

Bilans saisonniers et annuels des émissions de N₂O de deux sites agricoles dans le Sud-Ouest de la France impact des pratiques agricoles et de la variabilité météorologique et proposition d'une méthode de calcul

Contexte et résumé de l'étude

La méthodologie présentée dans le chapitre 2 a rendu possible le gap-filling des séries temporelles de FR-Lam entre Octobre 2011 et Décembre 2016 et de FR-Aur entre Janvier 2012 et Décembre 2016, et a permis d'obtenir un jeu de données journalières complet pour l'analyse de la dynamique à l'échelle du mois et des bilans des émissions de N₂O aux échelles de la saison et de l'année. Ce 3^{ème} chapitre de résultats présente la gamme de variation des émissions de N₂O de ces deux sites ainsi qu'une analyse des principaux facteurs d'influence mis en avant dans l'étude pour 5 cultures typiques de la région (blé, tournesol, colza, maïs ensilage, orge). Une nouvelle méthodologie de calcul des bilans est proposée à partir des relations empiriques identifiées entre variables explicatives et émissions de N₂O. Dans un contexte où le besoin d'inventaire des émissions de gaz à effet de serre et de quantification de leur atténuation potentielle à l'échelle d'un territoire est culminant, il est essentiel de pouvoir les réaliser avec le plus d'objectivité et de réalisme possibles.

Dans cette étude, j'ai examiné les effets de la fertilisation azotée, de la pluie et de l'irrigation, du développement des plantes, de la minéralisation de printemps et du travail profond du sol sur les émissions de N₂O. Les relations statistiques obtenues entre les facteurs de contrôle (seul ou en interaction entre eux) et les émissions de N₂O m'ont permis de mettre au point une équation empirique qui simule les émissions de N₂O à l'échelle de la saison et de l'année. Afin d'augmenter la généricité de l'approche inventaire proposée, les jeux de données issus de nos deux parcelles aux gestions contrastées ont été analysés ensemble sans distinction. J'ai ensuite comparé les bilans annuels d'émissions de N₂O obtenus à partir de l'approche empirique aux bilans annuels observés, puis à ceux calculés selon la méthode Tier 1 du GIEC.

Sur les 5 années de rotations étudiées, un total de 19.6 kgN ha⁻¹ de N₂O a été émis sur la parcelle de FR-Lam contre seulement 7.8 kgN ha⁻¹ sur celle de FR-Aur. Cet écart important est

le résultat de pratiques agricoles contrastées avec notamment la culture de maïs ensilage irrigué à FR-Lam, non cultivé à FR-Aur, qui s'est révélée être une culture très émettrice de N₂O ($7.95 \pm 0.40 \text{ kgN ha}^{-1} \text{ an}^{-1}$ pour la seule année 2014). Sur les deux parcelles, les cultures d'hiver présentent en moyenne les émissions de N₂O les plus faibles en comparaison aux cultures d'été. Concernant les cultures d'été (maïs et tournesol), le printemps est la saison où les émissions sont les plus fortes sur l'année en raison d'une forte disponibilité en azote minéral dans le sol issu de la minéralisation de printemps et en l'absence d'une végétation bien développée.

Cette étude met aussi en avant l'importance de la pluie et de l'irrigation dans l'explication des émissions de N₂O qui se trouvent être le dénominateur commun aux autres facteurs de contrôle étudiés. L'effet de chaque pratique agricole (travail de sol profond, fertilisation azotée, minéralisation de printemps des résidus, développement de la végétation) sur les émissions de N₂O est toujours pondéré par la disponibilité en eau (pluie + irrigation) avant ou après l'opération agricole en question.

Les équations empiriques mises au point pour simuler les émissions de N₂O observées se sont révélées très performantes pour modéliser les émissions sur les deux parcelles toutes cultures confondues. Cette étude est une première étape prometteuse vers un inventaire plus informé et précis des émissions de N₂O à l'échelle d'une parcelle agricole. Cette étude a fait l'objet d'un article qui a été soumis au journal « *Agricultural and Forest Meteorology* » en septembre 2020.

N₂O budget from 10 site-years measurement on two crop fields in southwestern France: impact of agricultural practices and calculation methodology

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Abstract

Agricultural managements play an important role in nitrous oxide (N₂O) emissions. However, their effects and their interactions with the meteorological variations have not been fully quantified yet. In this paper, the monthly observed N₂O emissions from 2 crop sites, Auradé (FR-Aur) and Lamasquère (FR-Lam), measured over 5 years (from 2012 to 2016) in the southwestern France were analysed along with their main key drivers. N₂O budgets were compiled and compared for 5 typical rotational crops of the region: winter wheat, barley, sunflower, rapeseed and irrigated maize. The analysis outcomes highlighted significant effect of Green Leaf Area Index (GLAI), tillage depth and N inputs (fertilisation, crop residues and cover crop incorporation) in interaction with water supply (rain and irrigation). The relations found between explanative variables and observed N₂O emissions were used to propose an original calculation methodology to simulate seasonal and annual N₂O budgets at crop plot scale with only few variables. Observed annual N₂O budgets varied from 1.0 ± 0.5 to 8.0 ± 0.4 kgN ha⁻¹ depending on crop species, agricultural practices and water supply distribution. Maize proved to be a particularly high emissive crop with annual budgets ranging from 2.4 ± 0.3 to 8.0 ± 0.4 due to nitrogen fertilisation combined with irrigation event and low crop development at the very beginning of growing season that enhance high soil N surplus. Results underlined that spring mineralisation, when soil is bare, may enhance high monthly N₂O emissions during summer cropping years. For the first time, the effect of deep tillage (> 20 cm) combined with previous mean rainfall on N₂O emissions was quantified and lead to an exponential and positive relationship with a R² score of 0.99. Superficial tillage has no effect whatever the rainfall

amount. The results of the proposed empirical approach to be far more efficient than the traditional IPCC Tier 1 methodology to estimate annual N₂O budgets with R² and RMSE scores of 0.96 and 0.43 kgN ha⁻¹ and of 0.37 and 1.56 kgN ha⁻¹, respectively. This original calculation methodology offers new perspective to inventory N₂O budget in cropland when no measurements exist. However, the approach needs to be evaluated on other sites with different climate, soil and agricultural conditions.

Keywords:

modelling, crop rotation, tillage, mineralisation, nitrous oxide, fertilisation, irrigation

Introduction:

At the global scale, agriculture represents 14 % of the total anthropogenic greenhouse gases (GHGs) emissions (Del Grosso et al., 2009), from which 46 % are due to nitrous oxide (N₂O) emissions from soils (United Nations Environment Program (UNEP), 2012). N₂O is a powerful and long-lived GHG with a high global warming potential, around 300-fold higher than carbon dioxide (CO₂), for an approximate residence time of 120 years in the atmosphere (IPCC, 2013) and it also contributes to stratospheric ozone depletion (Ravishankara et al., 2009, Portman et al., 2012). Atmospheric N₂O molar fraction has increased since the pre-industrial period due to an intensive growing use of synthetic nitrogen (N) fertilizers in agricultural practices in order to increase agro-ecosystem yields (Zheng et al., 2019; Davidson et al., 2009). Between 2007 and 2017, despite the urgent need to decrease GHG emissions (Conference of the Parties on its thirteenth session, 2007), atmospheric N₂O molar fraction has increased at a rate of 0.93 ppb per year at the global scale (WMO Greenhouse gas bulletin, 2018) meaning that N₂O emissions (production and transport near the surface) still increases.

N₂O production in soils is due to numerous processes involving interacting microbiological, physical and chemical drivers. Its intensity directly depends on soil nitrogen content, carbon substrate and dioxygen availability in soil (Robertson, 1989). Ammonium (NH₄⁺), nitrate (NO₃⁻), organic matter along with water content, which modulates dioxygen diffusion, were identified as the principal physical-chemical factors modulating the production of N₂O in agricultural soils (Hénault et al., 2005; Li et al, 2000; Parsons et al, 1993; Wiljer et Delwiche, 1954). Upward transport of N₂O from the bottom to the top soil and then to the atmosphere depends on soil properties, i.e. porosity and texture (in particular clay content)

which modulate water circulation and the major process allowing gas movement within soils, i.e. diffusion (Heincke and Kaupenjohann, 1999; Yoh et al. 1997, Ball et al., 2013). Agricultural practices (via N fertilizer type and quantity applied, irrigation, crop residues management, cover crop, tillage type, etc.) and meteorological conditions constitute a set of indirect abiotic and biotic controllers that modulate the intensity and timing of each direct driver on N₂O emissions.

Considering the agricultural practices, N fertilization is considered as the main key driver of observed N₂O emissions from agricultural soils in temperate regions. Numerous studies highlight that N₂O emissions increase with the amount of N applied on a field (McSwiney and Robertson, 2005; Hoben et al., 2010; Rosas et al., 2011; Reay et al., 2012; Yao et al., 2019). However, N₂O emissions magnitude and dynamics vary greatly in function of crop species (Yang et al., 2019a), fertilization schedule, fertilizers type (mineral, organic, slurry, manure) with less emissions for ammonium based fertilizer compared to the nitrate ones (Misselbrook et al., 2014; Harty et al., 2016; Zimmerman et al., 2018) and input modality with reduced N₂O emissions when the total N supply is split throughout the agricultural season (Deng et al., 2015; Aita et al., 2015). Irrigation strategy, amount and temporal distribution may also affect significantly N₂O emissions by modifying soil water content and N cycle (transport, mineralization, leaching, etc.) within the soil (Yang et al., 2019b, Franco-Luesma et al., 2019). Franco-Luesma et al. (2020) showed that these effects vary according to the irrigation system used: a sprinkler system enabled two-fold higher yield-scaled N₂O emissions (crop yield divided by N₂O budget) compared to a flood system in a Mediterranean Spanish site, due to lower water filled-pore space in the first case than in the second one. Moreover, previous meta-analysis carried out on Mediterranean climate cropping systems by Aguilera et al. (2013) and Sanz-Cobena et al. (2017) reported that the use of drip irrigation could be efficient in reducing direct N₂O emissions (on average 80% lower N₂O emissions than sprinkler systems). Tillage has also been reported to influence N₂O emissions in modifying several direct and indirect key drivers of production and diffusion like 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 reduced till-management, conventional tillage was reported to globally enhance higher N₂O emissions (Ball et al., 1999; Chatskikh and Olesen, 2007; Gregorich et al., 2008; Franco-Luesma et al., 2020). Rochette et al. (2008) showed in a meta-analysis from 25 field study that tillage effect can vary in function of soil type and that no-till only increases N₂O emissions compared to tillage when soils are poorly-aerated. Reduced or no-till management have been proved to provide several environmental

benefits like decreasing surface erosion (Oades, 1984), increasing water retention (Copeck et al., 2015) and carbon sequestration in the uppermost soil layer (Alvarez, 2005; Abdalla et al., 2013). Nowadays, no quantification of conventional tillage effect on N₂O emissions has been evaluated yet. All those controllers' interactions involve a non-linearity in N₂O emissions (Franco-Luesma et al., 2020). Indeed, the effect of one management on N₂O emissions can be inhibited or enhanced by another one: for example, an irrigation event would not have the same impact on a crop site managed with or without tillage (Sainju et al., 2012). In this context, much of the difficulty in managing and quantifying N₂O emissions lies in understanding and modeling the interactions among these controlling factors.

A fine quantification of GHG budget from the agricultural sector is crucial to detect mitigation efforts. However, in the absence of continuously monitored N₂O fluxes, the IPCC Tier 1 default emission factor remains widely used to estimate N₂O annual budget. This methodology only takes into account the annual amount of N input from fertilization and crop residues during a cropping year to calculate the annual N₂O budget. This approach is rather coarse since climate, soil and vegetation conditions and the other agricultural practices are not accounted for. Employing default emission factors may fail to properly represent the observed heterogeneity among local conditions (Reay et al., 2012). More complex models such as DNDC (DeNitrification-DeComposition) and DayCent are able to simulate N₂O fluxes at the plot scale and at a daily time step from crop fields (Li et al., 2012; Del Grosso et al., 2009). Their ability to well reproduce N₂O fluxes depends on the investigated temporal scale. For example, Abdalla et al. (2020) reported high correlation score between observed and simulated N₂O emissions at the annual scale ($R^2 = 0.91$) but poor results at daily time step when using DNDC. In addition, complex models have the disadvantage of requiring multiple parameters and input data like soil properties, climate, plant type, seeding date and rate and management operations that are difficult to obtain especially at large scale.

The long-term field monitoring of daily N₂O emissions is an asset to improve our understanding of the multifactorial short and/or long-term control of those fluxes (Dhadli et al., 2016; Hénault et al., 2012). Indeed, soil N₂O emissions are known to have an important spatial and temporal variability, making them really difficult to catch and predict thoroughly (Hénault et al., 2012). In this context, this study aims (1) at improving N₂O emissions understanding from some typical crops cultivated in southwestern France (winter wheat, rapeseed, sunflower and irrigated maize), (2) at analyzing the interacting effects of both agricultural practices and meteorological variability on seasonal N₂O budget and (3) at evaluating a new simple and

accessible empirical model to estimate seasonal N₂O budget at the crop plot scale. To fulfill that purpose, we benefited from a unique long time series of daily N₂O efflux (from 2011 to 2016) measured in the South West of France on two crop sites with contrasted management.

2. Material and methods

2.1. Study sites

In this study we used a dataset collected at Lamasquère (FR-Lam) and Auradé (FR-Aur) sites in the South West of 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. 4.1). Both experimental plots are part of the Regional Spatial Observatory South West (OSR SW), the regional Zone Atelier Pyrénées-Garonne (ZA PYGAR), the national research infrastructure Critical Zone Observatories: Research and Applications (OZCAR; Gaillardet et al., 2018) and the Integrated Carbon Observation System (ICOS) European network. Each site is therefore fully equipped with instruments to monitor greenhouse gas fluxes, meteorological, radiation and soil variables (see Béziat et al, 2009 and Tallec et al, 2013 for details).

FR-Lam crop site is part of an experimental dairy farm (Domaine de Lamothe, INP) following a winter wheat – irrigated maize crop rotation located in a plain. The soil is mainly clayey (50.3% clay, 35.8% silt, 11.2% sand, 2.8% organic matter). FR-Aur crop site is part of a grain farm following a winter wheat – rapeseed – barley – sunflower rotation located in a hilly area. The soil is mainly silty (30.8% clay, 48.3% silt, 19.2% sands, 1.6% organic matter).

At FR-Lam, the management is intensive with exportation of all aboveground biomass for mulching stable and/or feeding herd. An annual irrigation of approximately 150 mm is applied in summer when maize is cultivated. The annual amount of nitrogen (N) applied varies from 105 (wheat 2013) to 228 (maize 2015) kgN ha⁻¹ and from 100 (wheat 2016) to 145 (maize 2012) kgN ha⁻¹ for mineral and organic fertilisation respectively. N input modalities vary according to crop species. A catch crop was introduced between 21 August to 6 December 2013. At FR-Aur, only grain is exported while straw is left on the field. The plot receives only mineral N fertilisation with annual amount varying from 0 (sunflower) to 206 (rapeseed) kgN ha⁻¹. For both sites, mineral fertilization is split into 3 or 4 applications for winter wheat with a few weeks between and twice for maize at FR-Lam but in a shorter timing (typically one week) than for winter wheat.

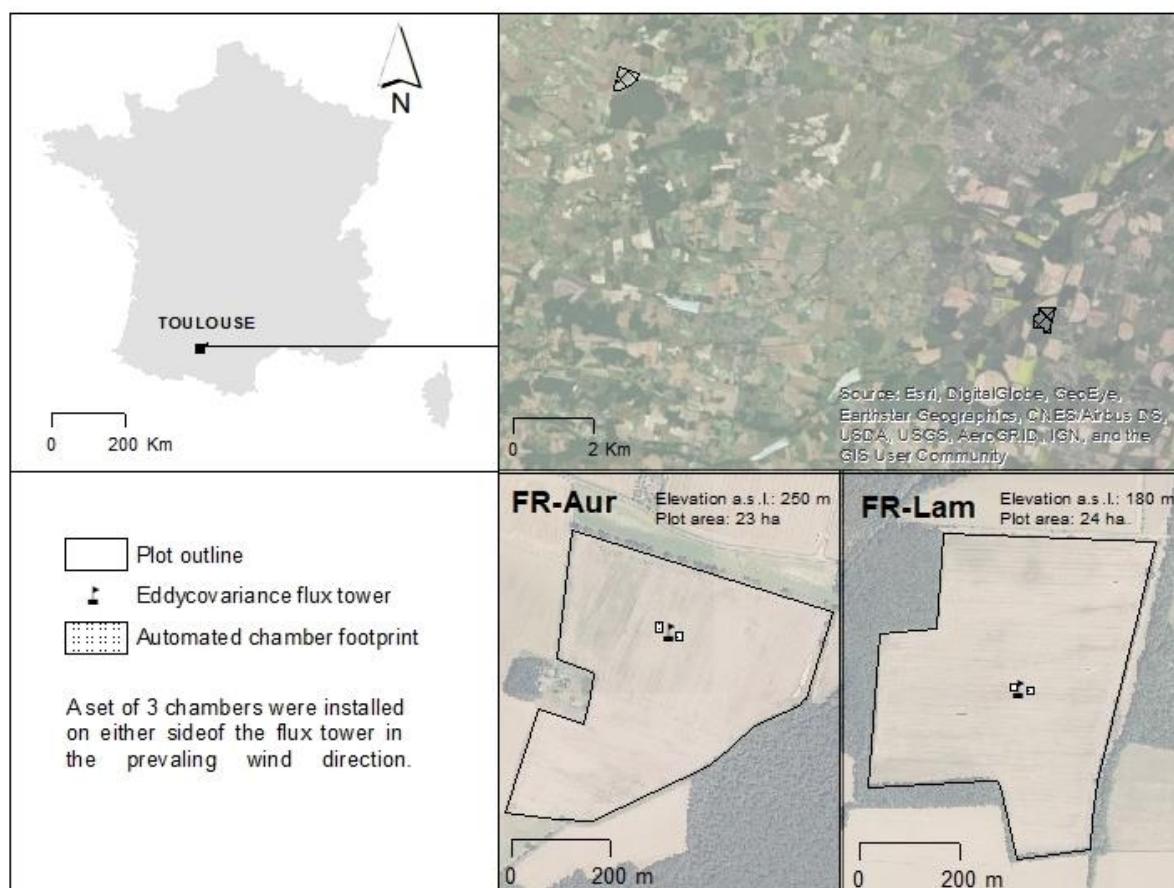


Fig. 4.1. Description and location of FR-Lam and FR-Aur site with the experimental devices' disposal on each site.

2.2. Climate and seasonal water supply variability

The climate on both sites (separated from 12 km, see Fig. 4.1) is a temperate climate with oceanic and Mediterranean influences with mild winter, strong heat and very low rainfall in summer, followed by very sunny autumns. Spring is usually rainy with a mean of 64 ± 28 mm month⁻¹. From 2012 to 2016, the mean annual rainfall was of 610 ± 156 mm and the mean annual temperature of 13.1 ± 0.4 °C on both sites.

Over the period of study, the amount of rain was quite similar on both sites (Fig. 4.2). However, during summer time, when maize was cultivated at the FR-Lam site 150 mm of irrigation was applied. Water supply (rainfall + irrigation) varied between a minimum and a maximum of 50 and 190 mm, 100 and 300 mm, 50 and 130 mm, 75 and 320 mm during autumn, spring, summer and winter seasons respectively. The late autumn was often as dry and warm as the summer period while spring and winter shared similar high wet conditions with particularly very high precipitation in 2013 (Fig. 4.2). Winter 2012 was particularly dry

compared to other years. These figures illustrate well the highly variable and contrasted climatic season and year.

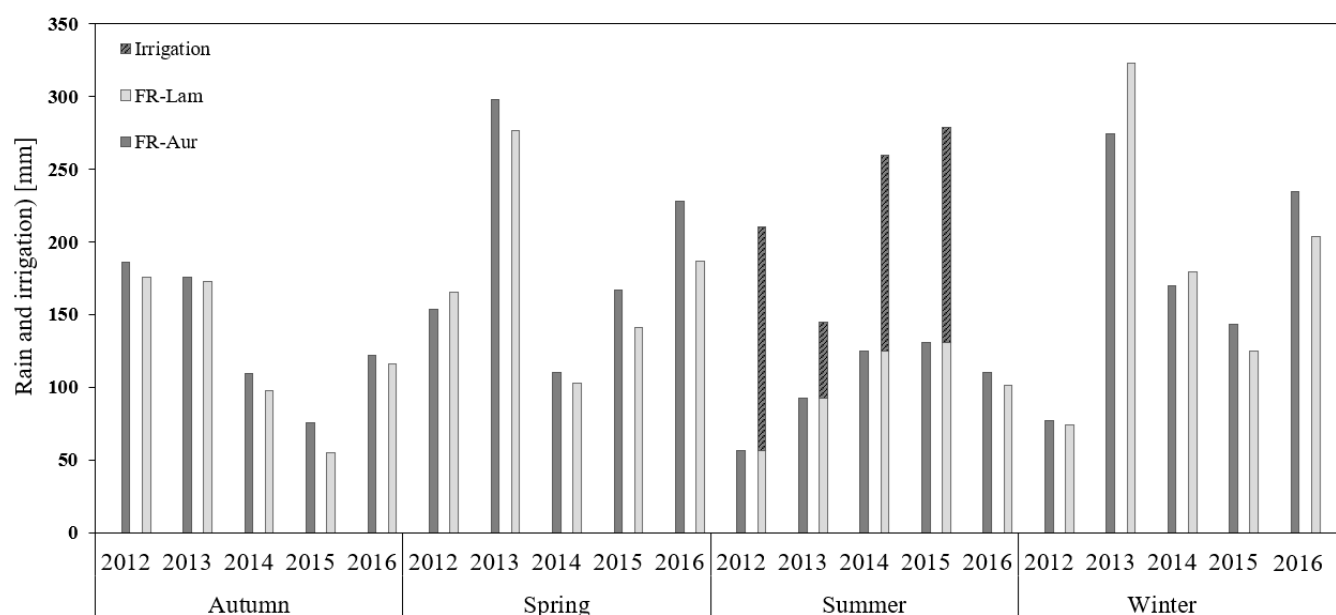


Fig. 4.2. Seasonal water supply (rain and/or irrigation) and water index (difference between the seasonal rain and irrigation of a given year and the mean seasonal rain and irrigation over the 5 years) on FR-Aur and FR-Lam

2.3. N₂O emissions measurements and computation

2.3.1. Automated chambers set up

To measure N₂O emissions, 6 stainless steel automated chambers (covering an area of 1610 cm²) were installed on each plot according to a closed dynamic set up (Peyrard et al., 2016, Tallec et al., 2019). The chambers have an elongated shape (70 cm x 23 cm x 10 cm) to be easily installed in the crop inter-rows where they are placed at a maximum 10 cm soil depth. The chambers were removed before and redeployed after each tillage and harvest. The chambers are open most of the time and automatically closed alternatively 17.5 minutes every 6 hours, i.e. four cycles a day (00h00, 06h00, 12h00, 18h00), to measure the N₂O accumulation into each chamber. A pump maintains a continuous air circulation in the circuit extraction in and out of the chamber at a constant flow rate (1 L.min⁻¹). A fan also enables air homogeneity in the chamber during the measurements (i.e. after the lead of the as tightly closed).

2.3.2. Flux calculation

The N₂O flux, ϕ , is calculated for each chamber and each cycle following Eq. (4.1) to (4.2):

$$\phi = \frac{H}{V_M} C_{max} k \quad (4.1)$$

$$C(t) = C_0 + C_{max}(1 - e^{-k(t-t_0)}) \quad (4.2)$$

with ϕ the N₂O efflux (nmole.m⁻².s⁻¹), C_{max} the asymptote of $C(t)$, k the speed factor equivalent to a gas diffusion coefficient (min⁻¹), $C(t)$ the N₂O molar fraction at time t , C_0 the initial molar fraction of N₂O at chamber closure in ppb, t_0 the first time at which the exponential regression is fitted to the measurement (min⁻¹), H the air height (cm) inside the closed chamber and V_M the molar volume of an ideal gas under normal temperature and pressure conditions, i.e 24.1 L mol⁻¹ at 20°C.

Daily N₂O effluxes were defined as the mean value of all available fluxes during the day.

2.4. Ancillary data monitoring

2.4.1. Vegetation dynamic

Vegetation dynamic was monitored each year using destructive measurements to quantify the green leaf area index (GLAI), the aboveground biomass produced and the total nitrogen content. According to the cultivated crop species, sampling protocol consisted in collecting vegetation five times during a growing season on 10 to 20 subplots inside a footprint area representative of the crop plot: 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. For each sampling date, GLAI was measured with a planimeter (Li-3100C, LI-COR, Lincoln, Nebraska, USA). Aboveground dry biomass and related nitrogen content were determined at the end of the growing season. The latter allowed quantifying the total nitrogen absorbed by the crop over its growing season and the crop residues nitrogen content after harvest or the organic N return after cover crop incorporation into soil. Nitrogen from below ground biomass residues was calculated following the Chapter 11 of IPCC Guidelines for National Greenhouse Gas Inventories (2006).

2.4.2. Meteorological data

Both sites are equipped with complete meteorological and flux stations. Matter (CO₂, H₂O) and energy fluxes, meteorological data (temperature, rain, pressure, etc) radiation and soil variables (temperature, water content and heat fluxes) were available at a half-hourly time step. 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 gap-filling procedure as described in Bigaignon et al. (2020). For the main purpose of this study, the only meteorological variables used were the air temperature and the amount of rain. Soil water content (SWC) is widely recognised to control N₂O emissions but the SWC datasets on both sites were not complete enough to satisfy our needs and then discarded from the analyses. Rain, especially integrated over a month, is a good proxy of SWC conditions on clayey-silty soil and constitute an enough high relevant explanative driver of the N₂O emissions variations. Moreover, the amount of rain has the advantage to be easy to assess and could facilitate the generalisation for the proposed inventory approach in this study.

2.5. N₂O budgets

For the purpose of our study, monthly, seasonal and annual budgets were calculated by cumulating daily N₂O emissions. The seasonal scale allowed evaluating the contribution of each season on the annual N₂O budget. The aim of calculating annual N₂O budgets was to compare the different cropping years with each other and with the IPCC Tier 1 estimation. Seasons were defined as January-February-March for winter, April-May-June for spring, July-August-September for summer and October-November-December for autumn. Annual budgets were calculated for the 12 months of each cropping year, i.e. from the beginning of October to the end of September of the following year. For years where slurry was applied at the FR-Lam site (September 2012 and September 2015), the related and significant immediate N₂O emissions recorded in September were allocated to the following annual crop N₂O budget, i.e. winter wheat 2013 and winter wheat 2016. As no data was available from October to December 2011 at the FR-Aur site, annual N₂O budget for the period 2011 – 2012 was calculated by simulating the missing period with the empirical function described in section 3.4.4 chapter 4.

2.5.1. Gap-filling of N₂O emissions datasets

Because of hardware dysfunction or field operations, such as tillage and harvest, chambers were removed many times, some data were missing which means that the number of available N₂O flux measurement per day to calculate a daily N₂O flux varied from 0 to 24. Finally, from October 2011 to December 2016 at FR-Lam, from 1919 theoretical daily N₂O flux values, only 1529 remained. The whole dataset included 62% of “highly” representative daily values (calculated with 12 to 24 measurements a day), 23% of “moderately” representative daily values (calculated with 6 to 12 measurements a day) and 15% of potentially “poorly” representative daily values (calculated with 1 to 6 measurements a day). At FR-Aur, from January 2012 to December 2016, over 1827 theoretical daily N₂O flux values, only 1091 remained. The whole dataset included 54% of “highly” representative daily values, 16% of “moderately” representative daily values and 30% (mainly due to the year 2014, see section 2.4 chapter 4) of potentially “poorly” representative daily values.

In the aim of estimating consistent N₂O budgets, 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 functioning period and for each site to maximize the performance of the gap-filling procedure. Bare soil was discriminated from growing season period, the latter being also divided according to the cultivated crop (for details see Bigaignon et al., 2020).

From 2013-08-21 to 2014-10-14, N₂O emissions measurements were stopped at the FR-Aur site, resulting in a huge gap in the data. This gap period included two bare soil periods and a winter wheat growing season. As the ANNs developed for the FR-Aur site fitted well the observations in the same functioning period, we decided to gap-fill this period with two ANN equations based on all available data. To do so, an ANN equation was developed using all data from winter crop periods (wheat 2011-2012, rapeseed 2012-2013 and barley 2013-2014) and then used to gap-fill the winter wheat 2013-2014 growing season. Another ANN equation was created by gathering all data from bare soil periods to gap-fill the two bare soil periods.

2.5.2. Uncertainty calculation on the N₂O budget

Uncertainty was calculated using the standard deviation σ :

$$\sigma = \sqrt{\frac{1}{n} \sum_{i=1}^n (x_i - \bar{x})^2} \quad (4.3)$$

with n the number of values used to calculate the uncertainty, x_i the value at the position i and \bar{x} the average of the values taken by x_i . Daily uncertainties in N₂O fluxes related to the set up (spatial and temporal sampling protocol) were determined by calculating the standard deviation of all measurements available on that day ($\sigma_{measurements}$). If only 1 measurement was available on a day, the uncertainty on that day was calculated by taking the square root of the mean squared uncertainty of the six daily uncertainty values surrounding the given N₂O efflux value.

A daily uncertainty related to the gap-filling procedure ($\sigma_{gap-filling}$) was calculated using the standard deviation of the distance between simulated and observed N₂O effluxes (Bigaïgnon et al., 2020; Eq 5):

$$x_i = x_{simulated,i} - x_{observed,i} \quad (4.4)$$

with $x_{simulated,i}$ and $x_{observed,i}$ being the simulated and observed value at the position i , respectively.

Monthly and annual N₂O budget uncertainties were then calculated following Eq. 4.5:

$$\sigma = \sqrt{\sum \sigma_{measurements}^2 + \sum \sigma_{gap-filling}^2} \quad (4.5)$$

2.5.3. IPCC Tier 1 methodology

Annual N₂O budgets were estimated using the IPCC Tiers 1 methodology from October to October as 1 % of the total N added to the field (fertilisation, residues and cover crop incorporation) during the considered cropping year as expressed in the Chapter 11 of IPCC Guidelines for National Greenhouse Gas Inventories (2006). The IPCC factor was also compared to the actual Emission Factor (EF) from each cultivation year.

2.5.4. Nitrogen Use Efficiency, Yield-scaled N₂O emissions, N surplus

To evaluate the effect of soil N level on crop production and on N₂O emissions, and to determine potential interactions, three indexes were calculated: (1) Nitrogen Use Efficiency ($NUE_{agro} = \text{kgN kgN}^{-1}$) for each maize and winter wheat cropping years were calculated from an agronomical point of view and defined as the quantity of N in the entire plant at harvest (N_{abs}) divided by the amount of annual N supply, i. e. N fertilizer applied (organic and mineral) and residual N (crop residues and winter cover crop incorporation in the soil). NUE_{agro} was expressed as a percentage by multiplying value by 100; (2) Yield-scaled N₂O efflux (expressed

in g N₂O-N kg⁻¹ aboveground N_{uptake}) were determined for each maize and winter wheat cropping years by dividing annual N₂O budget (FN_2O_{annual}) by the total aboveground N content (N_{abs}); (3) A nitrogen surplus index ($N_{surplus}$, kgN ha⁻¹) was estimated for all cropping years by subtracting the aboveground N content to the annual N_{input} amount.

2.5.5. Statistical evaluation

The statistical evaluation of the relationship found in this study between the different environmental factors (LAI, water supply), technical operations (tillage, N input) and observed N₂O emissions and the models evaluation were carried out using the determination coefficient as the square of the Pearson correlation coefficient (R^2 , Eq. 4.6) and the Root Mean Squared Error (RMSE, Eq. 4.7) to evaluate the differences between the observed and the estimated values.

$$R^2 = \frac{(\sum_{i=1}^n (p_i - \bar{p})(o_i - \bar{o}))^2}{\sum_{i=1}^n (p_i - \bar{p})^2 \sum_{i=1}^n (o_i - \bar{o})^2} \quad (4.6)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^n (o_i - p_i)^2}{n}} \quad (4.7)$$

2.5.6. Optimisation procedure of agricultural practices effects on N₂O emissions and empirical model parametrisation

In this study, the effects of agricultural practices effect on N₂O emissions were investigated. As it is known that it exists a lag between N₂O emissions and the context that trigger those emissions, an investigation to fix the average period during which a process is leading to N₂O emissions was carried out. Thus, the number of days that need to be taken into account to evaluate the effects of fertilisation, N residue, tillage and water supply on N₂O emissions was defined after an optimisation procedure that aimed to get the highest R^2 score for the fit of the empirical equation (Eq. 4.9) presented in section 3.4.4 chapter 4. This optimisation consisted to test and fix the number of days to calculate (1) the mean water supply (W_{mean}) related to a fertilisation, a spring mineralisation or a tillage event, (2) the mean GLAI ($GLAI_{mean}$) related to a fertilisation and (3) the cumulative N₂O emissions (F_{N_2O}) resulting from these events with a time window of 1 to 100 days.

3. Results

3.1. Dynamic of monthly cumulated N₂O emissions

Monthly cumulated N₂O efflux varied between 0.03 ± 0.07 and 2.93 ± 0.25 at the FR-Lam site and between 0.03 ± 0.00 and 1.14 ± 0.08 kgN ha⁻¹ at the FR-Aur sites (Fig. 4.3). At the cropping year scale, the monthly N₂O emissions dynamic did not follow any seasonal pattern. The dynamic depended more on the crop type, the water supply and the agricultural practices.

At both sites, monthly cumulated N₂O emissions were low during bare soil periods, when no technical operations occurred, ranging from 0.05 ± 0.00 to 0.17 ± 0.02 kgN ha⁻¹ at the FR-Lam site and from 0.03 ± 0.00 to 0.11 ± 0.08 kgN ha⁻¹ at the FR-Aur site. During the growing seasons, the highest emissions were mainly observed during summer crop cultivation whatever the site (maize in 2012, 2014 and 2015 at FR-Lam; sunflower in 2016 at FR-Aur). The strongest peaks in N₂O emission were observed at the FR-Lam site during the maize growing periods with monthly cumulated emissions reaching 1.1 and 1.0 kgN ha⁻¹ in May and June 2012, respectively, 1.5 and 3.0 kgN ha⁻¹ in May and June 2014, respectively, and 1.5 kgN ha⁻¹ in June 2015. No large cumulated N₂O emissions were observed in May 2015 at FR-Lam before maize was grown where the monthly cumulated N₂O emissions did not exceed 0.11 kgN ha⁻¹ that year.

During winter crops, the highest monthly cumulated N₂O emissions was 4 times lower than for the summer crops, ranging from 0.08 ± 0.00 to 0.75 ± 0.06 and from 0.03 ± 0.00 to 0.76 ± 0.05 kgN ha⁻¹ at FR-Lam and FR-Aur, respectively.

Depending on the previous month soil humidity conditions and following deep tillage (deeper than 20 cm depth; T, Fig. 4.3), monthly cumulated N₂O emissions peaks occurred after tillage either in September 2013, January 2014 or in December 2016 at FR-Lam leading to monthly N₂O budget of 0.5, 0.8 and 1.1 kgN ha⁻¹, respectively. Those effects were not observed after a superficial tillage (lower than 20 cm depth, data not shown).

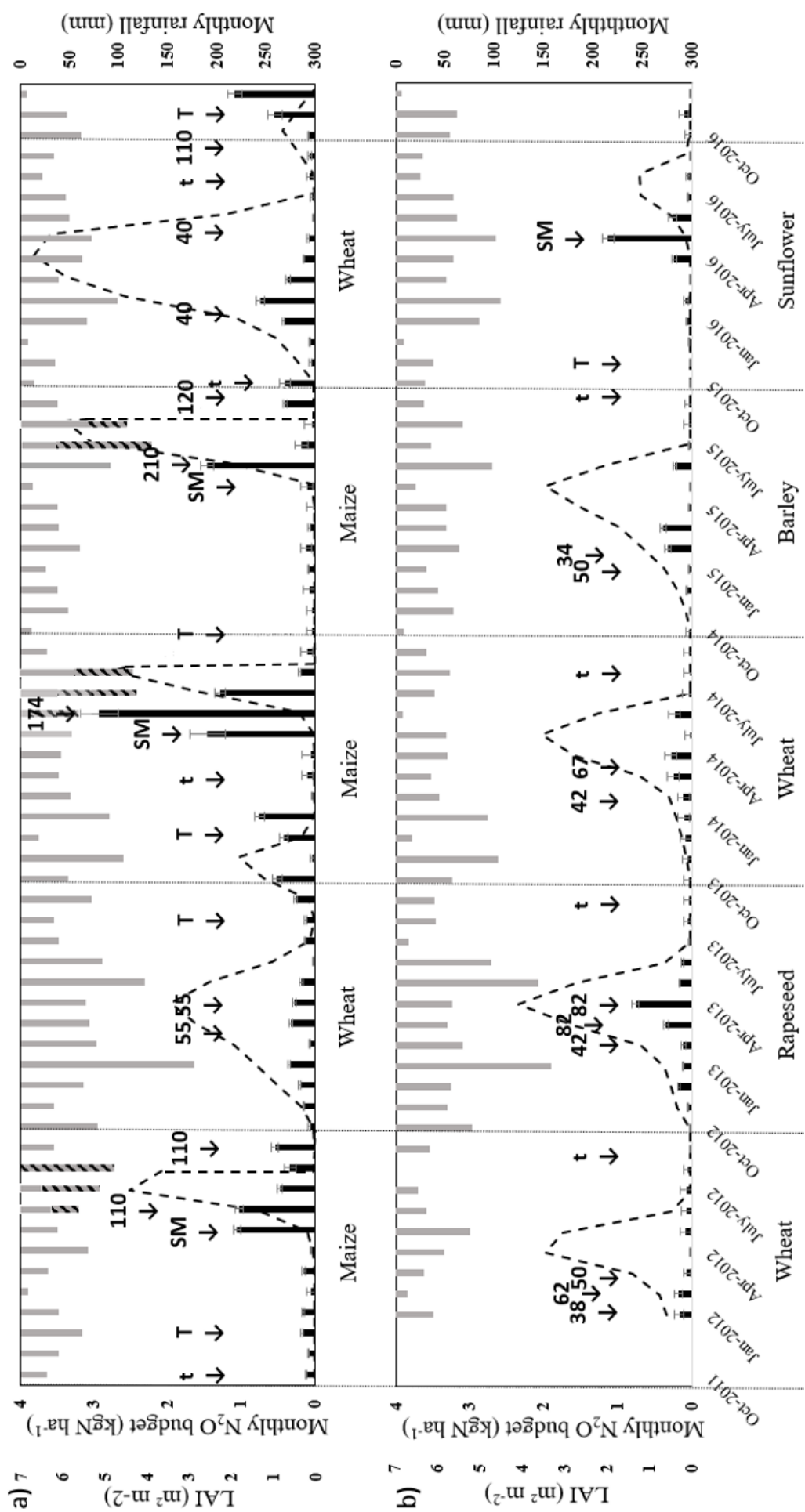


Fig. 4.3. Monthly cumulated N₂O emissions (dark grey bars), mean rain (light grey bars), mean rain + irrigation (dashed light grey bars), GLAI (dashed line) and field operations from October 2011 to December 2016 at FR-LAM (a) and FR-AUR (b) (T, tillage > 20cm; SM, Spring Mineralisation). Bars indicate uncertainties of the cumulated monthly N₂O emissions calculated according to the section 2.4.2 chapter 4.

3.2. Annual N₂O budget and contribution of seasons

Annual budget – Table 4.1 summarises the N₂O annual budgets calculated at both sites along with the annual nitrogen supply, NUE_{agro} , yield scaled-N₂O efflux and $N_{surplus}$. On the whole period of study, summer crops tended to present higher annual N₂O budgets than winter crops with values ranging from 2.1 ± 0.1 (sunflower 2016) to 8.0 ± 0.4 (maize 2014) for summer crops and from 1.0 ± 0.5 (wheat 2012) to 2.8 ± 0.1 (wheat 2016) kgN ha⁻¹ for winter crops. N₂O annual budgets were greater at FR-Lam than at FR-Aur with values ranging from 2.4 ± 0.1 to 8.0 ± 0.40 and from 1.0 ± 0.5 to 2.1 ± 0.1 kgN ha⁻¹ respectively. Moreover, annual N₂O budgets of winter crops were higher at FR-Lam than at FR-Aur, even if nitrogen application were equivalent or higher at FR-Aur.

Seasonal N₂O emissions contribution to annual budget – Winter and spring seasons appeared as the most contributing seasons to the annual N₂O budget for winter and summer crops, respectively (Fig. 4.4). Regarding winter crops, the winter season contribution was on average higher than 40% of the annual N₂O emissions and ranged between 33% (winter wheat 2013 at FR-Aur) and up to 61% (winter wheat 2016 à FR-Lam) of the annual emissions. However, at FR-Aur, rapeseed and winter wheat 2013 presented higher spring N₂O emissions than winter with contributions of 51 and 42 % to the annual N₂O budget. Concerning summer crops, spring season accounted for more than 50% of the annual N₂O budget whatever the cropping year for maize crop at FR-Lam and reached 80 % of annual N₂O emissions for the sunflower crop at FR-Aur. Autumn contributed to approximately 30% of annual N₂O emissions when slurry was spread at the FR-Lam site before a winter wheat crop.

Emission factors variability – Based on the measurement and the resulting annual N₂O budget, the emission factors varied between 0.35 and 3.35 % (for the winter wheat of 2012 and the sunflower of 2016 at FR-Aur respectively) with a mean value for the 10 site-years of 1.12 ± 0.87 % showing a high dispersion (Fig. 4.4) around the classic and well-known coefficient of 1% usually used (IPCC, 2006).

Table 4.1. Annual N₂O budget (\pm standard deviation), NUE_{agro}, Yield scaled-N₂O efflux and nitrogen inputs per cultivated crop and site. Uncertainties correspond to the observations' standard deviations.

Site	Year	Crop	Annual N ₂ O budget (kgN ha ⁻¹)	NUE _{agro} (kgN kgN ⁻¹)	Yield scaled-N ₂ O emissions (gN kgN ⁻¹ uptake)	Nitrogen inputs (kgN)		
						Fertilizer	Crop residues	Total
FR-Lam	2012	Maize	3.8 \pm 0.1	75.3	14.1	255	106	361
	2013	Winter wheat	2.6 \pm 0.1	79.6	11.1	225	72	297
	2014	Maize	8.0 \pm 0.4	41.1	43.7	199	130 (+115 CC)	444
	2015	Maize	2.4 \pm 0.3	99.9	7.8	228	74	302
	2016	Winter wheat	2.8 \pm 0.1	67.7	12.3	220	120	340
FR-Aur	2012	Winter wheat	1.0 \pm 0.5	109.4	3.2	147	149	296
	2013	Rapeseed	2.1 \pm 0.1	107.2	6.2	206	106	312
	2014	Winter wheat	1.3 \pm 0.3	92.7	6.4	109	108	217
	2015	Barley	1.3 \pm 0.1	94.7	9.0	88	67	155
	2016	Sunflower	2.1 \pm 0.1	164.0	20.5	0	62	62

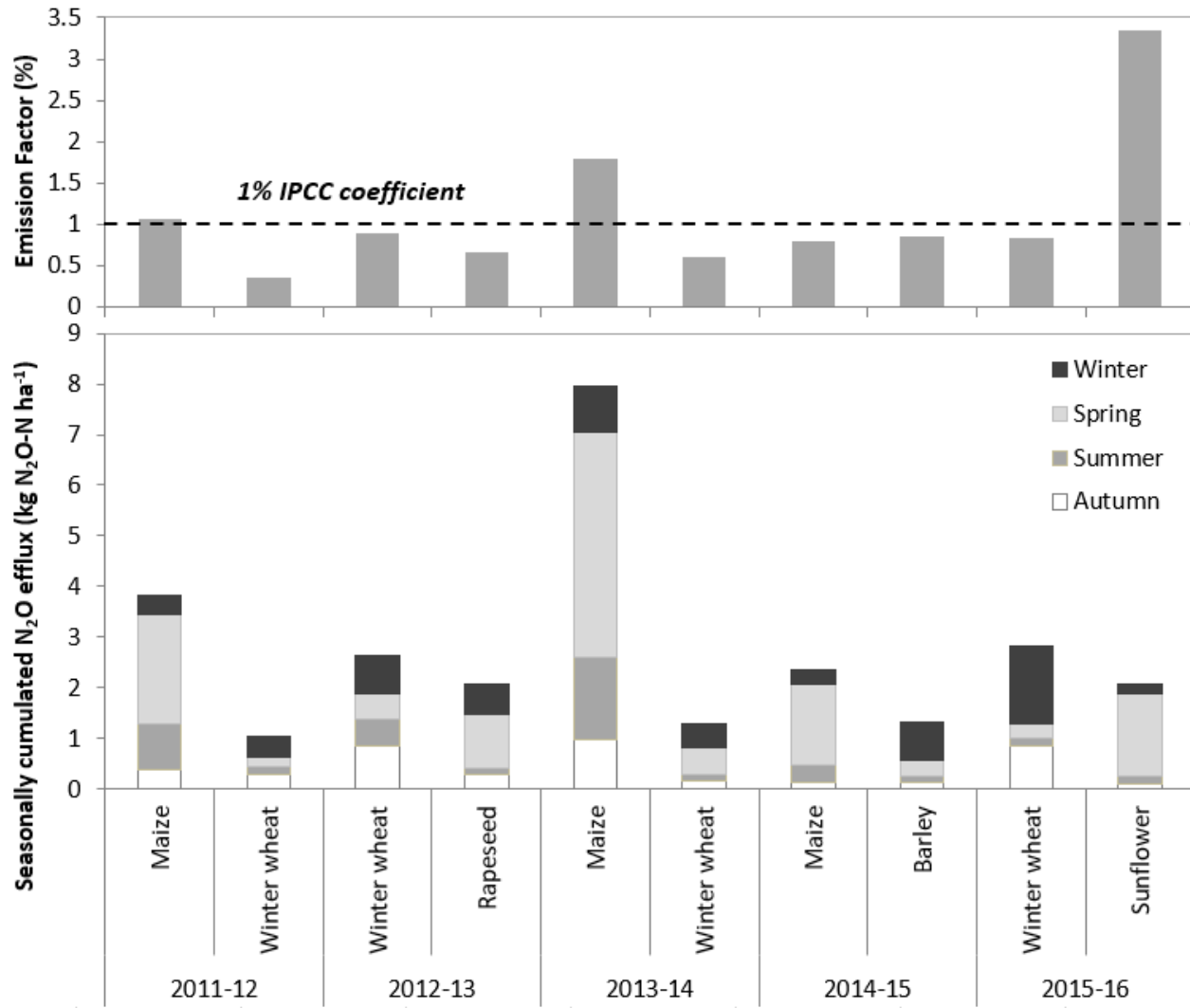


Fig. 4.4. Seasonally cumulated N_2O fluxes (lower panel) and annual emission factor (upper panel) according to year and cultivated species. For each cultivation year, the left side correspond to FR-Lam site and the right side to FR-Aur site.

3.3. Agronomical nitrogen use efficiencies versus yield-scale annual N_2O efflux

The N_2O annual budgets were compared to the N surplus (Fig. 4.5.a). On the 10 available cropping years, annual N_2O budgets increased with N surplus, ranging from 1.04 to 2.65 and from 2.66 to 7.96 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. 4.5.a and Table 4.1). On the contrary, yield-scaled N_2O efflux showed a significant negative relationship with NUE_{agro} . Yield-scaled N_2O efflux decreased from 43.7 to 7.8 and from 9.0 to 3.2 gN kg⁻¹ N uptake when NUE_{agro} increased from 68 to 110% and from 41 to 100% for maize and winter wheat, respectively (Fig. 4.5.b).

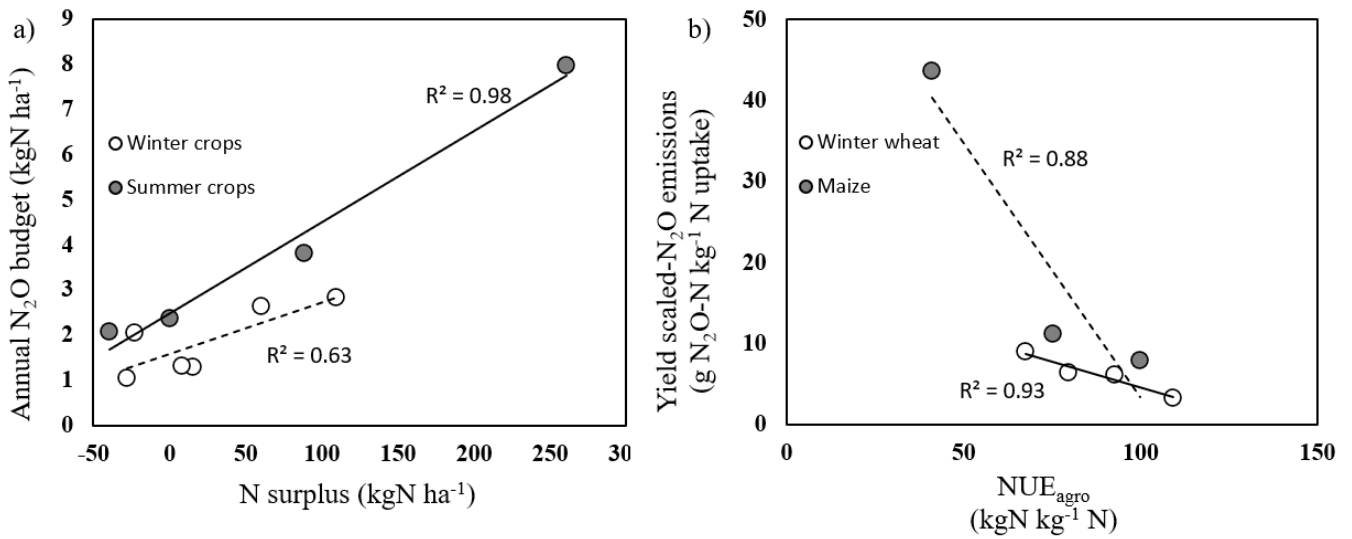


Fig. 4.5. Relationship between N surplus and annual N_2O emissions for summer and winter crops (a) and between agronomical nitrogen use efficiency (NUE, expressed as apparent recovery efficiency (in %) of applied N in kg N taken up per kg N) and (yield-scaled) N_2O emissions for maize and winter wheat crops only (b).

3.4. Management effects on N_2O budget at the FR-Lam and FR-Aur sites

The optimisation procedure described in section 2.4.5 chapter 4 lead to an optimal value of 31 days for the calculation of $Wmean$ and $GLAI_{mean}$ and of 41 for the calculation of the cumulative N_2O emissions F_{N_2O} . These values were fixed to build the relationships found in this study and the empirical model.

3.4.1. Nitrogen fertilisation and crop development

As expected, N fertilisation events modulated the intensity of N_2O emissions in function of the GLAI and the amount of water supply (R+I). The corresponding effect of a fertilisation event (either synthetic or organic) $FN_{day-fert}$ was quantified and calculated following Eq. 4.8. The formalism depends on the vegetation development and water supply:

$$\begin{aligned}
 FN_{day-fert} &= N Wmean_{31d}, \text{ if } GLAI_{mean}_{31d} < 1 \\
 FN_{day-fert} &= \frac{N Wmean_{31d}}{LAI_{31d}}, \text{ if } 1 < GLAI_{mean}_{31d} < 2 \\
 FN_{day-fert} &= \frac{N Wmean_{31d}}{2}, \text{ if } GLAI_{mean}_{31d} > 2
 \end{aligned} \tag{4.8}$$

with N representing the amount of mineral or organic nitrogen applied (in kgN ha⁻¹), $Wmean_{31d}$ and $GLAI_{mean}_{31d}$ being the mean water supply from R+I (in mm d⁻¹) and the

mean GLAI (in $m^2 m^{-2}$) from the day of N application until 30 days after, respectively. The calculated $FN_{day-fert}$ values were applied on a temporal window of 41 days. If different fertilisation events occurred in the same temporal window, the values of the $FN_{day-fert}$ components were summed. In these cases, $FN_{day-fert}$ presented in Fig. 4.6 corresponds to the average of the $FN_{day-fert}$ values over 41 days after a fertilisation event.

The relationship between the cumulated N_2O emissions over 41 days ($F_{N_2O_{41d}}$) and $FN_{day-fert}$ was found significant with a R^2 of 0.91 (Fig. 4.6).

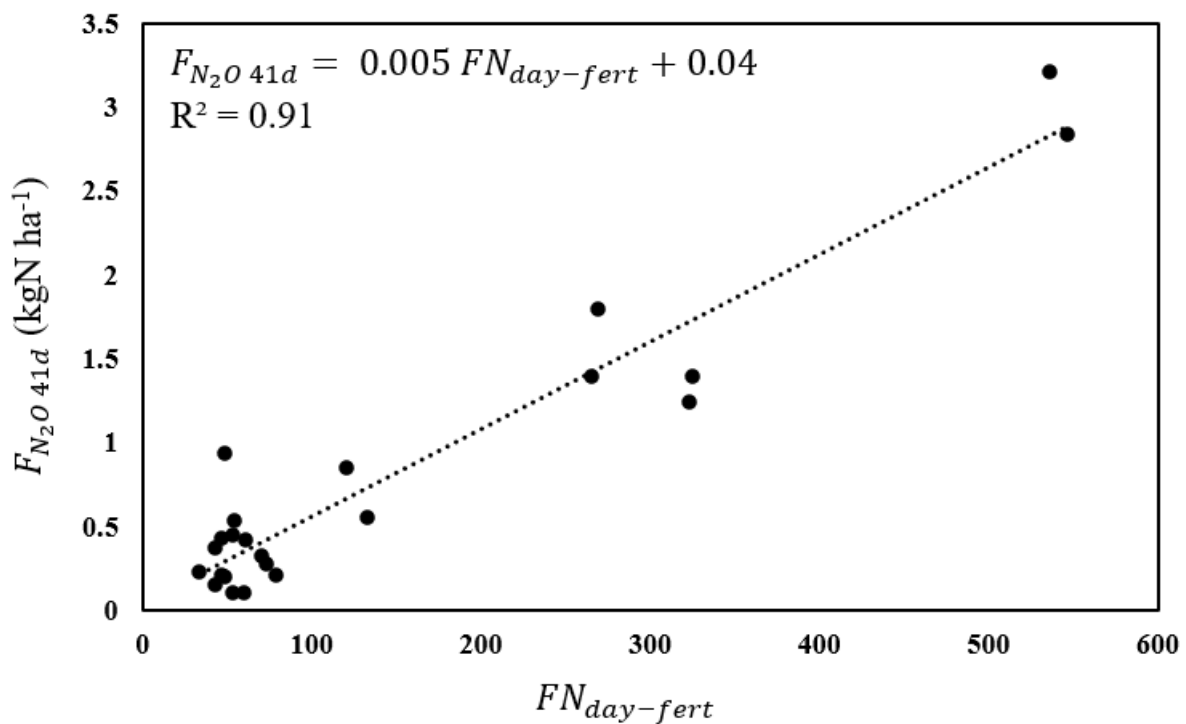


Fig. 4.6. Cumulated N_2O emissions over 41 days ($F_{N_2O_{41d}}$ in $kgN ha^{-1}$) in function of $FN_{day-fert}$ after a fertilisation event. The equation and R^2 are referring to the linear regression line in the figure.

3.4.2. Nitrogen residues and spring mineralisation

At both sites, four spring mineralisation effects on N_2O emissions were observed (3 at FR-Lam, 1 at FR-Aur) with large amount emitted in spring (Fig. 4.3) before a summer crop during periods when no field operations occurred and when vegetation was poorly developed ($GLAI < 0.05$). The assumption was that this event was due to spring mineralisation of the previous crop and/or cover crop residues and was dependent on the residues' nitrogen content (N_{res} , in $kgN ha^{-1}$) and rainfall amount. As organic matter mineralisation is highly influenced by temperature and degree days accumulation (Van Schöll et al., 1997; Delin and Engström,

2009), a test was carried out to estimate best the date or degree-day sum from which N_{res} should be taken into account.

Degree-day accumulation was defined as the cumulative value of $\frac{T_{max} + T_{min}}{2}$ from the 1st of January to the day when the spring mineralised N should be applied, with T_{max} and T_{min} as the maximum and minimum air temperature of each day (in °C), respectively. R^2 between $N_{res} Wmean_{31d}$ and $F_{N_2O\ 41d}$ was calculated at each date corresponding to the degree-day sum from 600 to 1100 °C by considering an increment of 10 °C. The optimum degree-day sum to estimate the effect of N_{res} mineralisation on N_2O emissions was found to be of 1040 °C with a R^2 of 0.85 (Fig. 4.7).

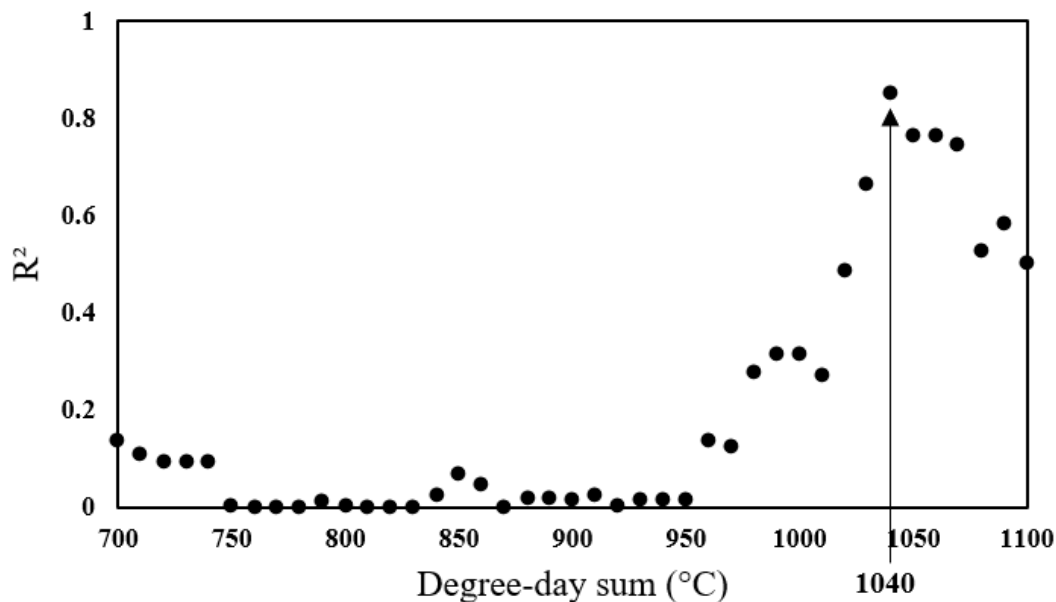


Fig. 4.7. R^2 calculated between $N_{res} Wmean_{31d}$ and $F_{N_2O\ 41d}$ in function of the degree-day sum from which applying N_{res} for the year 2012, 2014 and 2015 in FR-Lam and for the year 2016 in FR-Aur. For a degree-day sum of 1040 °C, R^2 is of 0.85.

The correlation between $N_{res} Wmean_{31d}$ calculated after a degree-day sum of 1040°C is reached and $F_{N_2O\ 41d}$ was analysed (Fig. 4.8). A linear relationship was found with a R^2 of 0.85. It should be note that N_{res} , experimentally measured, does not represent the real level of mineral nitrogen released from the residues' incorporation at the considered degree-day sum, but it allowed establishing a relationship between N_{res} and $F_{N_2O\ 41d}$.

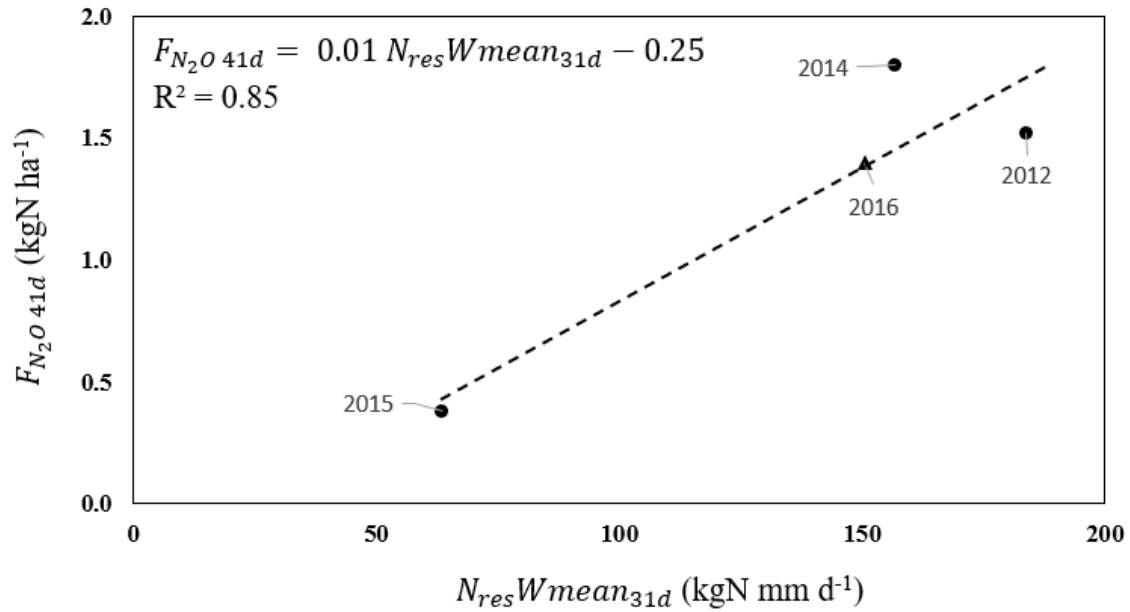


Fig. 4.8. Relation between the residual nitrogen content from the previous crop residues (N_{res} in $kgN\ ha^{-1}$) weighed by the mean waterfall over 31 days after the date determined from the 1040 degree-day accumulation starting on January the 1st ($W_{mean_{31d}}$ in $mm\ d^{-1}$) and the cumulated N_2O emissions over 41 days after that date ($F_{N_2O\ 41d}$ in $kgN\ ha^{-1}$) during a maize (FR-Lam, ●) or a sunflower (FR-Aur, ▲) cropping year. The equation and R^2 are referring to the linear regression line in the figure.

Table 4.2 summarizes the variables used to establish the relationship shown in Fig. 4.8. N_{res} values ranged from 62 to 245 $kgN\ ha^{-1}$ at FR-Aur in 2016 and at FR-Lam in 2014, respectively, when $F_{N_2O\ 41d}$ ranged from 0.38 to 1.80 $kgN\ ha^{-1}$ at FR-Lam in 2015 and at FR-Lam in 2012, respectively. The non-linearity between N_{res} and $F_{N_2O\ 41d}$ illustrates the importance of taking into account $W_{mean_{31d}}$ that modulates the impact of N_{res} on $F_{N_2O\ 41d}$ (Fig. 4.8).

Table 4.2. Values of N_{res} , $W_{mean_{31d}}$ and $F_{N_2O\ 41d}$ during the observed spring mineralisation effect on FR-Lam and FR-Aur from the a cumulative degree-day calculation of 1040°C and the associated date. Residues + CC means that the Cover Crop nitrogen content was also included in the total nitrogen residues.

Location	FR-Lam	FR-Lam	FR-Lam	FR-Aur
Date	11/05/2012	23/04/2014	03/05/2015	27/04/2016
N_{res} ($kgN\ ha^{-1}$)	106	245 (residues + CC)	74	62
$W_{mean_{31d}}$ ($mm\ d^{-1}$)	1.48	0.75	0.86	2.43
$N_{res}\ W_{mean_{31d}}$	157	184	63	150
$F_{N_2O\ 41d}$ ($kgN\ ha^{-1}$)	1.80	1.52	0.38	1.40

3.4.3. Tillage depth

Tillage proved to have an impact on N_2O emissions depending on the depth and the mean water supply over 31 days before the operation (Fig. 4.9). Data from both sites were put together to investigate the impact of tillage on N_2O emissions and also split according to the tillage depth (more than 20 cm depth versus less than 20 cm depth). The relationship between the cumulated N_2O emissions over 41 days ($F_{N_2O_{41d}}$) following a deep tillage with the mean water supply from R+I over 31 days before such a field operation ($Wmean_{31d-before}$, in $mm\ d^{-1}$) was found significant and exponential with a R^2 of 0.99 (Fig. 4.9). On the opposite, no significant relationship was found when reduced tillage was done (Fig. 4.9).

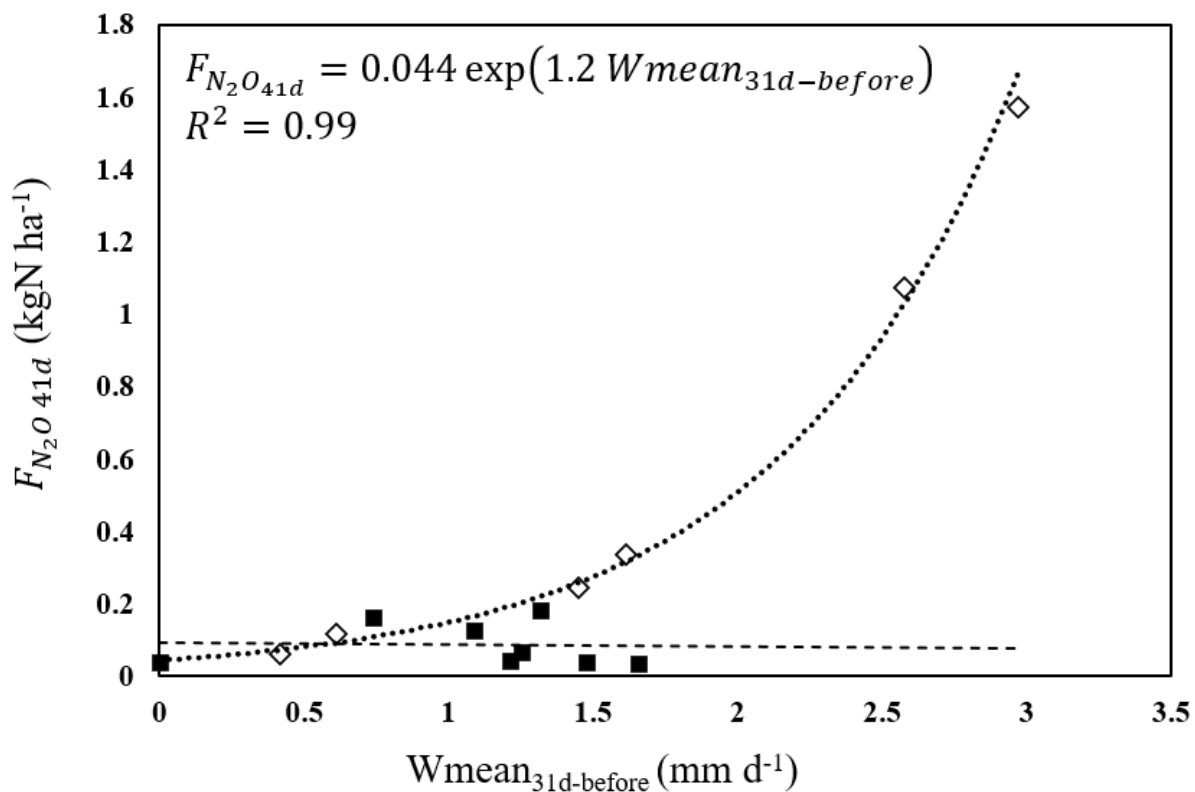


Fig. 4.9. Effect of mean rain over 31 days before a deep tillage ($Wmean_{31d-before}$ in $mm\ d^{-1}$) higher than 20 cm (■) or a reduced tillage lower than 20 cm (◇) on cumulated N_2O budget over 41 days after this tillage ($F_{N_2O_{41d}}$ in $kgN\ ha^{-1}$). The equation and R^2 are referring to the exponential regression line in the figure.

3.4.4. Empirical modelling of seasonal N_2O budget

Based on the relationship found between nitrogen fertilisation combined vegetation index and water supply with observed N_2O emissions, an empirical equation was developed to

reproduce observed seasonal N₂O budget (kgN ha⁻¹) by adding nitrogen residues and deep tillage effect following equation 9 to 12:

$$F_{N_2O \text{ season-sim}} = 0.0137 F_{N_{avg}} + 0.22 \quad (4.9)$$

$$F_{N_{avg}} = \frac{\sum_1^n (F_{N_{day-fert}} + F_{N_{day-res}} + F_{N_{day-till}})}{n} \quad (4.10)$$

where $F_{N_2O \text{ season-sim}}$ is the simulated seasonal N₂O budget, $F_{N_{avg}}$ is the average of the daily sum of the three $F_{N_{day}}$ components (defined and described hereafter) over a given season (*i.e.* three months). $F_{N_{day}}$ is a response function integrating changes in N supply, GLAI, tillage and cumulative amount of water supply from rain and irrigation (R+I). Then the formalism of the $F_{N_{day}}$ term varies according to field operation type (tillage, fertilisation or irrigation), rain and crop development (Eq. 4.8, 4.11 and 4.12). The effects of the three $F_{N_{day}}$ components were considered independently for the calculation of $F_{N_{day}}$. The calculated $F_{N_{day}}$ values were applied on a temporal window of 41 days. If different events occurred in the same temporal window, the values of the $F_{N_{day}}$ components ($F_{N_{day-fert}}$, $F_{N_{day-res}}$ and $F_{N_{day-till}}$) were summed.

$F_{N_{day-fert}}$ formalism depends on N fertilisation application rate, vegetation development and water supply and is calculated following Eq. 4.8.

In order to homogenise the effect of nitrogen residues and deep tillage on N₂O emissions according to the effect of nitrogen fertilisation, the equations showed in Fig. 4.8 and Fig. 4.9 were scaled to be equivalent to a nitrogen fertilisation effect.

Therefore, $F_{N_{day-res}}$ is calculated as follow before a summer crop when the degree-day accumulation reaches 1040 °C starting from the 1 January:

$$\begin{aligned} F_{N_{day-res}} &= 1.45 N_{res} W_{mean_{31d}} - 50, \text{ if } 1.45 N_{res} W_{mean_{31d}} > 50 \\ F_{N_{day-res}} &= 0, \text{ if } 1.45 N_{res} W_{mean_{31d}} \leq 50 \end{aligned} \quad (4.11)$$

The threshold of 50 here is used to avoid negative value of $F_{N_{day-res}}$.

In the case of a deep tillage, $F_{N_{day-till}}$ is calculated as follow:

$$F_{N_{day-till}} = 2.23 \exp[1.55 W_{mean_{31d-before}}] \quad (4.12)$$

Correlation between FN_{avg} and N_2O emissions were carried out at the seasonal scale.

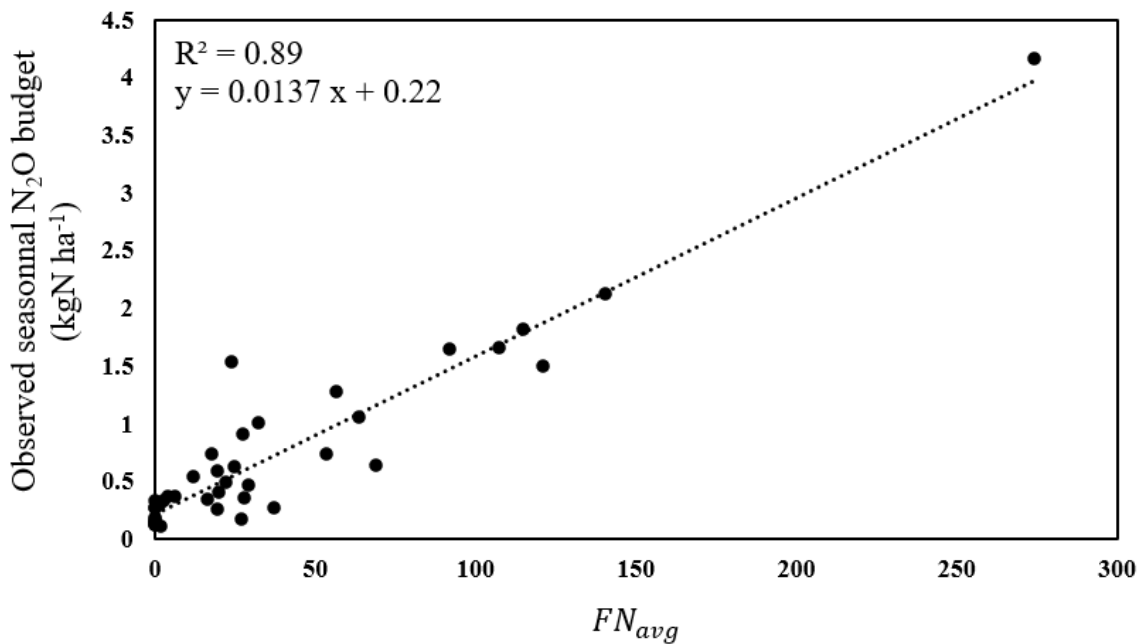


Fig. 4.10. Empirical linear relationship between the observed seasonal N_2O budget ($F_{N_2O \text{ season sim}}$ in $kgN \text{ ha}^{-1}$) and FN_{avg} (FN parameter averaged at the seasonal scale).

The linear regression and high associated R^2 score presented in Fig. 4.10 proved to well link the seasonal N_2O emissions with vegetation development, agricultural management and rainfall variability. These results lead to the empirical equation (Eq. 4.9) that simulates N_2O budgets at the seasonal scale.

3.5. N_2O budget simulation

3.5.1. Seasonal scale

The empirical equation (Eq. 4.9) presented in section 3.4.4 chapter 4 and developed using data from both sites proved to simulate well the observed N_2O seasonal budget whatever the magnitude of the monthly cumulated N_2O fluxes with R^2 scores of 0.87 and 0.92 and RMSE of 0.33 and 0.12 $kgN \text{ ha}^{-1}$ at FR-Lam and FR-Aur, respectively (Fig. 4.11), even if the sites have contrasted management and N_2O budgets ranges. Based on observations, N_2O seasonal budgets varied from 0.17 ± 0.06 to 4.17 ± 0.36 and from 0.11 ± 0.01 to $1.65 \pm 0.10 \text{ kgN ha}^{-1}$ at FR-Lam and FR-Aur, respectively. Based on simulations, they varied from 0.22 to 3.98 and from 0.22 to 1.48 $kgN \text{ ha}^{-1}$ at FR-Lam and FR-Aur, respectively. The lowest and highest simulated N_2O seasonal budgets are of the same order of magnitude when compared to the observations (Fig. 4.11).

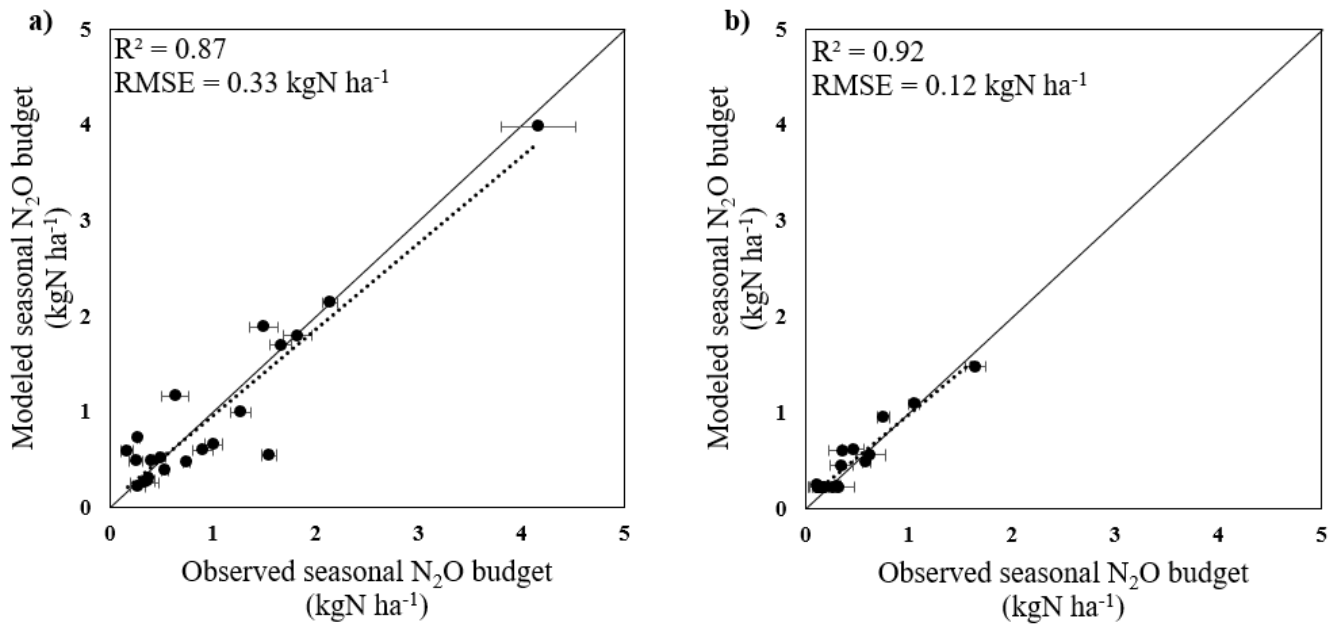


Fig. 4.11. Comparison between observed and modelled seasonal N_2O budget ($kgN\ ha^{-1}$) on FR-Lam (a) and FR-Aur (b). Linear regressions are represented by the dotted lines. Bars indicate uncertainties of the seasonal N_2O budgets calculated according to the section 2.4.3 chapter 4.

3.5.2. Annual scale: comparison with IPCC methodology

As no data were available from October to December 2011 at FR-Aur, the cropping year 2011 – 2012 at the FR-Aur site was removed from the analysis.

As expected from the previous results at the seasonal scale, estimating annual N_2O budget defined from October 1st to September 30th with the empirical model proved to be relevant. When binding data from both sites together, the empirical model reached a high R^2 score of 0.96 and a low RMSE of $0.43\ kgN\ ha^{-1}$ (Fig. 4.12. a) and estimated well the range of annual N_2O budgets. The proposed empirical model gave better results than the IPCC Tiers 1 methodology approach that predicted more scattered results with a low R^2 of 0.37 and larger RMSE of $1.56\ kgN\ ha^{-1}$ (Fig. 4.12. b).

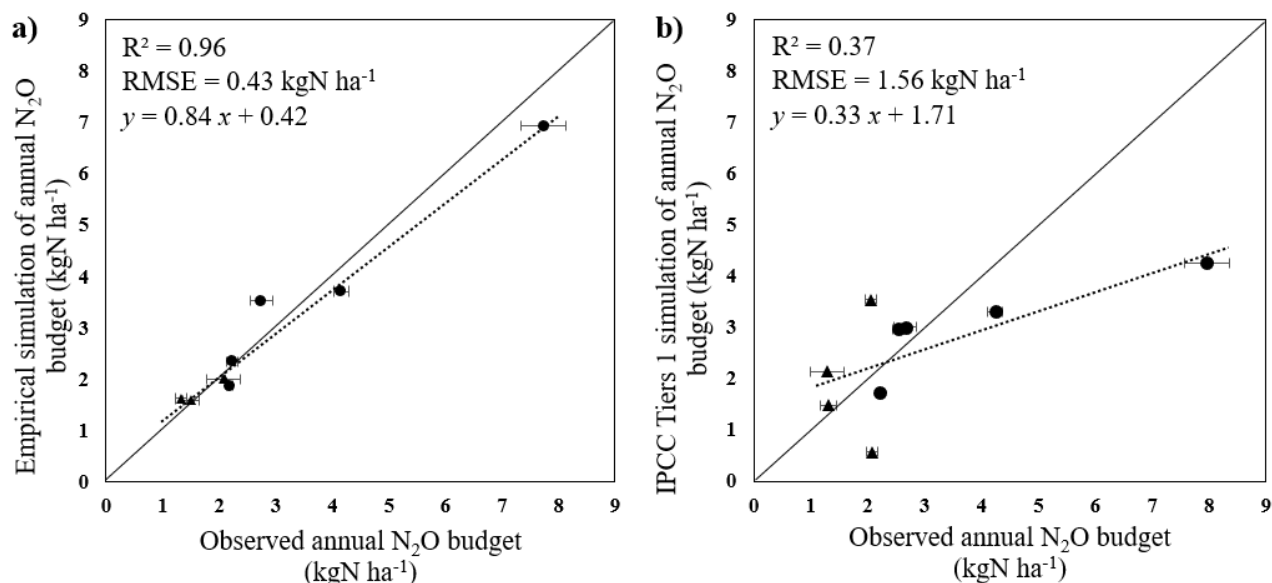


Fig. 4.12. Observed annual N_2O budgets at FR-Lam (●) and FR-Aur (▲) as a function of the empirical simulations of the annual N_2O budget (a) and of the IPCC Tiers 1 simulation (b). y represents the linear regression equations that are represented by the dotted lines. Bars indicate uncertainties of the annual N_2O budgets calculated according to the section 2.4.3 chapter 4.

4. Discussion

4.1. Effect of crop rotation on N_2O fluxes

Over the 5 cropping year's rotations, $19.6\ kg\ N_2O-N\ ha^{-1}$ were emitted at the FR-Lam site whereas only $7.8\ kg\ N_2O-N\ ha^{-1}$ were emitted at the FR-Aur site. The higher 5-years N_2O budget observed at FR-Lam compared to FR-Aur (Table 4.1) may partly be explained by crop rotations and differences in agricultural practice. Maize, that was grown over three cropping years at FR-Lam, proved to be a high emitting crop compared to the others. Indeed, due to its specific rapid growth which obstructs most technical operations because of important vegetation height, farmers usually apply the N plant requirements once or twice over a short time period at the very beginning of its growing season when the plants N demand is very low compared to its phenological stage. This, combined with irrigation and/or high rainfall events produced optimal conditions for denitrification to happen and therefore important N_2O emissions. For example, the large emissions that were observed in June 2012, 2014 and 2015 at the FR-Lam site followed nitrogen fertilisation of 110, 174 and 210 kgN , respectively. They also correspond to low vegetation development in May with LAI of 0.2, 0.02 and 0.06 $m^2\ m^{-2}$, medium LAI development in June with values of 1.3, 0.3 and 2.1 $m^2\ m^{-2}$ in 2012, 2014 and

2015 and multiple irrigation events. Moreover, at FR-Lam, before the sowing of maize, spring mineralisation occurred during long bare soil periods, increasing soil mineral N availability for other processes than crop production. Wagner-Riddle et al. (1997) also showed that spring season was the main contributor of annual N₂O emissions from barley, soybean, canola and corn crops sowed in May due to spring thawing with monthly emissions that can reach 3 kgN ha⁻¹.

Winter crop periods also proved to be more emissive at FR-Lam compared to FR-Aur (Fig. 4.4). These differences between both sites can be explained by the fact that in autumn before winter crop were sown, no deep tillage occurred at the FR-Aur site in contrast to the FR-Lam site. Mutegi et al. (2010) had the same results from a Danish crop site during a winter barley cultivation with higher N₂O emissions after conventional tillage in comparison to reduce and no till in autumn. Also, high amount of N fertilisation was applied at the FR-Lam site in September 2013 (110 kgN ha⁻¹) and 2016 (120 kgN ha⁻¹) before winter wheat crop was sown causing large N₂O emissions (Fig. 4.3). On the opposite, at the FR-Aur site, N fertilisation was split and applied throughout the vegetation period with lower amounts (ranging from 40 to 82 kgN) compared to FR-Lam. Also, vegetation was already well developed at FR-Aur during N applications with LAI ranging from 0.52 to 6.2 m² m⁻².

Except for the 2013 – 2014 maize crop at FR-Lam, values of N₂O annual budgets at our sites were in the range of those obtained by Loubet et al. (2011) in their long-term experiment in northern France, where values of N₂O budgets were of 0.8, 1.5 and 3.0 kgN ha⁻¹ yr⁻¹ during winter wheat, barley and maize cropping, respectively. Their winter crops budgets were close to those obtained at FR-Aur and their maize crop budget is similar to our 2014 – 2015 maize budget. These results tend to show that maize cropping years have a higher risk of large N₂O budgets compared to winter crops. Also, this comparison tends to show that the N₂O emissions that were observed for the 2013 – 2014 maize crop at FR-Lam were exceptional.

Two kind of key drivers modulating seasonal N₂O emissions were identified at different time scales in this study: medium term key drivers (N crop residues) and short-term key drivers (tillage, fertilisation, presence of vegetation, rain and irrigation). Among all the key drivers identified in this study, rain and irrigation, which control soil water content and soil O₂ concentration, proved to be the most important, the other ones having an effect on N₂O fluxes only when combined with these two ones. These variables were also recognised as the main key driver in a managed irrigated and fertilised eucalyptus forest in Australia (Martins et al., 2015). Skiba and Smith (2000) also showed that the IPCC methodology uncertainties in

estimating N₂O budgets can only be alleviated by including rainfall and water filled pore space into the budget equation.

4.1.1. Residual nitrogen and spring mineralisation impact on spring N₂O efflux: effect of crop type

This study highlighted and quantified for the first time the impact of previous crop or winter cover crop residues and related spring mineralised N on N₂O efflux (Fig. 4.8). Based on the 10 crop-years measurements, it clearly appeared that large emissions occurred before a summer crop (maize or sunflower) during the spring season when the soil was bare or when the vegetation was little developed in April/May. Spring was the period which contributed the most significantly to the annual N₂O budgets for summer cropping year (from 50 % to 80 %). The magnitude of the N₂O emissions was highly correlated with the amount of residual nitrogen released from the previous crop combined with the amount of precipitations during the month following the date when a cumulative temperature of 1040°C is reached from the 1 January (Fig. 4.8). N₂O emissions were really low in Spring 2015 at FR-Lam compared to 2012 and 2014 in FR-Lam and compared to 2016 at FR-Aur while N residues were similar to those from the FR-Aur site in 2016 (Table 4.2). The main difference between those two cropping years was the rainfall: indeed, over the considered periods, the rainfall was of 102 mm at FR-Aur in 2016 when it was only of 12 mm at FR-Lam in 2015.

The absence of such significant spring emissions when winter crop was cultivated suggests that the presence of well-developed vegetation helped to reduce the N losses to the environment, as nitrate leaching but also as N₂O production and emission. Plants' roots (rapeseed, wheat or barley) were able to valorise the mineral nitrogen compounds by extracting soil mineral nitrogen progressively released over the spring mineralisation period (Mackay and Barber, 1986), thus preventing their access to bacteria that can produce N₂O. If nitrogen supply amount and crop requirements are not well synchronized over the spring period, NUE would be low and enhance losses of soil N surplus to the environment (Van Eerd et al., 2018) partly as potential important N₂O emissions if the amount of rain and/or irrigation allows it. The negative correlation observed for maize between total produced biomass and annual N₂O budget per unit of N input (fertilizer + residues) on FR-Lam site typically corroborates that finding. In our case, an increasing soil N availability did not boost the maize biomass production but favoured N₂O production. This phenomenon was also particularly observed during the sunflower cropping year. Despite the fact that sunflower did not receive nitrogen fertilisation

and that the amount of the previous crop residues' nitrogen was the lowest, important N₂O fluxes were recorded during May 2016 spring period before which the amount of precipitation was also one of the highest recorded over the study period. Those results highlight the importance and the need to take into account the water effect during the spring season on potential N₂O fluxes and may reconsider the classic existing inventory approach. In that case, the IPCC Tiers 1 methodology based on the total nitrogen input only strongly underestimated the annual N₂O budget of summer cropping year as it does not take into account the meteorological variabilities that influence the N cycling and related mineralisation. Beside that inventory issue, our results highlighted the need to manage crop establishment more adequately with the soil N offer to improve NUE and crop yield and reduce N losses via N₂O production as shown in our study, especially for summer crop. Moreover, the intensity with which previous residual nitrogen is mineralized during the next spring cropping year would vary according to the residual C input amount. Mary et al. (2020) found a high correlation between the mineralization rate of organic matter and the amount of residual C inputs whatever the crop. Based on that recent finding we looked at the potential effect of the amount of C input (kgC ha⁻¹) from previous and/or intercrop cover residues on the following spring N₂O fluxes preceding a summer crop. We found a positive correlation (on only four points, data not shown) between the amount of residual carbon and spring N₂O fluxes. That positive correlation was also found with an increasing amount of residual nitrogen (Fig. 4.8). *In situ*, denitrification rates depend greatly on the availability of labile carbon compounds in soil and plant residues represent the main inputs of carbon contents to agricultural soils (Senbayram, et al. 2012). Knowing the quantity of C incorporated into the soil after harvest is a key driver of mineralisation (Mary et al. 2020) and would help to predict the potential mineralisation rate and anticipate the mineral nitrogen availability for the following spring season and adapt the agricultural practices accordingly. Spring mineralisation effect indirectly witnessed via high spring N₂O budget on both sites in May before a summer crop was in accordance with the results of Honeycutt and Potaro (1990) who found that plant nitrogen residues from a previous crop harvested in July were mineralised around May of the next cropping year in the New England region (USA).

4.1.2. Autumn N₂O flux intensity explained by the tillage depth combined with the previous mean water supply

Tillage deeper than 20 cm depth combined with rainy and wet conditions before the operation proved to impact N₂O emissions ($R^2 = 0.99$), which intensity depended on the

previous mean water from R+I. Deep tillage was mainly operated during the second part of autumn, preceding a summer cropping year. Such operations are done to prepare the soil and benefit from mechanical action of winter frost on clay soil. These results are in accordance with those from Tian et al. (2012) and Chatskikh et al. (2008) who showed an increase in N₂O fluxes after a deep tillage of 40 and 20 cm after, respectively for each study, when compared to no-till after a rain of 70 and 40 mm during the month preceding the tillage, respectively. A first underlying hypothesis to explain the following N₂O emission is that N₂O production occurred deeper into the soil than 20 cm on our sites, where the soil layer is more compacted and, combined with the duration of the supplying water from R+I, favoured the creation of anaerobic conditions for denitrification process. The deep tillage operation provoked a destruction of the soil layer that can favour the diffusion of N₂O from deep layers to the surface. Moreover, the absence of increased N₂O fluxes observed mainly at FR-Aur after a superficial tillage could be attributed to an increased physical protection of the deeper layer and which could result *in fine* to a lower organic matter decomposition (Balesdent et al., 2000), or to smaller C inputs (and available labile carbon compounds) due to the absence of mechanical incorporation of crop residues (Mary et al., 2020; Senbayram et al., 2012) over years in the deeper layer where denitrification activity and subsequent N₂O production mostly occur. This hypothesis is consistent with the study of Müller et al. (2004) who showed that N₂O production in a temperate grassland occurred between 20 to 50 cm depth. However, a second underlying hypothesis to explain increased N₂O emissions after deep tillage is that the resulted soil disturbance increased soil aeration and decomposition rate that led to more substrates (dissolved organic carbon, ammonium, nitrate, ...) released into the soil that stimulate nitrification and denitrification (Li et al. 2010).

4.1.3. Irrigation effect

Timing of irrigation combined together with high nitrogen availability (from residues and/or fertilizer) clearly enhanced high N₂O emissions during spring and summer time, especially from June to the end of July and to a lesser extent in August. In southwestern France, the climate is submitted to Mediterranean influence. Thus, precipitation during summer period, when the highest temperatures occur, is scarce, and most classic maize crops management, e.g. sowing in the middle of spring, requires irrigation to achieve worthwhile yields. Several studies reported irrigation effect on N₂O emissions in semi-arid/arid region (Aguilera et al., 2013, Sanz-Cobena et al., 2017, Yang et al., 2019b) and pointed out that irrigation optimisation represent an important tool to mitigate N₂O fluxes from irrigated fields in these regions from 40 to 70%.

Aguilera et al. (2013) on their meta-analysis of studies from multiple countries with a Mediterranean climate showed N₂O fluxes variation from 1.2 ± 1.0 (low water irrigation) to 4.0 ± 2.6 (high water irrigation) kgN ha⁻¹ yr⁻¹ from these studies. Yang et al. (2019b) found the same range in their results from a potato field in Inner Mongolia, China with fluxes from 1.5 ± 0.2 (sprinkler irrigation) to 4.3 ± 0.3 (flooded irrigation) kgN ha⁻¹ yr⁻¹.

As expected, high soil N level combined with high water level due to irrigation enhanced N₂O emissions from 0.1 ± 0.1 to 2.9 ± 0.3 kgN ha⁻¹ month⁻¹ for August 2015 and June 2014 maize cropping year respectively on FR-Lam site. Moreover, Bergstermann et al. (2011) in a laboratory experiment showed that antecedent soil moisture before an irrigation operation influenced N₂O efflux from an arable soil. Rewetting dry soil increased the emissions of N₂O as compared to wetting a moist soil which may be due to several (interacting) processes : an increase in C and N substrate availability (Ruser et al. 2006; Bergstermann et al., 2011) with associated respiratory O₂ consumption, microbial stress (Kieft et al., 1987; Fierer and Schimel, 2003), soil organic matter exposure by physical disruption of aggregates (Goebel et al., 2005) and a reduction of diffusional constraints (Schjønning et al., 2003).

4.2. Fertilisation effect on N₂O emission depends on N-use efficiency: NUE_{agro} versus yield-scaled N₂O efflux

Studies on GHG emissions and agronomical N use efficiencies should not be disconnected as both are closely related to fertilization and soil management. Some efforts to link both points of view and find consensus to reach an agronomical and an environmental compromise should be made. Van Groenigen et al. (2010) introduced the concept of linking agronomic productivity to environmental sustainability in looking and focusing on the relationship between N₂O efflux and crop productivity like the amount of N₂O-N emitted per kg of aboveground N-crop absorbed, denoted as 'yield-scaled' N₂O efflux. Consistently with the previous meta-analysis of Van Groenigen et al. (2010), a positive relation between N surplus and annual N₂O budgets and negative relation between N use efficiency and yield-scaled N₂O efflux were found on both sites of our study. An increase in N₂O efflux when N applied or available exceeds N crop demand is a common response that has been observed in a few field studies (Bouwman et al., 2002; Grant et al., 2006; Van Groenigen et al. 2010). It is interesting to note here that within the range of a N surplus of approximately 90 kg ha⁻¹ to a deficit N surplus of approximately -8 kg ha⁻¹, annual N₂O efflux ha⁻¹ were lower for winter crops than for summer crops. This underlines the capacity for winter crops to take up the progressively

mineralized nitrogen and valorise with more efficiency than summer crops the moderate rates of applied N during their growing season. The regression line in Fig. 4.5.b, showing a linear negative relation between NUE and yield-scale N₂O efflux, is based on only three points for maize and winter wheat. The strong fit of the regression equations should not therefore be over-emphasized. However, this trend is an indication that agronomic aims of increasing fertilizer N use efficiency may cohabit with GHG efficiency by reducing N₂O fluxes. The N loss partly as N₂O efflux in the detriment of agronomical performances was especially important for maize 2014 and winter wheat 2016 at the FR-Lam site. For winter wheat 2016, although well developed ($LAI > 2 \text{ m}^2/\text{m}^2$) when N fertilisation was applied and given the fact that the most contributed period to the annual N₂O budget for that crop species was winter time, those results suggest that either the offer exceeded the demand, or the process of plant N uptake was in competition with N₂O production processes with the soil moisture conditions being in favour of N₂O fluxes during the following month of this technical operation. In conclusion, a portion of available mineral nitrogen has been transformed into N₂O and not valorised by the plant.

Considering maize cropping years, and given the high contribution of the spring period to the annual N₂O budget, the stronger negative relation between both N use efficiency and yield-scaled N₂O efflux supported the need to improve and to optimise the synchronisation of summer crop seeding and development with spring soil N offer. Maize 2014 was a special high emitting cropping year, mostly because of very important monthly N₂O efflux during spring and summer compared to other years. This exceptional emission could result from very concomitant factors favourable to N₂O production processes: important spring mineralized nitrogen higher than other years (due to previous winter cover crop incorporation), application of a high amount of nitrogen fertilisation at a very early stage of development ($LAI < 0.5 \text{ m}^2/\text{m}^2$) and a high water amount from rain and irrigation. Timing of summer crop sowing together with adequate N application, at a LAI stage higher than $1 \text{ m}^2 \text{ m}^{-2}$ for example, may help to increase mineral-N (from fertilizer and spring mineralisation) use efficiency and by the way reduce N₂O efflux and any other way of N losses (Van Groenigen et al., 2010; Quemada et al. 2013). In our study based on the only 6 available cropping years, we were not able to quantify at which levels of N application the yield scaled N₂O efflux would be the smallest either for maize or winter wheat as Van Groenigen et al. (2010) did.

Finally, in opposition with the IPCC Tiers 1 approach, an increase of annual N_{input} does not automatically lead to an increase of N₂O annual budget (Table 4.1). Thus, on annual scale, considering N-surplus instead of total N_{input} sounds more precise to improve inventory

methodology. N-surplus is a variable which integrates effect of total N_{input} together with agricultural practices and meteorological variability which both influence the crop N use efficiency.

4.3. Towards an improved N₂O budget estimation

The new methodology proposed in this study to estimate N₂O budget proved to be really effective on our sites with very good statistical scores at both seasonal and annual scale. It also showed an important improvement when compared to the IPCC Tiers 1 methodology with more precise estimation (Fig. 4.12). Even if the Tiers 1 has the benefit to be easy to use, it has a critical lack of accuracy. Our methodology is a compromise between both as it needs really few input variables (LAI, maximum and minimum daily temperature, daily water from R+I and technical operations) that can be easily obtained when compared to other mechanistic models and has the advantage to be quite accurate. The chosen simulation's input variables made possible the generalisation of this empirical model. However, this methodology based on an empirical approach specifically developed for southwestern France conditions needs to be tested on other locations under contrasted soil characteristics, climate and agricultural practices conditions. For example, thawing has been reported to have an important impact on N₂O emissions from temperate agro-ecosystem (Müller et al., 2002; Wang et al., 2016). Snow cover or soil freezing is really scarce in Southwestern France; thus, this potential effect has not been quantified in our study quantifying this effect in the model can help to improve it.

Furthermore, to approach territorial scale inventories, this methodology offers new perspectives by taking advantage of high spatial and temporal remote sensing products (large scale land use and LAI maps) together with regional statistical management data and SAFRAN meteorological data to enrich, make easier and re-fine existing national methodology inventories (Fieuzal et al. Submitted, IPCC 2006) of croplands specifically.

Conclusion

This study, based on a long and continuous time series of N₂O fluxes (10 cropping years), identified different variables affecting N₂O efflux and quantified for the first time their effect more precisely. From the seasonal scale analysis, reduced tillage and crop residues management combined with water from rain and irrigation revealed a large potential for reducing net GHG efflux.

Moreover, a model using these different variables was developed to quantify N₂O seasonal and annual budget at the plot scale. Based on obtained results, this study leads to the conclusion that on our sites (i) water from R+I was the most important variable influencing N₂O fluxes ; (ii) when combined with water from R+I, nitrogen residues mineralisation during a summer crop cultivation year and deep tillage mostly during autumn have an important impact on N₂O fluxes ; (iii) and the model we developed which takes into account water from RI, fertilisation, crop development, spring mineralisation and deep tillage gave far better results than the IPCC Tiers 1 methodology. We think that this empirical model may not be site specific and could be used on other crop fields to estimate N₂O seasonal or annual budget. Testing this approach on different sites will help to validate it.

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