travel distance between the chamber and the analyzer (20 m). The set-up design necessarily induced a disturbance of the gas diffusion from the soil after chamber closure (Mello and Hines 1994; Kroon et al. 2008). Given these experimental conditions, no linear fitting could be applied and an exponential regression model (derived from the Fick's law of diffusion) was preferred (Eq. (1)).

$$C(t) = C_{\max} + (C_0 - C_{\max})\left(e^{-kt}\right) \tag{1}$$

with $C(t)$ the N_2O molar fraction at time t, C_0 the initial molar fraction of N_2O and t_0 the first time point when the exponential regression is fitted to the measurement, C_{max} the asymptotic molar fraction of N_2O at infinite time and k the rate constant. The parameter C_{max} and k were adjusted.

N_2O fluxes per chamber and per cycle were then calculated from the determined slope in Eq. (2):

$$F_{N_2O-chamber} = \frac{V_{chamber}}{A}\ \frac{M_m}{V_M} k(C_{max} - C_0)\left(e^{-kt}\right) \tag{2}$$

$V_{chamber}$ is the chamber volume, A is the chamber area, $(C_{max} - C_0)\ k$ the change in concentration within the chamber headspace over time, M_m the molar weight of N in N_2O, V_M the molar volume of an ideal gas under normal temperature and pressure conditions. The flux values were expressed in g N ha^{-1} day^{-1}.

As the flux is not constant and only the initial emission corresponds to the "natural" flux occurring without the chamber closure we retained then $F_{N2O-chamber} = F_{N2O} - (t = 0)$ in Eq. (3):

$$F_{N_2O-chamber} = \frac{V_{chamber}}{A}\ \frac{M_m}{V_M}(C_{max} - C_0)\ k \tag{3}$$

$F_{N2O\text{-}chamber}$ were curated on the basis of goodness-of-fit statistics and visual inspection. Data were removed in case root mean square error between N_2O molar fraction values predicted by the model and the values observed exceeded 20 ppb, or if the rate constant k value was outside the 0.001–0.2 range. The N_2O flux detection limit over the 17.5 min cycle was estimated around 3.9 g N_2O ha^{-1} day^{-1} according to the method described by Neftel et al. (2007).

Daily N_2O fluxes ($F_{N2O\text{-}day}$) were defined as the mean value of all available fluxes during a day (4 cycles x 6 chambers). We considered the daily values calculated with 1 to 5, 6 to 11 and 12 to 24 measurements per day to be "poorly", "moderately" and "highly" representative, respectively. The poorly representative values were removed and gap-filled according to Bigaignon et al. methodology (2020).

From October 2011 to December 2016 (1919 days) at FR-Lam, 1529 daily N_2O fluxes were available. Of these, 67%, 20% and 13% were "highly", "moderately" and "poorly" representative, respectively. At FR-Aur, from January 2012 to the end of September 2013 and from the beginning of October 2014 to the end of December 2016 (1462 days in total for both periods), 1091 daily N_2O fluxes remained. Of these, 57%, 18% and 26% were "highly", "moderately" and "poorly" representative. We decided to discard all "poorly" representative daily values.

2.3.3. Gap-filling of N_2O emissions datasets

Some daily data were missing because of hardware dysfunction, field operations implying the removal of chambers or data removal after quality check (20% and 25% of gap in the time series of FR-Lam and FR-Aur site, respectively). In the aim of estimating consistent N_2O emissions, the dataset used in this study was gap-filled using the methodology described in Bigaignon et al. (2020) which combines linear interpolation and artificial neuronal networks (ANN). A specific ANN was created for each operating period and for each site to maximize the accuracy of the gap-filling procedure. Bare soil and growing season periods of each rotation crop were specifically addressed by the methodology (for details see Bigaignon et al., 2020).

From 2013–08–21 to 2014–10–14, N_2O emissions measurements were stopped at the FR-Aur site, resulting in a huge gap in the data, including two bare soil periods and a winter wheat growing season. As the ANNs developed for the FR-Aur site fitted well with the observations made during operating periods, we decided to gap-fill the winter wheat 2013–2014 growing season with an ANN equation using all data from the other 3 winter cropping periods and another ANN equation was created by gathering all data from bare soil periods to gap-fill the two bare soil periods.

2.4. Seasonal and annual N_2O emissions

Seasonal and annual emissions were calculated by cumulating daily N_2O emissions. The seasonal scale allowed the contribution of each season to annual N_2O emissions to be assessed (January-February-March for winter, April-May-June for spring, July-August-September for summer and October-November-December for autumn). The aim of calculating annual N_2O emissions (from the beginning of October to the end of next September) was to compare cropping years with one another and with the IPCC Tier 1 estimation (De Klein et al., 2006). Two exceptions were made as liquid manure was applied at the FR-Lam site in September 2012 and September 2015: the immediate and significant N_2O emissions recorded in September were allocated to annual N_2O emissions from the following crops, i.e. winter wheat 2013 and winter wheat 2016. As no data was available from October to December 2011 at the FR-Aur site, N_2O emissions were estimated by averaging autumn emissions from the other 3 winter cropping years (2013, 2014 and 2015) which showed a similar pattern.

2.5. Uncertainties calculation

Gap-filling uncertainties and measurement uncertainties were calculated separately and then compiled. Mean daily uncertainties related to the gap-filling procedure ($U_{FN2O-day-gapf}$) were estimated according to Eq. 4:

$$U_{FN2O-day-gapf} = \sqrt{\frac{1}{n}\sum_i \left(x_{simulated,i} - x_{observed,i}\right)^2} \tag{4}$$

$x_{simulated,i}$ and $x_{observed,i}$ are the simulated and observed values at the position i, respectively.

For daily uncertainties related to measurement, we considered the spatial variation to be the main contributor. Therefore, we first calculated the daily standard uncertainty per chamber ($U_{\mathrm{FN2O-temporal}}$) related to temporal variation using Eq. (5):

$$U_{\mathrm{FN2O-day-temporal}} = \frac{SD}{\sqrt{n}} \tag{5}$$

SD is the standard deviation, including Bessel's correction, related to N_2O flux temporal variation and n is the number of $F_{\mathrm{N2O\text{-}chamber}}$ values in a day, per chamber. We then applied the same equation to estimate uncertainties due to spatial variation $U_{FN2O-day-spatial}$ by calculating SD of daily $F_{N_2O-chamber}$ average. Total daily uncertainty ($U_{FN2O-day-meas}$) was then integrated by considering independently $U_{FN2O-day-temporal}$ and $U_{FN2O-day-spatial}$ according to Eq. (6):

$$U_{FN2O-day-meas} = \sqrt{\sum U^2_{FN2O-day-temporal} + \sum U^2_{FN2O-day-spatial}} \tag{6}$$

For multiple chamber measurements over a large field, spatial variation is expected to be the main source of uncertainty. To assess spatial variability, we selected days when all 6 chambers were well operational and calculated the average and the uncertainties due to spatial variation of $F_{N2O\text{-}day}$. For both sites, the absolute daily standard uncertainty related to spatial variation increases positively with daily flux intensity (data not shown). The relative daily standard uncertainty among the 6-chamber set is comparable at both sites, reaching an average of 37 ± 16% (n = 387) at FR-Lam, and 33 ± 17% (n = 250) at FR-Aur (Fig. 2).

To calculate cumulative uncertainties, we considered consecutive daily data integrated over a month as dependent. Consequently, monthly cumulative uncertainties were calculated by summing absolute daily errors according to Eq. (7):

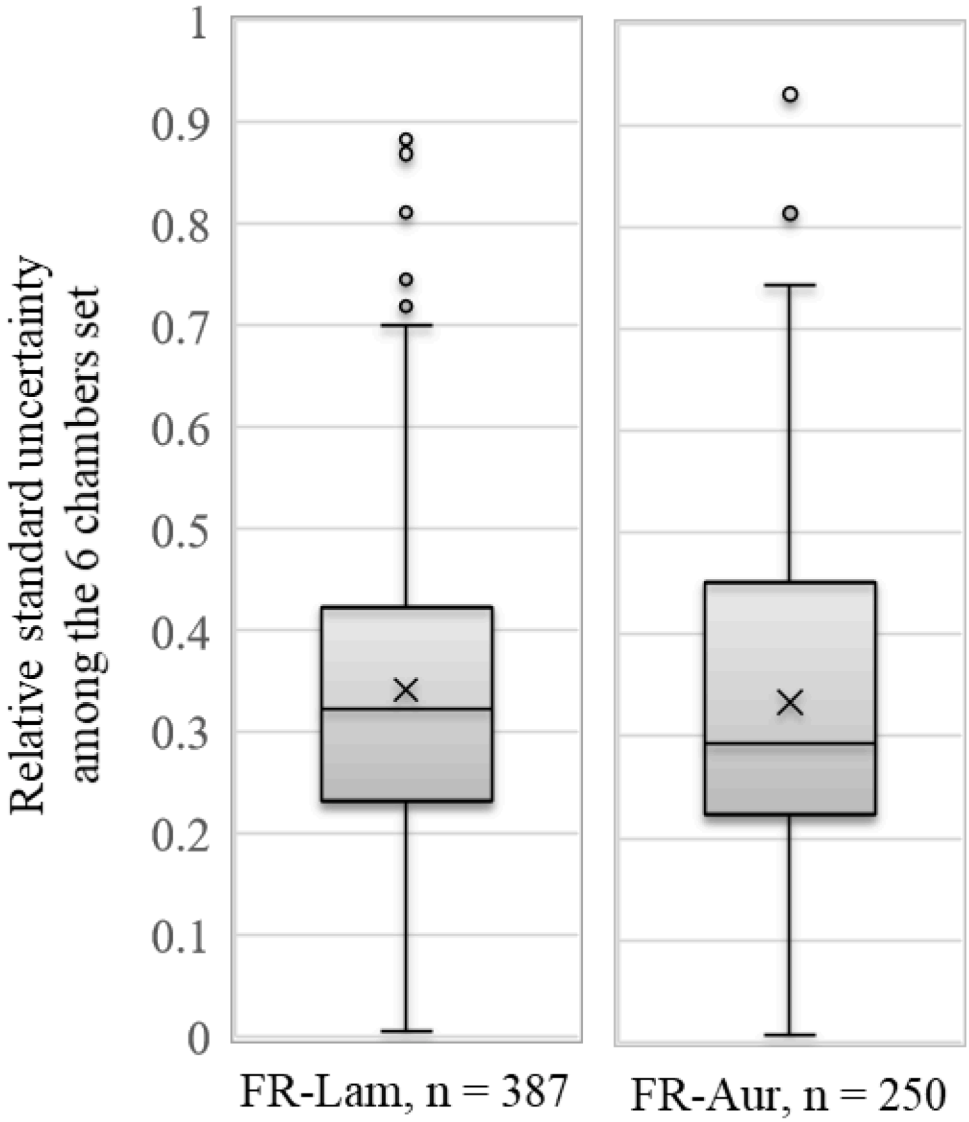

Fig. 2. Relative standard uncertainty among the 6 chambers set at FR-Lam (left panel) and FR-Aur site (right panel). The horizontal line near the middle of the box indicates the median. The x inside the box indicates the mean. The bottom of the box indicates the median of the bottom half or 1st quartile. The top of the box indicates the median of the upper half or 3rd quartile. The whiskers (vertical lines) extend to the minimum and maximum values.

$$U_{FN2O-month} = U_{FN2O-day-meas} + U_{FN2O-day-gapf} \quad (7)$$

As for the calculation of uncertainty on seasonal and annual emissions, we considered monthly emissions values as independent, without autocorrelation. Therefore, seasonal and annual uncertainties were calculated as the square root of the sum of squared monthly uncertainty.

2.6. Nitrogen use efficiency, N surplus

To assess the overall N fate in both agro-ecosystems, FR-Lam and FR-Aur, two indices were considered, based on the calculation methodology from Van Groenigen et al. (2010): (1) Nitrogen Use Efficiency (NUE_{agro}, kgN^{-1}) for each summer and winter cropping year: calculated from an agronomical point of view and defined as the amount of N in the entire plant (aboveground and belowground parts) at harvest (N_{abs}) divided by the amount of annual N supply, i. e. N-fertilizer applied (organic and mineral) and residual N (in crop residues and winter cover crop incorporated into the soil). (2) Nitrogen surplus ($N_{surplus}$) was estimated per hectare by subtracting the aboveground N content from the annual N_{input} amount.

2.7. Vegetation dynamic monitoring

Vegetation dynamics were monitored using destructive measurements to quantify green leaf area index (GLAI), aboveground biomass and total nitrogen content. For sampling, vegetation was collected five times per growing season on 10 to 20 subplots within the area of the 6 chambers set: 1 plant per subplot was collected for maize, rapeseed and sunflower; a length of 50 cm on a row was collected for wheat and barley. GLAI was measured on sampling day with a planimeter after separating the yellow or dead parts from the green parts (Li-3100C, LI-COR, Lincoln, Nebraska, USA). Aboveground dry biomass, yield and nitrogen content were determined at the end of each growing season. The latter allowed quantifying total nitrogen absorbed by the crop over its growing season and nitrogen exported at harvest. After harvest, the aboveground crop residues' nitrogen and carbon content were also estimated by collecting residues from 10 subplots of 50×50 cm within the same area of the crop plot. The organic N return after winter cover crop incorporation into the soil was determined. Nitrogen from belowground biomass residues was calculated according to chapter 11 of the IPCC Guidelines for National Greenhouse Gas Inventories ((De Klein et al., 2006).

2.8. Data analysis

We used an empirical multifactor index ($\frac{WFPS \times N_{supply}}{GLAI}$), combining the cumulative effect of WFPS and of soil mineral N availability weighed by the crop development index, GLAI, to assess the correlation with N_2O emission intensity. N_{supply} is the total amount of mineral and/or organic nitrogen applied (in kgN ha^{-1}), *WFPS* and *GLAI* are the mean WFPS (in %) and the mean GLAI (in m^2 m^{-2}) over the whole three months of the season, respectively. N_{supply} includes N input from fertilizers over the whole three months of the season and also N input from the incorporated crop and/or cover crop organic residues. As organic matter mineralization highly depends on temperature and degree days accumulation (Van Schöll et al., 1997; Delin and Engström, 2010), an experiment (data not shown) was carried out to estimate the best time to study the residual N effect. The optimum degree-day sum to determine the effect of residual N on N_2O emissions was found to be 1040 °C, which occurs on average in early April. In the present study, it should be noted that the amount of N_{supply} from the incorporated crop and/or cover crop organic residues does not represent the effective amount of available mineral N but the total amount measured before incorporation into the soil.

The relationships between variables (see Figs. 5, 6, 7 and 9) were analysed using the Spearman's rank coefficient of correlation (Spearman, 1904). The Spearman's rank coefficient of correlation (*Rs)* is a nonparametric measure of rank correlation (statistical dependence of ranking between two variables). It allowed assessing to what extent the relationship between two variables could be described using a monotonic function by measuring the strength and direction of the association between both ranked variables. The significance of *Rs* was checked by using the Student's t-Test with a 95% confidence threshold.

3. Site properties, climate and management practices

3.1. Soil properties

According to the textural triangle of Malterre and Alabert (1963) the respective soil of FR-Lam and of FR-Aur sites are clay-silty (50.3% clay, 35.8% silt, 11.2% sand, 2.8% organic matter) and clayey to sandy-clay (30.8% clay, 48.3% silt, 19.2% sands, 1.6% organic matter). Bulk density strongly varies in time, especially near the surface on a depth of 0–30 cm, along with rainfall, field operation type and *in fine* soil water content. Integrated on a depth of 0–30 cm, respective bulk densities measured periodically on FR-Lam and FR-Aur sites vary between 1.12 ± 0.12 and 1.58 ± 0.02 and between 1.33 ± 0.09 and 1.73 ± 0.04. Integrated on a depth of 30–60 cm, respective bulk densities on FR-Lam and FR-Aur sites vary less, between 1.52 ± 0.06 and 1.63 ± 0.02, and between 1.6 ± 0.08 and 1.64 ± 0.07. According to the Soil World Reference Base for Soil Resources (WRB), both sites have a Luvisol soil type. Respective pH values measured in June 2020 in the upper soil layer (0–20 cm) of FR-Lam and FR-Aur sites were 6.3 ± 0.4 and 6.6 ± 0.5.

3.2. Local climate and seasonal water supply variability

The climate on both sites is temperate with oceanic and Mediterranean influences, with mild winters, rainy springs, very hot summers with very low rainfall, followed by very sunny autumns. A very close by MeteoFrance weather station (Lherm-Muret) determined a mean annual rainfall of 617 ± 101 mm and a mean annual temperature of 13.7 ± 0.6 °C over the past 24 years on both sites. However, over the studied period, highly variable and contrasted climatic seasons and years occurred (Fig. 3), providing a large panel of climatic conditions to evaluate the impact of agricultural practices and water supply on N_2O

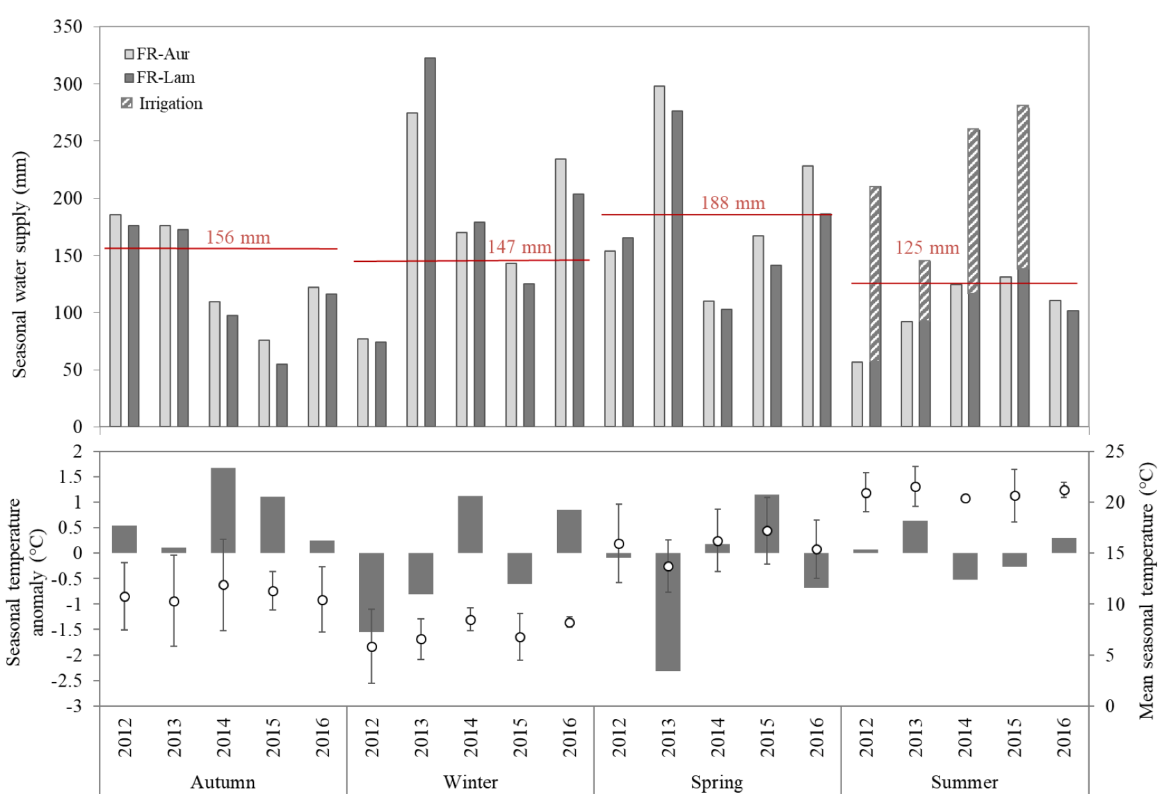

Fig. 3. (a) Seasonal water supply (rain and/or irrigation) on FR-Lam (dark gray bars) and FR-Aur (light gray bars), and mean annual water supply over the 24 past years (red lines) based on a nearby Meteofrance meteorological measurement station. Irrigation amounts are represented by white striped bars. (b) Mean seasonal temperature (right axis, bars are standard deviations) and mean seasonal temperature anomaly of the 5 studied cropping years calculated from the mean seasonal temperature over the past 24 years (left axis). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

emissions. The amounts of rain were quite similar on both sites. Autumn was often as dry as the summer period, while there were similar amounts of rainfall in spring and winter, with particularly high precipitations in 2013 (Fig. 3). Winter 2012 was particularly dry compared to other years. During summer time, the FR-Lam site received around 150 mm additional water when irrigated maize was cultivated. Mean seasonal temperature over the study period varied from a minimum of 5.8 ± 3.6 °C to a maximum of 21.6 ± 2 °C. Overall, Autumns were warmer than the seasonal normal, while other seasons showed more variability over the years. It should be noted that temperatures during winter 2014 and 2016 were higher than normal for the season with mean values of 8.5 ± 1.1 and 8.2 ± 0.5 °C, respectively and that temperatures during winter 2012-spring 2013 were lower than the seasonal normal with mean values of 5.8 ± 3.6 °C and 13.8 ± 2.6 °C, respectively.

3.3. Agricultural practices

At FR-Lam, the management is intensive with exportation of all aboveground biomass for use as bedding material or as feed for the herd in the stable. An annual irrigation of approximately 150 mm is applied in summer when maize is cultivated. Annual N fertilization amounts were as follows: mineral N application varied from 105 (wheat 2013) to 228 (maize 2015) kgN ha^{-1}, whereas organic N application (from livestock manure, liquid or solid) varied from 100 (wheat 2016) to 145 (maize 2012) kgN ha^{-1} (See Table 1 for details). A catch crop was introduced from August 21st to December 6, 2013. At FR-Aur, only the grain is exported while the straw is left on the field. The plot receives only mineral N fertilization with annual amount varying from 0 (sunflower) to 206 (rapeseed) kgN ha^{-1} (See Table 2 for details). Sunflower received no N fertilization given that the low N requirements of this crop are often met by soil contents.

On both sites, soil tillage is regularly carried out at a depth depending on the objectives which may be stubble cultivation, weed control, seedbed preparation, seeding, liquid manure incorporation, etc. In the present study, we focused only on the impact of tillage depth. Based on both plots management information given by farmers and the definitions given in Schneider et al. (2017), we defined tillage deeper than 20 cm as deep tillage, inducing changes in the soil profile, either deep-mixing or deep-ploughing, the latter especially during autumn before a summer cropping year. Superficial tillage (less than 20 cm deep) was defined as subsoiling decreasing the bulk density without turning or mixing soil horizons. Cultivated species and field operations (seeding, harvest, tillage depth, and nitrogen input amount) and the corresponding schedule, are reported in Tables 1 and 2, and illustrated in Fig. 4.

4. Results

4.1. N_2O emission dynamics

Daily N_2O emissions at FR-Lam and FR-Aur sites varied between a common minimum of 0.86 ± 0.09 and a maximum of 399 ± 199 and of 151 ± 73 gN ha^{-1} day^{-1}, respectively (Fig. 4). As expected, daily N_2O emission dynamics varied according to water supply and agricultural practices. Several moderate to high N_2O emissions periods, short or persistent were observed concomitantly with N-fertilizer application or deep tillage. Emissions intensity depended on nitrogen, irrigation, rainfall amounts and inherent WFPS. For winter crops, the highest daily N_2O emission reached 101 ± 58 gN ha^{-1} day^{-1} and was observed for winter wheat in February 2016 at FR-Lam site, when WFPS was high (55%) and synthetic N-fertilizer had been applied one month earlier. For summer crops, the highest daily emission peaks occurred during spring and reached 174 ± 30, 399 ± 199, 160 ± 50 and 151 ± 73 gN ha^{-1} day^{-1} for maize 2012, 2014 and 2015 and for sunflower 2016, respectively, whereas for winter crops, spring emissions only reached 33 ± 29 and 52 ± 30 gN ha^{-1} day^{-1} for wheat 2013 and rapeseed 2013 at FR-

Table 1
Agricultural practices at FR-Lam site: field operations schedule, type of operations (tillage (specified depth), seeding, mineral and organic fertilization (specified amounts), irrigation (specified amounts), harvest).

Crop	Field Operation	Date	Type	Amount/ depth
Maize 2012	Tillage	01/09/2011	Superficial	5 cm
		07/12/2011	Deep	25 cm
		07/02/2012	Superficial	20 cm
		25/04/2012	Superficial	10 cm
	Seeding	27/04/2012		
	Fertilization	15/09/2011	Solid manure	145 kgN ha^{-1}
		30/05/2012	Mineral fertilization	110 kgN ha^{-1}
	Irrigation	29/06/2012		30 mm
		11/07/2012		30 mm
		23/07/2012		30 mm
		02/08/2012		30 mm
		20/08/2012		30 mm
	Harvest	23/08/2012		
Winter Wheat 2013	Tillage	17/10/2012	Superficial	15 cm
	Seeding	29/10/2012		150 kgN ha^{-1}
	Fertilization	04/09/2012	Liquid manure	120 kgN ha^{-1}
		22/02/2013	Mineral fertilization	55 kgN ha^{-1}
		14/04/2013	Mineral fertilization	50 kgN ha^{-1}
	Harvest	22/07/2013		
White Mustard 2013	Tillage	16/08/2013	Deep	25 cm
		20/08/2013	Superficial	20 cm
	Seeding	21/08/2013		
Maize 2014	Tillage	06/12/2013	Cover destruction	
		10/12/2013	Deep	25 cm
		19/03/2014	Superficial	15 cm
		14/05/2014	Superficial	10 cm
	Seeding	15/05/2014		
	Fertilization	09/04/2014	Mineral fertilization	24.7 kgN ha^{-1}
		03/06/2014	Mineral fertilization	102 kgN ha^{-1}
		11/06/2014	Mineral fertilization	72 kgN ha^{-1}
	Irrigation	15/06/2014		25 mm
		20/06/2014		25 mm
		20/07/2014		25 mm
		27/07/2014		25 mm
		12/08/2014		25 mm
				25 mm

(continued on next page)

Table 1 (*continued*)

Crop	Field Operation	Date	Type	Amount/ depth
		25/08/2014		
		09/09/2014		25 mm
	Harvest	22/09/2014		
Maize 2015				
	Tillage	25/09/2014	Superficial	20 cm
		21/10/2014	Deep	25 cm
		26/11/2014	Deep	25 cm
		16/04/2015	Superficial	15 cm
		05/05/2015	Superficial	10 cm
	Seeding	05/05/2015		
	Fertilization	05/05/2015	Mineral fertilization	18 kgN ha^{-1}
		01/06/2015	Mineral fertilization	105 kgN ha^{-1}
		11/06/2015	Mineral fertilization	105 kgN ha^{-1}
	Irrigation	01/06/2015		30 mm
		24/06/2015		30 mm
		05/07/2015		30 mm
		17/07/2015		30 mm
		26/08/2015		20 mm
	Harvest	08/09/2015		
Winter wheat 2016	Tillage	10/10/2015	Superficial	15 cm
		29/10/2015	Superficial	10 cm
	Seeding	20/10/2015		
	Fertilization	23/09/2015	Liquid manure	100 kgN ha^{-1}
		22/01/2016	Mineral fertilization	40 kgN ha^{-1}
		29/03/2016	Mineral fertilization	40 kgN ha^{-1}
		02/05/2016	Mineral fertilization	40 kgN ha^{-1}
	Harvest	20/07/2017		
	Fertilization	09/08/2016	Solid manure	110 kgN ha^{-1}
White mustard + Vetch 2016	Tillage	10/08/2016	Deep	25 cm
		14/11/2016	Deep	25 cm
	Seeding	17/08/2016		

Table 2
Agricultural practices at FR-Aur site: field operations schedule, type of operations (tillage (specified depth), seeding, mineral and organic fertilization (specified amounts), irrigation (specified amounts), harvest).

Crop	Field Operation	Date	Type	Amount/ depth
Winter wheat 2012	Tillage	25/07/2011	Superficial	5 cm
		22/09/2011	Superficial	15 cm
		10/10/2011	Superficial	15 cm
	Seeding	21/10/2011		
	Fertilization	19/01/2012	Mineral fertilization	36 kgN ha^{-1}
		02/03/2012	Mineral fertilization	62 kgN ha^{-1}
		28/03/2012	Mineral fertilization	50 kgN ha^{-1}
	Harvest	14/07/2012		
Rapeseed 2013	Tillage	01/09/2012	Superficial	15 cm
	Seeding	16/09/2012		
	Fertilization	23/02/2013	Mineral fertilization	42 kgN ha^{-1}
		20/03/2013	Mineral fertilization	82 kgN ha^{-1}
		10/04/2013	Mineral fertilization	82 kgN ha^{-1}
	Harvest	05/07/2013		
Winter wheat 2014	Tillage	26/09/2013	Superficial	15 cm
	Seeding	26/10/2013		
	Fertilization	25/02/2014	Mineral fertilization	42 kgN ha^{-1}
		25/03/2014	Mineral fertilization	67 kgN ha^{-1}
	Harvest	10/07/2014		
Barley 2015	Tillage	08/08/2014	Superficial	15 cm
	Seeding	20/10/2014		
	Fertilization	15/01/2015	Mineral fertilization	50 kgN ha^{-1}
		21/02/2015	Mineral fertilization	34 kgN ha^{-1}
	Harvest	27/06/2015		
Sunflower 2016	Tillage	23/09/2015	Superficial	15 cm
		17/11/2015	Deep	40 cm
	Seeding	20/04/2016		
	Harvest	23/09/2016		

Lam and FR-Aur sites, respectively.

At both sites, monthly cumulated N_2O emissions were low during bare soil periods, when no technical operations occurred ranging from 0.05 ± 0.02 to 0.17 ± 0.09 kgN ha^{-1} month^{-1} at the FR-Lam site and from 0.03 ± 0.08 to 0.11 ± 0.33 kgN ha^{-1} month^{-1} at the FR-Aur site. Overall, the highest monthly emissions were observed in spring of summer crop years on both sites. Monthly values reached 1.06 ± 0.33 and 1.01 ± 0.41 kgN ha^{-1} month^{-1} in May and June 2012, 1.46 ± 0.70 and 2.92 ± 1.11 kgN ha^{-1} month^{-1} in May and June 2014 and 1.46 ± 0.43 kgN ha^{-1} month^{-1} in June 2015 for maize at FR-Lam and 1.12 ± 0.53 kgN ha^{-1} month^{-1} in May 2016 for sunflower at FR-Aur site. The intensity of spring N_2O emissions varies with control factors. Indeed, we found a positive correlation between cumulative spring N_2O emissions and N_{supply} amounts (synthetic N-fertilization + crop residues N pool) together with WFPS and weighed by GLAI values (Fig.5). WFPS displays a moderate range of variation (from 40.1 to 52.0%), whereas N_{supply} and GLAI vary much more (from 0 to 302 kgN ha^{-1} and from 0.4 to 6.1 m^{2}m^{-2}, respectively). With coefficients of variation greater than 75%, these last two control factors are the main contributors to the empirical multifactor index, which shows a marked positive correlation with cumulated spring N_2O emissions (Fig. 5). The observed trend appears promising, but further measurements are needed.

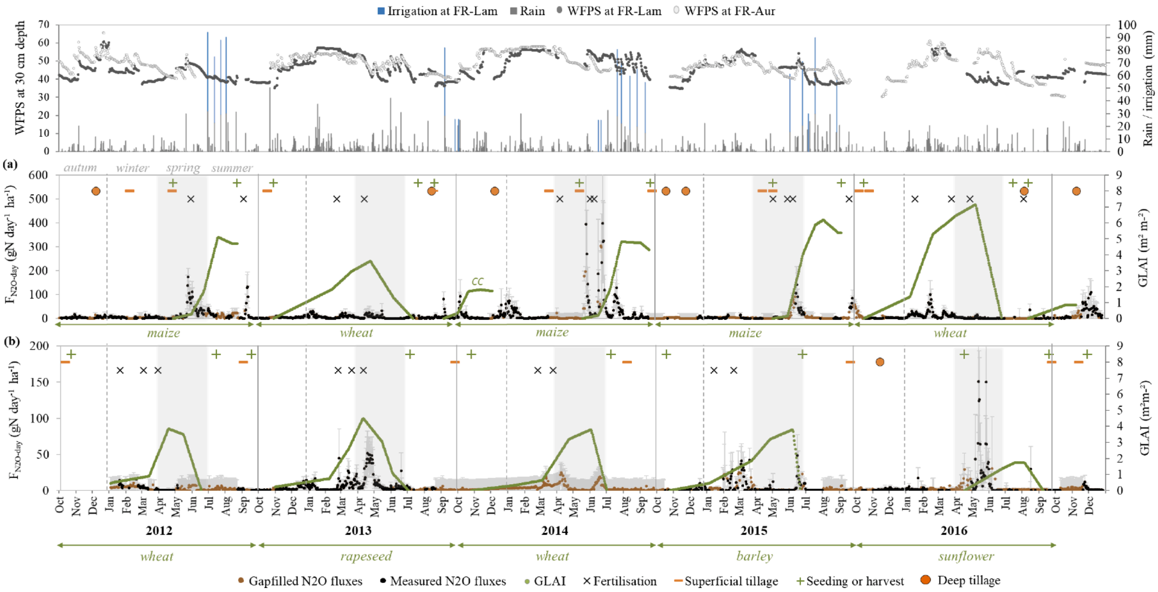

Fig. 4. Dynamics of N_2O emissions, water filled pore space at 30 cm depth, water supply (rain + irrigation) and GLAI, on a daily basis, from October 2011 to December 2016 at FR-Lam (a) and FR-Aur (b) sites. Symbols indicate field operations i.e. tillage, N-fertilizer application, seeding and harvest (see Tables 1 and 2). Error bars (in gray) indicate uncertainties of measured (black dots) and simulated (brown dots) N_2O emissions calculated according to the methodology described in Section 2.5. Scales are different because N_2O emissions are much higher at FR-Lam than at FR-Aur.

N_2O emissions were significantly lower for winter than for summer cropping years, with maximum monthly cumulated values of 0.44 ± 0.26 to 0.75 ± 0.36 kgN ha^{-1} month^{-1} in January and February 2016 and 0.44 ± 0.19 and 0.76 ± 0.38 kgN ha^{-1} month^{-1} in March 2015 and April 2013 at FR-Lam and FR-Aur sites, respectively.

We found that, over the growing season, monthly N_2O are positively correlated with mean monthly WFPS at 30 cm depth, particularly for maize and sunflower (Fig. 6).

We also found that deep tillage operations lead to N_2O emission levels positively correlated with the soil WFPS. Cumulated monthly N_2O emissions after deep tillage vary from 0.04 ± 0.01 to 0.17 ± 0.07 kgN ha^{-1} month^{-1} along with increasing WFPS from 35% to 54% (Fig. 7).

Also, in the case of a concomitant supply of nitrogen in December 2013 and in January 2014, then in November and in December 2016 at FR-Lam, after cover crop destruction and incorporation into the soil by deep tillage (Fig. 4, Table 1) under very wet conditions (54 and 48% WFPS, respectively), cumulated monthly N_2O emissions reached high levels of 0.44 ± 0.31, 0.76 ± 0.42, 0.56 ± 0.43 and 1.09 ± 0.62 kgN ha^{-1} month^{-1}, respectively. For superficial tillage, we found no significant effect on N_2O emissions, regardless of soil WFPS.

4.2. Annual N_2O emissions and seasonal variations

Annual emissions - Table 3 summarizes N_2O annual emissions for both sites along with annual nitrogen supply, NUE_{agro} and $N_{surplus}$. Over the whole study period, overall higher emisions were observed during summer cropping years than winter cropping years. Indeed, annual N_2O emissions for summer crops ranged from 1.90 ± 0.57 (sunflower 2016) to 7.96 ± 1.73 (maize 2014) kgN ha^{-1}, and remained lower for winter crops, ranging from 0.95 ± 0.88 (wheat 2014) to 2.91 ± 0.55 (wheat 2016) kgN ha^{-1}. N_2O annual emissions werehigher at FR-Lam than at FR-Aur with values ranging from 2.31 ± 1.04 to 7.96 ± 1.73 and from 0.95 ± 0.88 to 2.06 ± 0.65 kgN ha^{-1}, respectively. Moreover, annual N_2O emissions of winter crops were higher at FR-Lam than at FR-Aur.

Seasonal variations in N_2O *emissions* - We observed that winter and spring are the seasons contributing the most to annual N_2O emissions for winter and summer crops, respectively (Fig. 8). For winter crops, winter N_2O emissions accounted for 40% of annual emissions on average, ranging from 28% (wheat 2013 at FR-Lam) to 58% (barley 2015 at FR-Aur) of annual emissions. However, we observed a possible delay: at FR-Aur, rapeseed 2013 and wheat 2014 showed higher N_2O emissions in spring than in winter, with respective springtime contributions of 51 and 41% of annual N_2O emissions. For summer crops, spring N_2O emissions accounted for more than 60% of annual N_2O emissions for all maize crops at FR-Lam and even reached 78% of annual N_2O emissions for sunflower 2016 at FR-Aur. Autumn contributed approximately 30% of annual N_2O emissions when slurry was spread at FR-Lam site before a winter wheat crop.

Emission factors variability – Based on annual N_2O emissions, the calculated emission factors range from 0.32 to 3.09% (for winter wheat of 2012 and sunflower of 2016 at FR-Aur, respectively) with a mean value of 1.1 ± 0.80% for the 10 site-years, close to the IPCC default coefficient (De Klein et al., 2006) but however showing a high dispersion.

4.3. Agronomical nitrogen use efficiencies versus annual N_2O emissions

Annual N_2O emissions were compared to N surplus (Fig. 9). On the 10 available cropping years, annual N_2O emissions increased with N surplus, ranging from 0.95 ± 0.88 to 2.91 ± 0.55 and from 1.90 ± 0.57 to 7.96 ± 1.73 kgN ha^{-1} when N surplus ranged from −28 to 110 and from −39 to 261 kgN ha^{-1} for winter crops and for summer crops, respectively (Fig. 9 and Table 3). In contrast, we found a significant negative correlation of annual N_2O emissions with NUE_{agro}. Annual N_2O emissions decreased from 2.91 ± 0.55 to 0.95 ± 0.88 and from 7.96 ± 1.73 to 1.9 ± 0.57 kgN—N_2O ha^{-1} for corresponding NUE_{agro} rising from 68 to 109% and from 41 to 164% for winter crops and summer crops, respectively (Fig. 9).

5. Discussion

5.1. Variation in N_2O emissions: influence of crop rotation choice and agricultural practices

Over the 5 cropping year's rotations, 19.65 ± 2.25 kg N_2O—N ha^{-1} were emitted at the FR-Lam site whereas only 7.60 ± 2.04 kg N_2O—N ha^{-1} were emitted at the FR-Aur site (see Table 3). Our hypothesis that the dairy farm plot emits more N_2O than the grain farm is thus validated.

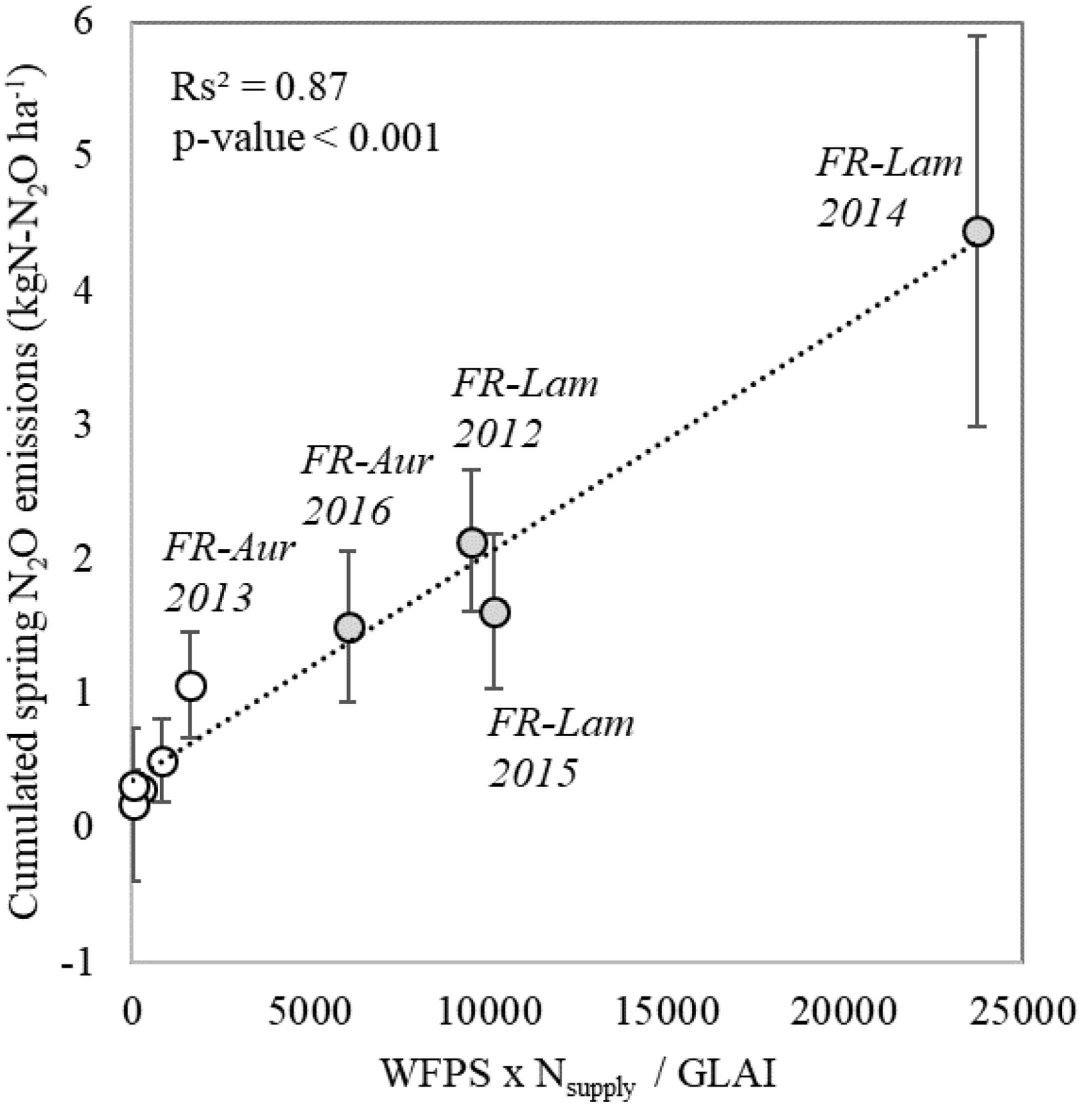

Fig. 5. Cumulated spring N_2O emissions versus water filled pore space (WFPS) at 30 cm depth multiplied by the potentially available soil N weighed by green leaf area (GLAI) for winter crops (white circle) and summer crops (gray circles). Error bars indicate uncertainties of cumulated N_2O emissions (see 2.5 for methodology).

This may partly be explained by crop rotations and differences in agricultural practices. Maize, that was grown over three cropping years at FR-Lam, proved to be a high emitting crop compared to the others. For example, the emission peaks that were observed in June 2012, 2014 and 2015 at the FR-Lam site followed nitrogen fertilization of maize with 110, 174 and 210 kgN, respectively. Daily fluxes registered in our study were similar to those of Dhadli et al. (2016) in an irrigated maize–wheat cropping system in Northern India, where the highest emissions of N_2O were observed during early vegetation growth periods, coinciding with N-fertilizer application. In our study, N_2O emission peaks also coincide with low vegetation development in May (GLAI of 0.2, 0.02 and 0.06 m^2 m^{-2}), medium vegetation development in June (GLAI of 1.3, 0.3 and 2.1 m^2 m^{-2}) in 2012, 2014 and 2015 at FR-Lam, respectively and several irrigation operations. Due to the rapid growth of maize obstructing most technical operations, farmers typically apply plant nitrogen requirements in one or two applications at the very beginning of the growing season, when the phenological demand of the plants for nitrogen is very low. Moreover, at FR-Lam, before the sowing of maize, spring mineralization occurring during long bare soil periods increased soil mineral N availability for other processes than crop production. This, combined with irrigation and/or high rainfall events triggers denitrification and therefore important N_2O emissions. On the opposite, during winter cropping years, lower amounts of synthetic nitrogen fertilizer (ranging from 40 to 82 kgN) were split and applied throughout the growing season period, when vegetation was already well developed, with respective GLAI ranging from 2.3 to 7.1 and from 0.5 to 4.1 m^2 m^{-2} at FR-Lam and FR-Aur sites. Winter crops also proved to be more emissive at FR-Lam compared to FR-Aur (Table 3 and Fig.7). These differences between both sites can be explained by the fact that in autumn before winter crop were sown, high amount of liquid manure was applied at the FR-Lam site in September 2013 (110 kgN ha^{-1}) and 2016 (120 kgN ha^{-1}), immediately causing major N_2O emissions peaks (Fig. 4). Organic N-fertilizers, especially liquid manure (or slurry), is known to cause high and often immediate N_2O emissions, the intensity and persistence of which depend on organic fertilizer type and the application technique (Severin et al., 2016; Herr et al., 2019). N-slurry application further stimulates microbial activity through the additional supply of labile carbon, which can lead to exacerbated anaerobic conditions due to the high oxygen consumption from the mineralization of slurry carbon (Giles et al., 2012; Van Nguyen et al., 2017).

Overall, the annual N_2O emission values at our sites are within the range of those commonly reported in the literature, apart from one specific crop year (2014 maize at FR-Lam). Loubet et al. (2011) measured annual N_2O emissions of 0.8, 1.5 and 3.0 kgN ha^{-1} yr^{-1} in a

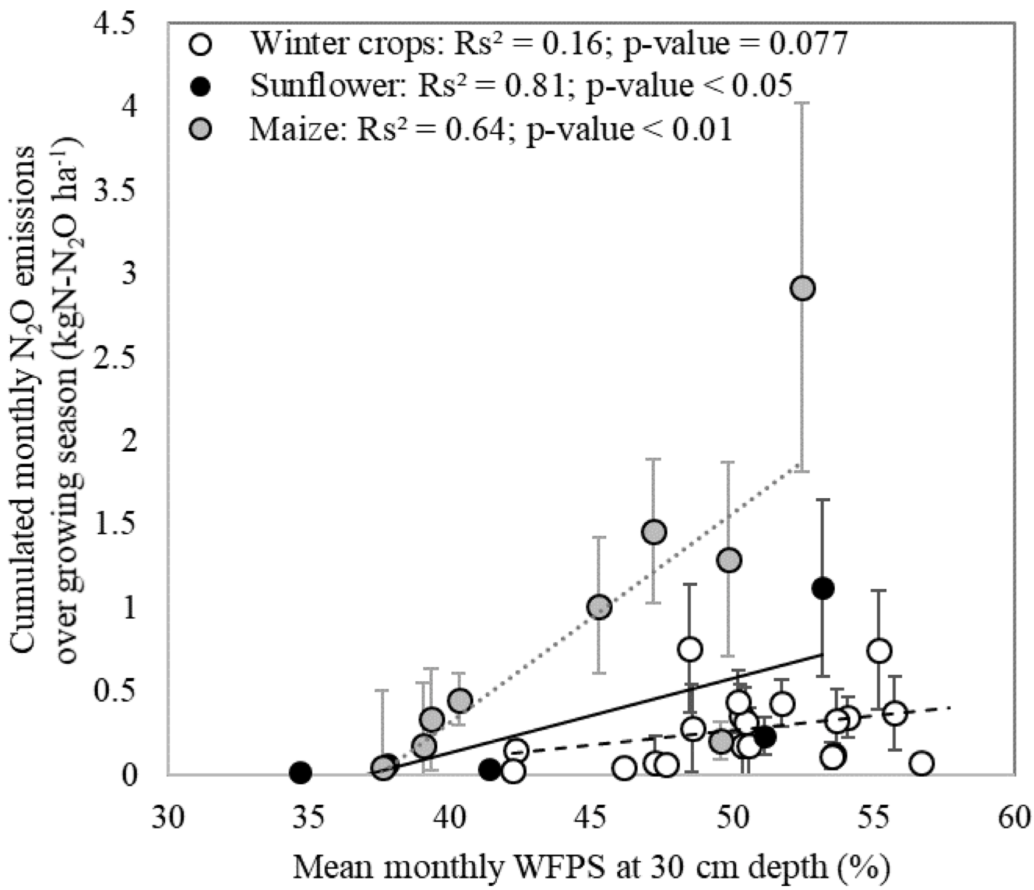

Fig. 6. Cumulated monthly N_2O emissions versus calendar monthly mean WFPS at 30 cm depth over the growing seasons of maize, sunflower and winter crops. Error bars in gray indicate uncertainties (see 2.5 for methodology).

long-term experiment in northern France during winter wheat, barley and maize cropping, respectively. Dhadli et al. (2016) recorded significantly higher cumulative N_2O emission than in the present study during the irrigated maize cropping season (3 months and a half), reaching up to 5.99 N_2O—N kg ha^{-1} with same amount of total N input but with 662 mm irrigation, more than twice that of our study. The annual N_2O emissions measured during winter cropping years in this study were also similar to those reported in other countries. Kaiser and Ruser (2000) reported, in a meta-analysis from multiple crop sites in Germany, mean N_2O emissions values ranging from 1.68 (barley) to 2.82 (sugar beet) kgN ha^{-1} yr^{-1}. Maas et al. (2013) measured a mean annual N_2O emissions of 3.0 kgN ha^{-1} from two winter wheat crops in Manitoba (Canada). During an experiment carried out in the same region than in our study but with different N supply management, Peyrard et al. (2016) reported annual N_2O emissions from a sunflower crop plot similar to our results (sunflower 2016 at FR-Aur, no N input) with annual values reaching around 1.2 and 1.7 kgN ha^{-1} under low N input (96 kg N ha^{-1} year^{-1}) and very low N input (33 kg N ha^{-1} year^{-1}) respectively. Moreover, they also registered spring emissions before synthetic-N fertilizer application. Since N supply was higher in their experiment, annual N_2O emissions should have been higher than those measured for sunflower 2016 at FR-Aur. The main difference is that the precipitation amount was much lower in spring 2012 (150 mm), the year of their experiment, than in spring 2016 (225 mm). Finally, as observed for maize crop at FR-Lam, spring mineralization occurring during a long bare soil period before the sowing of sunflower has led to a high level of free mineral nitrogen in the soil. This, combined with high rainfall events in spring 2016, triggered denitrification and therefore a persistent period of peak N_2O emissions of up to 150 gN ha^{-1} day^{-1}.

5. 2 crop residues-N effect on spring N_2O emissions

This study highlights for the first time the effect of nitrogen from crop residues on N_2O emissions in the subsequent spring (Fig. 5). Our results support the hypothesis that spring emissions were higher during a summer cropping year than during a winter cropping year, due to reduced development of vegetation during spring and the inherent free mineral nitrogen returned by crop residues decomposition from the previous cropping year and/or autumn cover crop. Our results show that N_2O emissions are particularly high in May for all summer crops despite the absence of synthetic-N fertilization. We hypothesize that these N_2O emissions are caused by the decomposition of residues from the previous crop (including cover crop), resulting in a spring mineralization effect. The results of Honeycutt and Potaro (1990) are in accordance with this spring mineralization effect, as they found that plant nitrogen residues from a previous crop harvested in July were mineralized around May of the next cropping year in New England (USA). Based on our 10 crop-years measurements, it clearly appears that the most significant emissions (from 50% to 80% of annual values) occurred before summer crops (maize or sunflower) during the spring season when the soil was bare or when the vegetation was little developed in April/May. The magnitude of spring N_2O emissions was strongly correlated with the amount of residual nitrogen released from the previous crop combined with the mean WFPS and weighted by the vegetation index of the crop plot (Fig. 5). Spring N_2O emissions levels were comparable in 2015 at FR-Lam and in 2016 at FR-Aur (Fig. 5), whereas soil N amount was higher at FR-Lam in 2015 than at FR-Aur in 2016 (see total nitrogen inputs in Table 3, 302 versus 62 kgN ha^{-1}, respectively). The main difference between both lies in water supply, and the inherent water filled pore space: indeed, the amount of rainfall and of WFPS was only

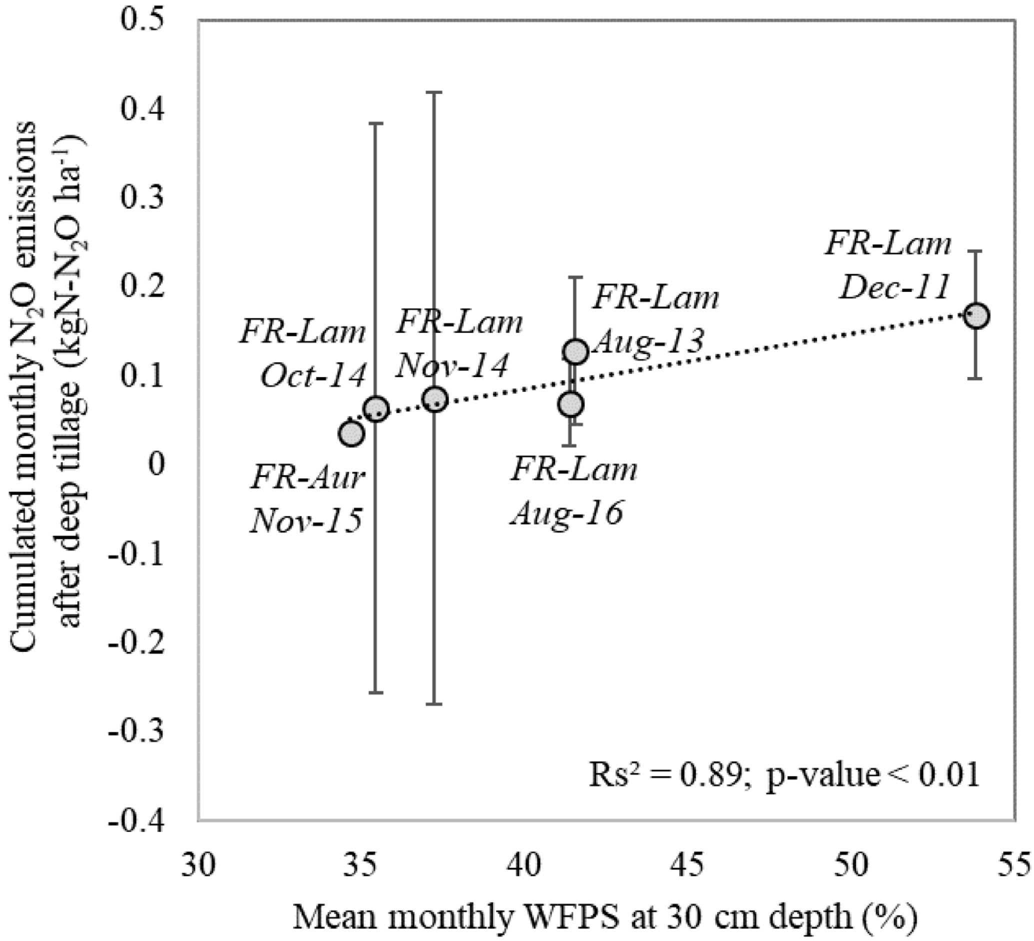

Fig. 7. Cumulated monthly N_2O emissions versus calendar monthly mean WFPS at 30 cm depth after deep tillage events. Error bars in gray indicate uncertainties (see 2.5 for methodology).

Table 3
Annual N_2O emissions (± standard error), NUE_{agro} (expressed as apparent recovery efficiency (in%) of applied N (in kg N taken up per kg N) and nitrogen inputs per cropping year and site. + CC means that the Cover Crop nitrogen content was also included in the total nitrogen residues.

Site	Year	Crop	Annual N_2O emissions (kgN_2O—N ha^{-1})	NUE_{agro} (%)	Nitrogen inputs (kgN ha^{-1}) Fertilizer	Previous crop residues
FR-Lam	2012	Maize	3.84 ± 0.70	75	255	104
	2013	Winter wheat	2.63 ± 0.43	80	225	72
	2014	Maize	7.96 ± 1.73	41	199	130 (+115 cc)
	2015	Maize	2.31 ± 1.04	100	228	74
	2016	Winter wheat	2.91 ± 0.55	68	220	120
FR-Aur	2012	Winter wheat	0.95 ± 0.88	109	147	149
	2013	Rapeseed	2.06 ± 0.65	107	206	106
	2014	Winter wheat	1.29 ± 1.51	93	109	108
	2015	Barley	1.40 ± 0.60	95	88	67
	2016	Sunflower	1.90 ± 0.57	164	0	62

49 mm and 46.9% at FR-Lam in April-May 2015, and 159 mm and 52.0% at FR-Aur in April-May 2016. In addition, maize had a more developed cover than that of sunflower (1.4 versus 0.5 m^2 m^{-2}, respectively), which may have contributed to a better valorization of the soil mineral N. The absence of significant spring emissions when a winter crop is cultivated suggests that well-developed vegetation helps reduce N losses to the environment (nitrate leaching as well as N_2O production and emission). Plants' roots (rapeseed, wheat or barley) extract the mineral nitrogen progressively released into the soil during the spring mineralization period (Mackay and Barber, 1986; Sapkota et al., 2012), thus preventing their access to bacteria that can produce N_2O.

5.3. Winter cover crop incorporation effect on autumn and winter N_2O emissions

Significant and persistent N_2O emissions over the two months following winter cover crop incorporation (by deep tillage) were observed at FR-Lam, corroborating the hypothesis that their deep incorporation into the soil during wet and mild meteorological conditions enhances N_2O emissions. With respective cumulated emissions of 1.2 and 1.65 kg N_2O—N ha^{-1} over two months in December 2013-January 2014 and November-December 2016 at FR-Lam, this phenomenon contributes significantly to annual N_2O emissions, and must therefore be taken into account for the GHG budget and agronomical performance assessment, contrary to the conclusion of Peyrard et al.

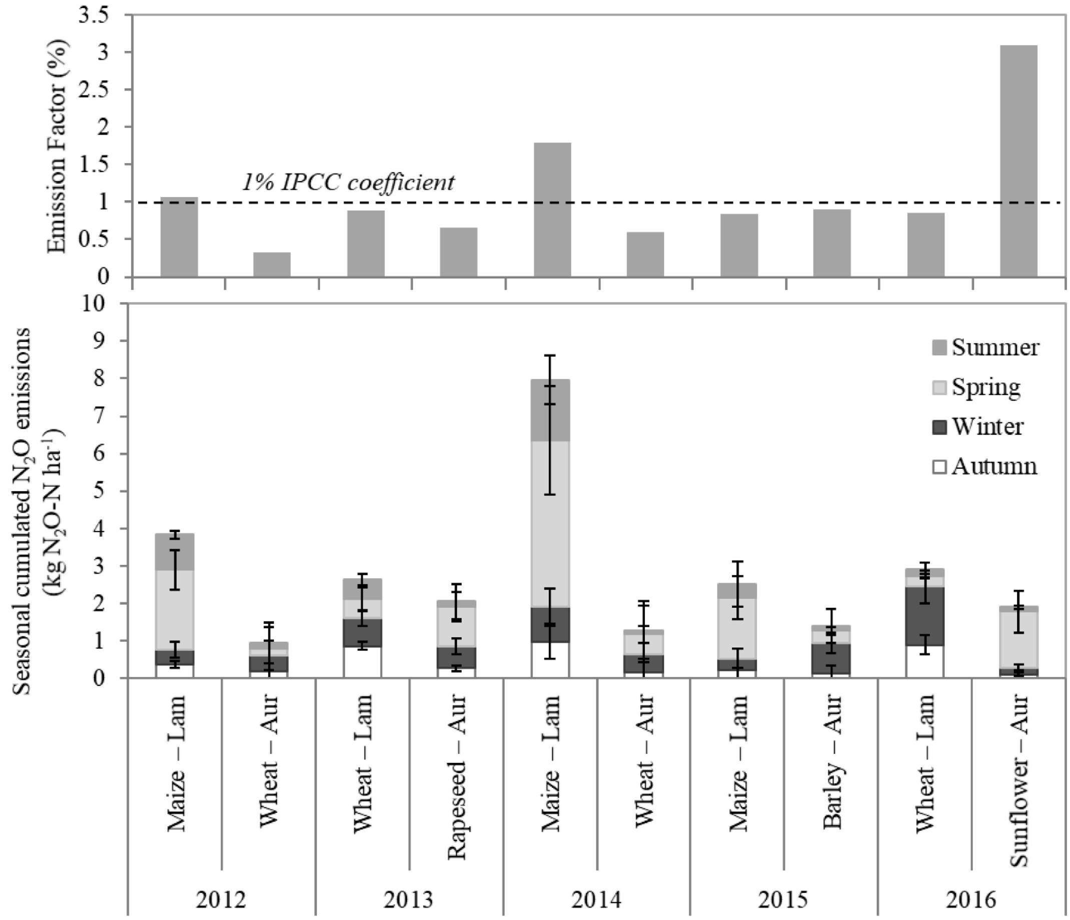

Fig. 8. Seasonal cumulated N_2O emissions (lower panel) and annual emission factor (upper panel) according to cropping years. Annual emission factors were estimated using the IPCC Tier 1 methodology as expressed in the Chapter 11 of IPCC Guidelines for National Greenhouse Gas Inventories (2006). It was calculated from October to October by dividing annual N_2O emissions by total N added to the field (fertilization, residues and cover crop incorporation).

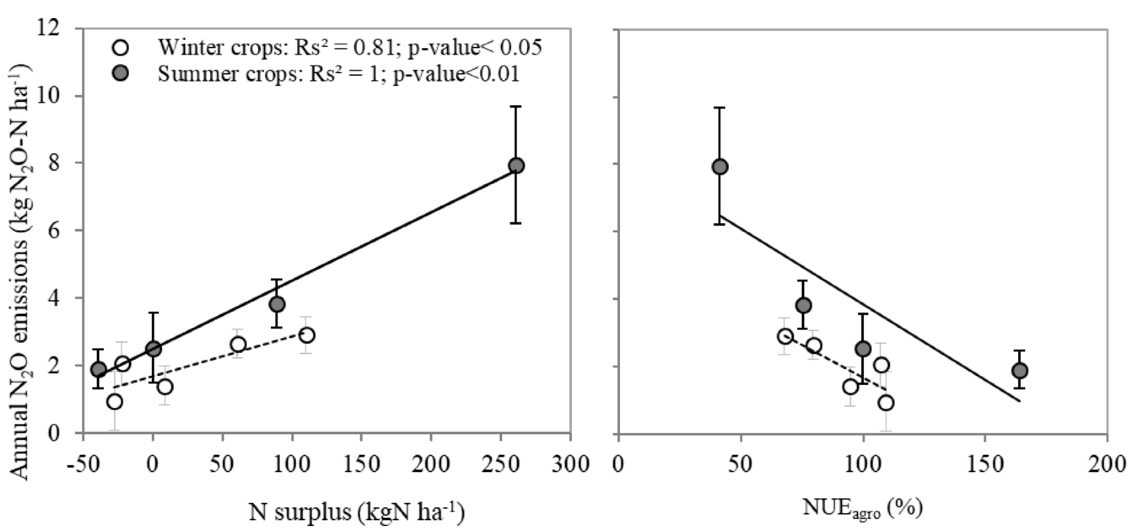

Fig. 9. Correlation between annual N_2O emissions and N surplus (left panel) and agronomical nitrogen use efficiency (NUE_{agro}), expressed as apparent recovery efficiency (in%) of applied N (in kg N taken up per kg N) (right panel) for summer and winter crops of both sites. Error bars indicate uncertainties (see 2.5 for methodology).

(2016). Again, the difference between Peyrard et al. observations (2016; contribution lower than 10 g N_2O—N ha^{-1}) and ours, under similar soil texture condition, probably lies in meteorological and edaphic conditions triggering N_2O emissions under certain circumstances. Indeed, amount of rain (and then WFPS) and air temperature were clearly higher in autumn 2016 (125 mm, 10.5 °C) and in autumn 2013-january 2014 (180 mm, 9.4 °C) than when Peyrard et al. conducted their experiment, in winter 2012 (80 mm, 5.8 °C). Likewise, Chatskikh et al. (2008)

showed an increase in N_2O emissions after a deep tillage of 20 cm on an arable plot following a rainy month of July. They attributed these emissions rather to increased diffusion from the sub-surface, where the gas is primarily produced, to the surface, due to improved soil aeration (Ball et al., 1999), than to increased production related to enhanced organic matter turnover. Shcherbak and Robertson (2019) showed that subsurface N_2O production, which occurs deeper than 20 cm below the surface, may be significant, accounting for up to 50% of total N_2O emissions. These results support our hypothesis that at the FR-Lam site, N_2O production occurs deeper than 20 cm below the surface, where the soil layer is more compact with low air-filled porosity and, combined with water supply, provides anaerobic conditions for denitrification processes (Ball et al., 1999). Besides, deep tillage completely disrupts the top soil layer, thereby promoting N_2O diffusion from the deep layers to the surface.

Furthermore, a second hypothesis to explain the increase in N_2O emissions after cover crop incorporation is that soil disturbance, i.e. soil profile mixing or disruption of the physical protection of the deeper layer, not only increases soil aeration, but also C inputs, decomposition rate, leading to an increase in substrates (dissolved organic carbon, ammonium, nitrate, …) released into deeper layer soil (Balesdent et al., 2000; Van Den Bossche et al., 2009; Lupwayi et al., 2011; Senbayram et al., 2012; Mary et al., 2020) and the subsequent nitrification and denitrification processes leading to N_2O production (Li et al., 2010). We wish to draw attention to the immediate and persistent increase in N_2O emissions caused by deep tillage and cover crop incorporation, which depends both on soil organic matter turnover and on transport processes affecting soil N_2O release.

5.4. Water supply effect on maize N_2O emissions

Soil water content was shown to strongly influence N_2O emissions, and to modulate the effect of others drivers. The impact of water supply appears most clearly during spring season and maize cropping season (Fig. 5 and 6), when the highest daily N_2O emissions were recorded. Irrigation operations, combined with high nitrogen availability (from residues and/or fertilizer), rainy days and elevated temperature, clearly enhance N_2O emissions (Sainju et al., 2012; Dhadli et al., 2016). In our study, this was most strikingly illustrated in 2014 during a maize cropping season at FR-Lam (Fig. 5). In agreement with the findings of Dhadli et al. (2016), higher soil temperature during the maize crop season than during the winter crop seasons (data not shown) under equivalent soil moisture probably boosted the soil biochemical reactions, accelerating microbial activity and intensifying denitrification. Several studies reported the effect of irrigation on N_2O emissions in semi-arid/arid region (Aguilera et al., 2013; Sanz-Cobena et al., 2017; Yang et al., 2019b) and pointed out that irrigation optimization is an important tool to mitigate N_2O emissions (40–70% decrease) in these regions. In their meta-analysis of studies from multiple countries with a Mediterranean climate, Aguilera et al. (2013) reported N_2O emissions ranging from 1.2 ± 1.0 (low irrigation) to 4.0 ± 2.6 (high irrigation) kgN ha^{-1} yr^{-1}. At the FR-Lam site in June 2014, N_2O emissions values reached 2.9 ± 1.11 kgN ha^{-1} $month^{-1}$, when maize was growing and soil nitrogen and moisture levels were high. The reason we found a stronger correlation between N_2O emissions and water supply in maize than in other studies may be because wetting dry soil increases N_2O emissions more than wetting a moist soil (Bergstermann et al., 2011). This may be explained by several interacting processes: an increase in C and N substrate availability (Ruser et al., 2006; Bergstermann et al., 2011) with associated respiratory O_2 consumption, a change in microbial functioning (Kieft et al., al.,1987; Fierer and Schimel, 2003), higher soil organic matter exposure by physical disruption of aggregates (Goebel et al., 2005). Southwestern France is under the influence of the Mediterranean climate. Precipitation is scarce in summer, when temperatures are highest and the soil is dry (see Section 3.2). Most classic maize crops management in the region, e.g. sowing in the middle of spring, requires irrigation of dry soil to achieve worthwhile yields. The combination of dry soil irrigation operations, high temperatures and soil mineral N availability favor N_2O production in southwestern France.

5.5. Fertilization effect on annual N_2O emission depends on N-use efficiency: NUE_{agro} versus annual N_2O emissions

GHG emissions and agronomical N use efficiencies are closely interrelated and highly dependent on fertilization and soil management, as shown by Van Groenigen et al. in 2010. In the present study, we found a positive correlation between surplus N and annual N_2O emissions and a negative correlation between N use efficiency and annual N_2O emissions, both especially marked for summer crops. An increase in N_2O emissions when N supply (applied or available) exceeds N crop demand has been observed in several field studies (Bouwman et al., 2002; Grant et al., 2006; Van Groenigen et al., 2010). Basically, when N supply and crop requirements are not well synchronized during spring, NUE is low and surplus soil N is lost to the environment (Van Eerd et al., 2010). It is of interest to point out that within the range of a nitrogen surplus of approximately 90 kgN ha^{-1} to a nitrogen deficit of approximately -8 kgN ha^{-1}, annual N_2O emissions were lower for winter crops than for summer crops (Fig. 9). This reflects the better uptake and use of progressively mineralized nitrogen by winter crops than by summer crops. The regression lines in Fig. 9, showing a negative correlation between NUE_{agro} and annual N_2O emissions, indicate that increasing NUE can serve both agronomic and ecological purposes by reducing N_2O emissions. Considering summer cropping years, given the high spring contribution to the annual N_2O emissions, the negative correlation between NUE_{agro} and annual N_2O emissions supports the need to improve synchronization of summer crop development with spring soil N offer. Wagner-Riddle et al. (1997) showed that spring contributes the most to annual N_2O emissions from barley, soybean, rapeseed and maize crops sowed in May due to spring-thaw gross N mineralization, with monthly emissions reaching 3 kgN ha^{-1}. They highlighted that when the soil was not bare, spring-thaw N_2O emissions did not occur, thereby supporting our finding that vegetation, when well developed, can inhibit N_2O emission peaks in spring by extracting available soil mineral N. Sowing summer crops earlier in the season, together with adequate N application, at a GLAI stage around 1 m^2 m^{-2} for example, may help increase mineral N (from synthetic fertilizer and spring mineralization) use efficiency and thus reduce N_2O emissions and other nitrogen losses (Van Groenigen et al., 2010; Quemada et al., 2013). Nitrogen losses in the form of volatilized NH_3, runoff and leached NO_3^- were not included in our assessment. We expect these to be positively correlated with N surplus and negatively correlated with NUE, along with N_2O emissions.

6. Conclusion

Overall, over the 5-years, the crop rotation on the dairy farm site resulted in higher cumulative N_2O emissions than the crop rotation on the grain farm. We have identified specific agricultural practices that meet dairy farm needs but lead to increased N_2O emissions: (1) late summer application of liquid manure to bare soil prior to a winter crop, (2) deep incorporation of a winter cover crop during a mild and wet autumn/winter, (3) frequent cultivation of irrigated maize (4) associated with a high amount of nitrogen applied in a short period of time when vegetation is not well developed. We have also identified one agricultural practice resulting in particularly high N_2O emissions at both sites: leaving the soil bare during spring before a summer crop results in a large pool of free and recently mineralized N, which is subsequently released, instead of being processed as spring crop biomass. Although the agronomic assessment of N_2O emissions needs further analysis with longer monitoring of the cropping system, our results point out that, since annual N_2O emissions of summer and winter crops decrease with increasing NUE_{agro}, crops growing with high NUE may mitigate annual N_2O emissions. In light of this finding, providing more ground cover in the spring, either through the introduction of an intermediate cover, or earlier sowing of summer crops in southwestern France, may help reduce N losses of mineralized spring-N (from previous crop residues) and improve agronomic N use efficiency. Lastly, we found that an increase of annual N_{input} does not automatically lead to an increase of annual N_2O emissions (Table 3). Thus, for the improvement of inventory methodology, $N_{surplus}$ is more relevant that N_{input}. $N_{surplus}$ is a variable which integrates the effects of total N_{input} together with agricultural practices and local climate variability, which both influence the crop N use efficiency.

Given the limited data available on the chemical properties of soils in this study, the next step is to run a deterministic model like STICS (Buis et al., 2011) to simulate mineral N availability dynamics (Yin et al., 2020), to thus assess the contribution of nitrification and denitrification to N_2O emissions (Hénault et al., 2005) and finally to simulate mitigation strategies (Plaza-Bonilla et al., 2016).

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.

Data Availability

Data will be made available on request.

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