Review of causes and sources for N₂O emissions and NO₃-leaching from organic arable crop rotations

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Abstract.

The emissions of nitrous oxide (N_2O) and leaching of nitrate (NO_3) from agricultural cropping systems have considerable negative impacts on climate and the environment. Although these environmental burdens are on average less per unit area in organic than in non-organic production, they are roughly similar per unit of product. If organic farming is to maintain its goal of being environmentally friendly, these loadings must be mitigated. We discuss the impact of possible drivers of N_2O emissions and NO_3 leaching within organic arable farming practice under European climatic conditions, and potential strategies to reduce these. Organic arable crop rotations are generally diverse with frequent use of legumes, intercropping and organic fertilizers. The soil organic matter content and share of active organic matter, soil structure, microbial and faunal activity are higher in such diverse rotations, and yields lower, than in non-organic arable cropping systems based on less diverse systems and inorganic fertilizers. Soil mineral nitrogen (SMN), N_2O emissions and NO_3 leaching are low under growing crops, but there is the potential for SMN accumulation and losses after crop termination, harvest or senescence. The risk of high N_2O fluxes increases when large amounts of herbage or organic fertilizers with readily available nitrogen (N) and degradable carbon are incorporated into the soil or left on the surface. Freezing / thawing, drying / rewetting, compacted and/or wet soil and mechanical mixing of crop residues into the soil further enhance the risk of high N_2O fluxes. N derived from soil organic matter (background emissions) do, however, seem to be the most important driver for N_2O emission from organic arable crop rotations and the correlation between yearly total N-input and N_2O emissions is weak. Incorporation of N rich plant residues or mechanical weeding followed by bare fallow increases the risk of NO_3 leaching. In contrast, strategic use of deep-rooted crops with long growing seasons or effective cover crops in the rotation reduces NO_3 leaching risk. Enhanced recycling of herbage from green manures, crop residues and cover crops through biogas or composting may increase N efficiency and reduce N_2O emissions and NO_3 leaching. Mixtures of legumes (e.g., clover or vetch) and non-legumes (e.g., grasses or Brassica species) are as efficient cover crops for reducing NO_3 leaching as monocultures of non-legume species. Continued regular use of cover crops has the potential to reduce NO_3 leaching and enhance soil organic matter but may enhance N_2O emissions. There is a need to optimise the use of crops and cover crops to enhance the synchrony of mineralisation with crop N uptake to enhance crop productivity, and this will at the same time reduce long-term risks of NO_3 leaching and N_2O emissions.

We use the following abbreviations: *BNF*: biological nitrogen fixation, *C*: Carbon, *CC*: cover crops, *CH₄*: methane, *EF*: Emission factor = % of N applied emitted as N_2O-N, *N*: nitrogen, *N₂O*: nitrous oxide, *NO₃*: nitrate, *PMN*: Potentially mineralizable N, *SMN*: Soil mineral nitrogen, *SOC*: Soil organic carbon, *SOM*: Soil organic matter.

and terminology: *Background emissions*: N_2O emissions that derive from N released from SOM as opposed to N added through fertilization, soil amendments or plant residue applied in the current year, *Degradable C*: Easily degradable carbon compounds, *Hot moments of N₂O flux*: Brief and disproportionally high short-term N_2O flux event due to the combination of multiple influencing factors (Molodovskaya et al., 2012), *N₂O emission*: The cumulative flux reported for one field treatment during the actual measurement period, *NO₃ leached*: NO_3 transported below the root zone, *PMN***:** The amount of SMN released from organic matter under ideal soil environmental conditions, *SMN:* The content of mineral N in the form or ammonium (NH_4) and NO_3 in soil.

1 Introduction

Biologically available nitrogen (N) or reactive N is limited in most natural terrestrial ecosystems. In modern crop production, addition of N fertilizer has become crucial to achieve high crop yields. This has resulted in cropping systems where a substantial proportion of the N added is lost to the environment, and where the excess reactive N threatens the quality of air, water and ecosystems (Robertson and Vitousek, 2009). The emissions of N_2O have considerable environmental impacts through the contribution to global warming and ozone depletion (Ravishankara et al., 2009), and about 16 to 20 Tg N_2O-N is emitted annually to the atmosphere. Of this, close to 40% is anthropogenic, and agriculture accounts for 67–80% of the anthropogenic N_2O emissions (Ussiri and Lal, 2013). About half of the anthropogenic N_2O emissions originate from cultivated soils (Stehfest and Bouwman, 2006). In addition, agricultural soils are sources of indirect N_2O emissions resulting from downstream microbial turnover of N from NO_3 leaching or ammonia volatilization (IPCC, 2006). NO_3 lost by leaching may also contaminate drinking water and lead to eutrophication of freshwater and marine ecosystems (Dalgaard et al., 2014).

The area under organic production increases worldwide (Willer and Lernoud, 2018). In Europe, 2.7% of the agricultural land is under organic farming, and in nine countries, 10% or more of the agricultural land is managed organically (Willer et al., 2018). In 2016, 43% (6 mill ha) of the organic farmed area in Europe was under arable crops.

Organic agriculture aims to be an environmentally friendly production system that sustains the health of soils, ecosystems and people. It should rely on ecological processes, biodiversity and nutrient cycles adapted to local conditions, rather than the use of inputs (IFOAM, 2019). Because of the serious consequences of N_2O emissions and NO_3 leaching, these environmental burdens are important issues also for organic farming, and there is a continued debate whether an organic mode of crop production enhances or reduces greenhouse gas (GHG) emissions and NO_3 leaching from agriculture (Lorenz and Lal, 2016; McGee, 2015). We have chosen to focus on arable systems because crop and soil management vary more between organic and non-organic production of arable crops than for grassland (Barbieri et al., 2017), and the yield gap is larger (De Ponti et al., 2012). These conditions will affect N_2O emissions and NO_3 from the systems. Previous reviews have compared the net N_2O emissions and NO_3 leaching from these systems. Lower area-scaled, but roughly similar yield-scaled emissions (slightly higher, similar or slightly lower) are commonly observed for N_2O emissions (Skinner et al., 2019; Skinner et al., 2014; Tuomisto et al., 2012,) and NO_3 leaching (Aronsson et al., 2007; Benoit et al., 2014; Kirchmann and Bergström, 2001; Stopes et al., 2002; Tuomisto et al., 2012) from organic versus non-organic arable crop production.

In this review we focus on the drivers for N_2O emissions and NO_3-leaching under organic arable crop rotations under European climatic conditions. There is insufficient robust field data on N_2O emission and NO_3 leaching within organic arable crop rotations to allow for a meta-analysis to quantify the impact of key causes, so here we use the available data to identify sources and causes of N_2O emission and NO_3 leaching in these rotations, as a basis for suggesting targeted mitigation strategies. We

define "organic arable crop rotations" as cropping systems with associated crop and soil management commonly used in European farms dominated by arable cropping and following the European Council Regulation (EC) No 834/2007s on organic farming (Council of the European Union, 2007). Among others, the use of synthetic N-fertilizers and N-inhibitors are prohibited, but manure and/or short-term leys may be used in these rotations. We designate "non-organic crop systems" as

5 arable cropping systems generally based on inorganic fertilizers, use of pesticides and often using narrow crop rotations, commonly called conventional farming.

Both globally and in Europe organic rotations are longer and more diversified than non-organic rotations (Barbieri et al., 2017). This is essential for nitrogen supply, and for pest and weed control (Stockdale et al., 2001). Barbieri et al. (2017) found that catch crops and undersown cover crops are 2.4 and 8.7 times more frequent in organic compared to conventional systems,

10 respectively. They further found that the share of pulses and temporary fodder crops (such as alfalfa, clover and ryegrass) were higher in organic than in non-organic crop rotations, and that the difference between organic and non-organic crop rotations was greater in this respect in Europe than in North America and globally. They further found that more legumes are included in fodder crops, and in catch crops, under sown cover crops and intercropping than in non-organic rotations. In addition to plant derived N, organic N are applied in manure or other organic fertilizers and amendments. The great diversity of N

15 mineralisation patterns among the organic fertilizers and crop residues is a challenge for farm management to synchronize the N release with plant N uptake. If N is released during periods with poor plant uptake, then the content of soil mineral nitrogen (SMN) and other easily available N can accumulate, creating a large risk of N losses through gaseous emissions or through leaching. Because N is mainly applied through plant residues and a limited amount of organic fertilizers in arable organic systems, the N-turnover from biological activity is crucial for the content and type of SMN. Plants and organic fertilizers are

20 also important sources of soil organic carbon (SOC). We address how supply and quality of organic matter in above- and belowground residues and organic amendments influence availability and type of SMN and degradable carbon.

Increasing the content of SOC enhances the risk of N_2O emissions (Li et al., 2005). This is true whether the soil has a high content of SOC or the content is increased by additions of organic matter to the soil and is caused by the tight link between SOC and microbial N_2O production (Sahrawat and Keeney, 1986). Because the impact of SOC on N_2O emission is

25 dependent on NO_3 content in soil (e.g. Weier et al., 1992; Li et al., 2005), we address how supply of N and carbon through organic inputs drive N_2O emissions in organic arable rotations. Based on IPCC (2006) most inventories and farm models assume that 1% of total N-input by fertilizers, manure and plant residues are emitted as N_2O-N. Skinner et al. (2014) found no correlation between total N-input and N_2O emission in organic systems. If this is a general trend, the total N-input cannot be used to estimate N_2O emissions from organic crop rotations. In non-organic cropping systems, peak fluxes of N_2O are

30 commonly observed shortly after fertilization with mineral fertilizers in moist soil (Smith et al., 2012), whereas in organic crop rotations the highest fluxes are often observed after incorporation of plant residues (e.g. Brozyna et al., 2013; Krauss et al., 2017b; Nadeem et al., 2012; Pappa et al., 2011; Skinner et al., 2019). Because of enhanced content of SOM and thus a larger impact of background emissions of N_2O in organic versus non-organic cropping systems (Skinner et al., 2014), increased background emissions are likely to have a major impact in organic-crop rotations. In order to design good

35 mitigation strategies, it is useful to know the relative importance of these two sources of N_2O emissions.

The main sources for NO_3 leaching are NO_3 from nitrification of plant residues and added organic matter, as NO_3-fertilizers are prohibited in organic systems. The crop N requirement dependency on soil organic matter turnover may lead to an asynchrony between crop nutrient demand and the mineralization of soil organic N, which enhances the risk of NO_3 leaching (Crew et al., 2005; Di and Cameron, 2002). Furthermore, soil cultivation for weeding or incorporation of plant residues have

40 been shown to influence NO_3 leaching (Askegaard et al., 2011). We discuss the main drivers for NO_3 leaching in organic arable cropping systems and the associated preventative measures.

We address the following questions for organic arable crop rotations:

1. How does supply and quality of organic matter in above- and below ground residues and organic amendments influence availability of easily available N and degradable C?

2. How does supply of easily available N and degradable C drive N_2O emissions and how can these be mitigated?

3. Is there a lack of correlation between total N-input and N_2O emission in organic arable crop rotations, and are total N_2O emissions primarily driven by background emissions or by episodes with high N_2O fluxes following N additions?

4. What are the main drivers for NO_3 leaching in organic arable systems and how can the leaching be reduced?

2. Methodology

Based on the authors own field trials, literature databases and searches through Google Scholar, we compiled data on agronomic management, soil properties and yield level of organic arable crop rotations and measurements of SMN, N_2O emissions and NO_3 leaching from field trials relevant to organic crop rotations, climate and soil conditions in Europe. For SMN and NO_3 leaching, we used the available literature and the data in Tables S1 and S3 to explore the importance of the determining factors for SMN and NO_3 leaching, and to identify factors of importance for NO_3 leaching in organic arable crop rotations. For SMN and NO_3 leaching the structure of the available data did not allow meaningful statistical analyses.

For N_2O we used data presented in Table S2 to explore the impact of background emissions and episodes with high N_2O fluxes. We aimed to analyse the impact of high emission events of N_2O fluxes on the total N_2O emission. However, we lacked daily measurements, and we lacked data for yearly periods. It was thus not possible to identify the full impact of hot moments as done by Molodovskaya et al. (2012). Because of the differences in measurement period, it was also not possible to make direct comparisons between the different field trials. To overcome this, we used a regression model based on N_2O emissions in the actual period and peak N_2O flux within this period, resulting in the following fitted model:

$$\ln(C/P) = -1.34 + 0.83 \ln(N+2), R^2 = 0.66, n = 97 \qquad \text{(Eq. 1)}$$

C is N_2O emission in the measurement period (-278 to 8566 g N_2O-N ha^{-1}); P is duration of the period (38 to 490 days); C/P express the average daily N_2O flux (-1.3 to 53.2 g N_2O-N ha^{-1} day^{-1}) and N is the highest N_2O-flux rate (0.1 to 605 g N_2O-N ha^{-1} day^{-1}) in the measurement period. The total analysis and data used is given in S4. One negative value for average daily N_2O flux (-1.3) in a barley/pea crop was removed from the analyses because it would have had a large impact on the results.

We also calculated the percentage contribution of the highest daily N_2O flux of the total N_2O emissions in the measurement periods for all trials presented in Table S2 (n=97), and correspondingly the sum of the fluxes for the days with the five highest flux rates as a percentage of the total N_2O emissions.

The impact of the following potential explanatory variables on the highest daily N_2O flux rates: clay (soil clay content %), pH (soil pH), SOC (soil organic C, g kg^{-1} dry soil), WFPS (soil water filled pore space, %), NO3 (soil content of NO_3, kg NO_3-N ha^{-1}) and NH4 (soil content of NH_4, kg NH_4-N ha^{-1}) and temp (soil temperature, °C) was calculated by stepwise regression (α to enter = 0.15; α to remove = 0.15) by Minitab 18. The selection of these variables was based on expected impact on N_2O emission (Sect. 4.1) and the data we were able to obtain from these studies. We used data from the following studies: Ball et al. (2007a), Baral et al. (2017), Brozyna et al. (2013), Chirinda et al. (2010), Krauss et al. (2017b), Li et al. (2015a), Nadeem et al. (2012b) and Pugesgaard et al. (2017). The N_2O flux data were log-transformed to achieve near normality and variance homogeneity: ln (daily N_2O flux +2), where daily N_2O flux = g N_2O-N $ha^{-1}day^{-1}$ is the highest N_2O flux rate during the actual measurement period (Table S2, Highest daily flux rate). The total analysis and data used is given in S5.

Based on the stepwise regression, we achieved the following fitted regression model:

$$\text{Ln (daily } N_2O \text{ flux + 2)} = -1.18 + 0.04 \text{ clay} + 0.06 \text{ SOC} + 0.03 \text{ WFPS} + 0.01 \text{ NO3} + 0.05 \text{ temp}, R^2=0.71, n=66 \qquad \text{(Eq. 2)}$$

We used a literature review to explore the impact of total N added and, N and C added through organic inputs from living plants, plant residues and organic fertilizers on N_2O emission as we did not have enough data on added N to be able to include this in the regression analyses.

3 Drivers of SMN and degradable C in organic arable crop rotations

3.1 Supply and quality of soil organic matter

Return of crop residues to the soil is standard practice in both organic and non-organic arable production; however, because of the more diverse crop rotations in organic production systems, larger and more diverse inputs of herbage from legume-based green manures, leys, cover crops (CC) and intercrops are returned to soil than in non-organic cropping systems commonly used in Europe (Gattinger et al., 2012). The N content in the crop residues of legume-based systems is typically higher than from non-legume systems (Watson et al., 2002). Commonly used external sources of organic inputs in organic cropping systems are animal manures and slurries, composts or biogas residues, and organic fertilizers based on animal manure or municipal waste (Løes et al., 2017). The great diversity of N mineralisation patterns among the organic fertilizers and crop residues result in a large variation in how much N and C that are rapidly degradable in the organic inputs.

Through the application of organic amendments and various crop residues from arable and forage crops, carbon (C) and N is applied to soil, and the soil organic matter (SOM) content is often higher in organic than in non-organic arable crop rotations (Marriott and Wander, 2006; Marinari et al., 2007; Gomiero et al., 2011; Gattinger et al., 2012; Aguilera et al., 2013; Hu et al., 2018). The quality of SOM differs between non-organic and the more diversified organic, arable crop rotation, with a higher share of labile SOM (Lynch, 2015) and thus easily degradable organic matter (C and N) in soils in organic crop rotations (Marinari et al., 2007; Marriott and Wander, 2006; Martyniuk et al., 2016). The higher content of degradable SOM in organic crop rotations is a valuable soil fertility asset as it provides a short-term pool for plant nutrient supply (Marriott and Wander, 2006; Martyniuk et al., 2016). SOM turnover rates vary with soil texture and climate, being higher when organic carbon is less protected from decomposers (low clay content) and in warm climates with suitable moisture (Burke et al., 1989).

3.2 Soil biological activity

Soil microbes contribute directly to plant residue decomposition and to mineralization and turnover of SOM, as well as earthworms and other soil fauna (Lubbers et al., 2013, Kuiper et al., 2013). The inflow of degradable organic matter provides substrate for soil organisms, and application of organic matter increases the growth of microbial communities, their enzyme activities and the microbial diversity compared to an unfertilised control or soil fertilized with only mineral fertilizer (Anderson and Domsch, 1989; Marinari et al., 2007; Thangarajan et al., 2013), although such changes in the topsoil may be slow processes (Petersen et al., 2013). Accordingly, higher biological activity has commonly been found in arable soils managed organically compared with non-organically (Mäder et al., 2002; Gomiero, 2013; Hartmann et al., 2015; Lori et al., 2017). In their meta-analysis, Lori et al. (2017) found that organic systems had 32% to 82% greater microbial biomass C, microbial biomass N, total phospholipid fatty-acids, and dehydrogenase, urease and protease activities than conventional systems. They found that when both organic and non-organic systems included legumes, the organic system displayed a higher microbial N content than the non-organic counterpart. In cases where only the organic systems contained legumes, the difference in microbial N between the two systems was even more pronounced. The abundance of earthworms can be twice as high in organic compared with non-organic systems (Pfiffner and Mäder., 1997; Filser et al., 1999; Hansen and Engelstad, 1999; Riley et al., 2008). More abundant earthworm populations are found when large amounts of animal manure or green manure are applied to soil (Hansen and Engelstad, 1999; Frøseth et al., 2014), when autumn ploughing is avoided (Pfiffner and Luka, 2007) and in the absence of tractor traffic (Hansen and Engelstad, 1999).

3.3 Soil N-dynamics within organic arable cropping systems

Crop N supply and SMN in organic farming relies largely on mineralisation of N in soil organic matter, N in organic amendments and crop residues and BNF of legume-based crops (Gattinger et al., 2012; Lorenz and Lal, 2016). A principal

sketch for N-dynamics in organic systems is given in Fig. 1 and an overview of performance of selected drivers of SMN in organic and non-organic crop rotations are given in Table 1.
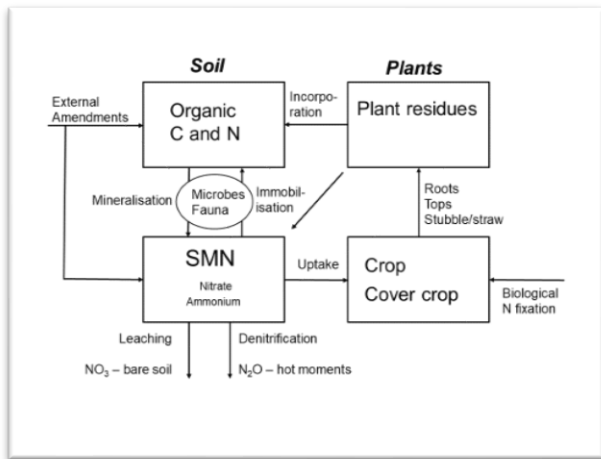
Fig.1. The principle of N-dynamic in organic crop rotations

Table 1. Performance of selected drivers of SMN in organic and non-organic cropping systems

Indicator	Organic arable systems	Conventional arable systems	*Source*
Main source of SMN	Mineralisation of crop residues and soil organic matter	Synthetic fertilizers	*Lorenz and Lal (2016)*
BNF in SMN supply	High	Low	*Kayser et al. (2010); Pandey et al. (2017)*
Size of the potentially mineralizable N pool	Usually high	Usually medium	*Poudel et al. (2002)*
Microbial biomass nitrogen content	High	Low	*Lori et al. (2017)*
Release of SMN	Slow and more continuous	Rapid with clear peaks	*Poudel et al. (2002)*
Concentration of SMN during growing season	Usually low	Usually high	*Brozyna et al. (2013) Frøseth et al. (2014) Krauss et al. (2017b)*
Concentration of SMN off growing season	Usually low, high only after termination of legume-rich crops	Usually high	*Hansen et al. (2007) Jończyk and Martyniuk (2017)*

Despite substantial inputs of N from BNF (Kayser et al., 2010; Pandey et al., 2017) and from organic amendments, the N-supply is often below optimum for plant growth in arable organic farming (Berry et al., 2002; Tuomisto et al., 2012). BNF may be a more important N source, than N from organic manures and fertilizers in organic arable cropping systems (Pandey et al., 2018).

5 The major release of plant available N from added organic matter depends on the C:N ratio, the mineral N content and degradability of the C of added plant residues manure, compost and other decomposed amendments. Bhogal et al. (2016) showed that for pig slurry and poultry layer manure with $C:N_{org}$ of 9-12:1, up to 70% of the organic N was mineralized after five growing seasons, whereas in cattle slurry and straw-based farmyard manure with $C:N_{org}$ of 10-21:1, only 10-30% of N was net mineralised. For crop residues with a high C:N content there will be initial immobilisation and the net mineralisation

10 may only start very late or after growing season of the main crop (Li et al., 2015b). The mineralization process of the more stable N can continue over years to decades. Simulation modelling has shown that even over a 20-year period, only 10-15% of organic N in applied manure may be taken up by crops, the rest being lost or retained in soil organic matter (Berntsen et al., 2007). The high microbial activity and high content of organic matter affect N-cycling. Hu et al. (2018b) found a higher net N mineralisation of added organic matter in soils having a prehistory of use of CC, indicating positive legacy effects of CC

15 use, which was attributed to a greater microbial biomass N in systems with use of CC.

The design of the rotation, as well as its management, influences the PMN pool. Working in three different organic arable systems, Spargo et al. (2011) showed that the PMN pool amounted on average for 315 kg N ha^{-1}. They showed that more diversified crop rotations resulted in larger PMN pools. Poudel et al. (2002) reported 112 and 56% greater PMN pool in the organic system in comparison to the conventional and low-input systems, respectively. Moreover, they observed slower and

20 more continuous release of mineral N in the organic systems compared to the more rapid release of mineral N from synthetic N fertilizers applied in non-organic systems. Moyo et al. (2016) reported higher PMN in soil under wheat following a cut and mulched red clover ley compared to after a ley where the residues had been removed, indicating the importance of total N input to the soil. The importance of total N input in plant residues for crop N uptake was also observed by Petersen et al. (2013).

25 SMN is found to be very low under grass-clover leys (Table S1, Watson et al., 1993; Nadeem et al., 2012; Brozyna et al., 2013; Frøseth et al., 2014; Krauss et al., 2017b), since grasses quickly take up soil NO_3 in the root zone (Brophy et al., 1987). Frøseth et al. (2014) observed low levels of SMN in a 1-year grass-clover ley, irrespective of whether the herbage was mulched or removed. After termination of a ley, the concentration of SMN usually increases (Table S1, Ball et al., 2007a; Brozyna et al., 2013; Krauss et al., 2017b). Even in the year following termination of a ley, the content of SMN can still be high (Hansen

30 et al., 2007; Jończyk and Martyniuk, 2017). Kayser et al. (2010) pointed out that N provided by spring ploughing of both 1-year grass-clover ley and 3-year grassland ley resulted in high concentrations of SMN (0-90 cm, 61 kg N ha^{-1} and 95 kg N ha^{-1}, respectively) in the following autumn after harvest of spring triticale. Much of this SMN may not have been available for the crops during spring and is likely to have been mineralised after the end of the growing season. In contrast, low concentrations of SMN were observed in topsoil and subsoil after spring ploughing of a 1-year grass-clover ley in four field

35 trials in Norway, in late spring and in autumn after harvest of a spring barley crop (Table S1) and in the following year (Frøseth, 2014, 2016).

4 Drivers of N_2O emissions in organic arable crop rotations

4.1 Mechanisms for N_2O emissions

Many processes contribute to N_2O production in soils, but the dominant mechanisms for N_2O emission in terrestrial agricultural

40 soils are the microbial processes of nitrification, nitrifier denitrification (as a result of incomplete nitrification) and denitrification (Firestone and Davidson, 1989; Butterbach-Bahl et al., 2013). Nitrification and denitrification are both

biological processes, thus the same mechanisms will cause N_2O emissions in organic as in non-organic farming systems. However, fertilisation, crop and soil management practices differ substantially between these two systems (Sect. 3), and the relative importance of the various triggers therefore differ (Sect. 6).

Nitrification is the microbial oxidation of NH_3 to NO_2 and ultimately NO_3, where N_2O is produced as by-product through some partially understood biotic and abiotic reactions of hydroxylamine (Anderson, 1964, Liu et al., 2017). Nitrifier denitrification occurs when NO_2 produced during nitrification is reduced to N_2O (by denitrifying organisms), instead of being oxidized to NO_3, under fluctuating oxic-anoxic conditions (Firestone and Davidson, 1989). Denitrification is the microbial anaerobic reduction of NO_3 via NO_2 to gaseous NO, N_2O and N_2, which are ultimately transported from soil to atmosphere. Denitrification is the main source of N_2O production in soils, as N_2O yield potential of denitrification is much higher (1-100%) than nitrification (0.1-1%) (e.g., Andersson et al., 1993, Butterbach-Bahl et al., 2013).

The ratio between the gaseous products of denitrification depends on NO_3 availability, oxygen availability in the soil and/or microsites, amount of easily decomposable carbon as an energy source, soil pH and microbial community structure (Bakken et al., 2012). Oxygen availability depends on soil microbial activity and gas diffusivity, which depends on soil moisture content, texture and density. Gas diffusivity is a promising predictor for N_2O fluxes from soils with varying bulk density as observed by Balaine et al. (2013), who found that the production of N_2O increased when the relative gas diffusivity was between 0.006 and 0.020 and the soil became anaerobic.

As explained above, risk of N_2O emissions increases as soil carbon content increases (Li et al., 2005). N_2O and N_2 production correlates with total organic C, water soluble C and mineralizable C in soil, but increased availability of C also decreases the ratio of N_2O/N_2 (Sahrawat and Keeney, 1986). Emissions from soil organic matter (background emission) will vary between years because of variations in temperature and precipitation (Hansen et al., 2014; Brozyna et al., 2013) and between different categories of crops due to different time windows with high SMN (Dobbie and Smith, 2003).

Organic amendments and plant residues that provide carbon that is easily decomposable by microbes may enhance microbial activity and deplete soil oxygen through enhanced soil respiration. In addition, degradable carbon is an energy source for denitrifying bacteria. In accordance with this, Köster et al. (2011) concluded that bacterial denitrification was the main process for producing N_2O during the first three weeks after application of biogas residues, and high carbon availability was an important cause for this. Li et al. (2016) concluded that denitrification was the main cause for N_2O emission after addition of legume-based residues. Several studies have shown higher rates of N loss through denitrification from soils treated with organic amendments such as manure, composts, and plant residues when compared to unamended or mineral N treated soils (Thangarajan et al., 2013). In line with this, incorporation of residues by tillage increases soil respiration and N_2O fluxes because of microbial stimulation (Krauss, 2017a).

Low soil pH inhibits the activity of N_2O reductase enzyme and thus $N_2O:N_2$ ratio increases (Liu et al., 2010). At higher soil pH, the denitrification rate is higher, but the $N_2O:N_2$ ratio is lower as a greater part is completely denitrified to N_2. At low temperatures, nitrous oxide reductase is hampered (Holtan-Hartwig et al., 2002), but on the other hand, denitrification rates are also reduced (Butterbach-Bahl et al., 2013).

4.2 Legumes during active plant growth

In general, unfertilized legumes have small N_2O emissions during their growing period, particularly when grown in mixtures with non-legumes. Low N_2O emission are found during growth of grain legumes (Dusenbury et al., 2008; Jensen et al., 2012; Jeuffroy et al., 2013; Pappa et al., 2011; Rochette and Janzen, 2005), green manure crops and CC (Baggs et al., 2000b; Brozyna et al., 2013; Peyrard et al., 2016; Li et al., 2015a; Shelton et al., 2018) as well as for grass-clover leys (Baggs et al., 2000b; Ball et al., 2002; Brozyna et al., 2013; Skinner et al., 2019; Krauss et al., 2017b; Nadeem et al., 2012). This is consistent with low SMN concentrations during growth (Sect. 3) and negligible N_2O emissions associated with BNF by the legume rhizobium

symbioses (Rochette and Janzen, 2005; Carter and Ambus, 2006). However, following termination or senescence of the legume crops, reactive N released from dying roots and nodules may lead to enhanced N_2O emission (Rochette and Janzen, 2005). Surface mulching of harvested herbage may theoretically enhance N_2O emissions due to mineral N released from the herbage. However, several studies show that mulching of grass-clover herbage on the growing ley only causes a slight increase in N_2O emissions (Brozyna et al., 2013; Möller and Stinner, 2009; Nadeem et al., 2012). None of these studies measured ammonia volatilization from mulched herbage, which could have been a major loss of mineralized N corresponding to the findings of Larsson et al. (1998). Volatilized NH_3 will be redeposited elsewhere and may result in increased N_2O formation downstream (IPCC, 2006).

4.3 Crop residues

As outlined in Sect. 4.1, there is an enhanced risk of N_2O emission from agricultural soils when easily degradable carbon and N are simultaneously available, and denitrification is probably the main source for this. Because legume-based crop residues also increase SMN (Sect. 3.3), increased N_2O emissions have been reported in field trials whether the residues are from grain legumes (Jeuffroy et al., 2013; Pappa et al., 2011), grass-clover (Baggs et al., 2000b; Ball et al., 2007a; Skinner et al., 2019; Nadeem et al., 2012; Brozyna et al., 2013), intercropped clover (Pappa et al., 2011) or CCs (Baggs et al., 2000b; Peyrard et al., 2016, Pugesgaard et al., 2017). The increase of N_2O fluxes after incorporation of crop residues and other plant material might however, be small and a negligible part of the total N_2O emissions (Peyrard et al., 2016; Pugesgaard et al., 2017; Shelton et al., 2018).

The C/N ratio of incorporated herbage do affect N_2O emissions, with higher emissions expected from herbage having low C/N-ratio (Chen et al., 2013). From this, one should expect higher N_2O emission from legume residues than from cereals or grasses (e.g. Rochette and Janzen, 2005). However, Larsson et al. (1998) observed the same N_2O-N EF (1% of applied-N) from mulched alfalfa (C/N-ratio 11) as from mulched grass with a C/N ratio of 21, but higher EF than from a mulched grass with a low N-content (C/N-ratio 36, EF = 0.1%). The N_2O fluxes might be high despite a high C/N-ratio when the carbon source is easily degradable as observed by fodder radish by Li et al. (2015a) (Table S2).

N_2O emissions may also be associated with previous incorporation of plant residues. In accordance with this, Skinner et al. (2019) observed enhanced N_2O fluxes after a maize crop succeeding a grass-clover ley. Measurement of N_2O fluxes shortly after incorporation of plant material, or measurements in the following year, only tell part of the story. Enhanced content of various fractions of SOM derived from crop residues, ley and CC (Sect. 3.2) are likely to increase the long-term background emissions of N_2O (Sect. 4.1). In a ten-year-old field experiment with and without legume-rich CC in the crop rotation, Pugesgaard et al. (2017) concluded that crop residues were important source of N_2O, and that mineralizable C, rather than N input, was the main driver for N_2O emission. Contrary to this, Peyrard et al. (2016) observed in a three-year low-input field trial that although N_2O fluxes increased for a few days after incorporation of CC, the contribution of such events to cumulative N_2O emissions were negligible. In their study, however, the CC treatments started when the N_2O measurements started. More studies in long-term experiments with continuous use of CC are needed to verify the actual impact of crop residues in a long-term perspective in various field situations, because addition of plant material to soil also affect soil structure, soil biological activity and N turnover.

4.3.1 Freeze/thaw – dry/wet

The mechanisms behind freeze/thaw have been comprehensively reviewed by Congreves et al. (2018) showing that the causes for N_2O emissions are different for these two mechanisms and that freeze/thaw has a larger impact on N_2O emissions in temperate agroecosystems than drying/rewetting. Wagner-Riddle et al. (2017) estimated that neglect of freeze/thaw emissions underestimate global agricultural N_2O emissions by 17 to 28%. Freezing/thawing of soil rich in organic matter and soil biota, or soil covered with plant residues may result in a N_2O boost as easily degradable C and N is released from cells through lysis

after frost. As summarized in the introduction and in Sect. 3 these conditions are particularly relevant for organic crop rotations. Flessa et al. (1995) observed that 46% of total annual N_2O emissions from a sunflower crop, solely fertilized with farmyard manure (12 Mg ha^{-1}) occurred during December and January, mainly due to high N_2O peak fluxes (650 g N_2O-N ha^{-1}day^{-1}) after thawing of the first freezing period during winter. Correspondingly, Westphal et al. (2018) did not observe any enhanced

5   N_2O fluxes after late summer incorporation of a ley dominated by alfalfa (0-10 g N_2O-N ha^{-1} day^{-1}), but fluxes were greatly enhanced during spring thaw in the following year (60 g N_2O-N ha^{-1}day^{-1}).

When CCs are killed by frost, N_2O fluxes will increase during thawing of the soil because of release of easily degradable C and N in the plant material. Li et al. (2015a) observed significantly higher emissions from the frost-sensitive fodder radish rich in readily degradable carbon than from other less frost sensitive CCs. Winter emissions were even greater when fodder radish

10   was harvested in late autumn (30 October), leaving only roots and stubble (Table S2). This suggests that N and C in roots of frost sensitive CCs can be an important source for N_2O emissions after thawing. Also, in leys frost may enhance N_2O emissions. Sturite et al. (2014) observed that enhance N_2O emission during thawing of a frozen grass-clover ley correlated with clover content in the ley.

Under drought conditions, the nitrification process prevails and N_2O is produced at very low rates. However, with rewetting

15   easily degradable N and C is mineralized, resulting in increased N_2O fluxes. Hansen et al. (2014) observed that the N_2O flux increased with increasing clover content during rewetting of a grass-clover ley after drought. Hence, both freezing/thawing and rewetting may have a large impact in organic systems.

4.3.2 Soil and tillage effects

N_2O emissions associated with crop residues are affected by tillage depth and soil type. Large N_2O emissions have been

20   observed when crop residues are placed near the soil surface in heavy soil (Peyrard et al., 2016, Krauss et al., 2017b) whereas in lighter soil types the highest emission has been observed either after rotary harrow (Baggs et al., 2000a) or after ploughing (Petersen et al., 2011). When the crop residues are squeezed and mixed with a rotary harrow, easily available N and degradable C become available for denitrifying bacteria in soil and the potential for denitrification is large. In line with this, Krauss et al. (2017b) observed high N_2O fluxes few days after weeds and crop residues were superficially incorporated with a rotary harrow

25   in a moist calcareous clay soil (WFPS 80 %) (Table S2, highest observed peak in single plot 800 g N_2O-N ha^{-1}day^{-1}). Similarly, Peyrard et al. (2016) observed enhanced N_2O fluxes (max rates 60 g N_2O-N ha^{-1}day^{-1}) up to several days after crop destruction when crop residues (sunflower, wheat, faba bean) were mulched or placed near the soil surface of a calcareous-clay, but not by ploughing or mechanical weeding. Baggs et al. (2000a) observed higher N_2O fluxes when lettuce residues were incorporated by rotary harrow than by ploughing (peak of 67 g N_2O-N ha^{-1}day^{-1}).

30   Restricted gas diffusivity is another possible explanation for the observed lower N_2O fluxes with deep incorporation of crop residues in dense soil. With reduced gas diffusivity more N_2O are likely reduced to N_2, in accordance with a general trend of a larger ratio of N_2O-N/(N_2O-N+N_2-N) close to the soil surface and smaller fluxes deeper in the soil profile (Sahrawat and Keeney, 1986). Kuntz et al. (2016) observed a decreased O_2 concentration at 8 cm soil depth and a corresponding reduction of N_2O to N_2 with surface application of carbon rich material. As another example, Petersen et al. (2011) found in their loamy

35   sandy soil that the largest fluxes were observed when residues were incorporated by ploughing compared to reduced tillage. Possible explanations for this could be that residues came directly into contact with mineral N from the injected slurry after ploughing, thus fostering enhanced microbial turnover of C and N and that in this soil, the aeration with O_2 was still available at plough depth.

4.4 Organic fertilizers

40   Organic fertilizers vary widely in the content and types of N and C compounds causing large variations in N_2O emissions after application. Animal slurries have a higher content of NH_4-N and contain more easily degradable N and C than solid manures

and composts and are thus stronger triggers for rapid N_2O emissions shortly after application (Charles et al., 2017, Sect. 3). In accordance with this, Krauss et al. (2017b) observed higher N_2O emissions shortly after application of cattle slurry than composted solid cattle manure. Correspondingly, in a field experiment with spring barley fertilized with various organic slurries, Baral et al. (2017) observed the highest N_2O EF in the treatment with highest application of organic matter, and thus

5   highest content of easily degradable C. Meijide et al. (2007) and Chantigny et al. (2007) found that the use of digested slurry, with lower content of degradable C compared to untreated pig slurry, reduced soil N_2O emissions by 25 and 50% respectively. The effect of organic fertilizers does depend on soil type and content of SOC. Degradable C applied with organic fertilizers will to a greater extent trigger microbial respiration and denitrification in a soil with low content of SOC than in a soil with high content of SOC (Chantigny et al., 2010, Pelster et al., 2012), whereas in a soil with high content of SOC the impact of

10   easily available N is higher (Petersen et al., 2008). Because of a higher content of SOC and a higher share of labile SOM in organic crop rotations compared with non-organic crop rotations (Sect. 3.1), the short-term effect of organic fertilizers with a high content of degradable C on N_2O emission are likely lower in organic than in non-organic crop rotations.

Also, the absence of synthetic fertilizers means that organic fertilizers are likely to have a smaller short-term impact on N_2O emission in organic than in non-organic crop production. Charles et al. (2017) found in a meta-analysis that the N_2O EF was

15   higher when soils received organic amendments in combination with synthetic fertilizers. They found EFs for liquid manures + synthetic fertilizers: 2.14 % ($\pm$ 0.53), composts + synthetic fertilizers: 0.37 % ($\pm$ 0.24), and the corresponding EFs for manure: 1.12 % ($\pm$0 .18) and compost: 0.00% ($\pm$ 0.17).

However, the long-term impact of manures is not included in these EFs. In contrast to the short-term fertilizer effect, a long-term fertilization with organic fertilizers may enhance N_2O emissions through enhanced background emissions. Chang et al.

20   (1998) observed that annual N_2O emissions increased with manure rate when different rates of solid feedlot manure and thus N-application were applied for 21 years. Their manure rate was far greater than would be applied under organic farming conditions, but the possibility for enhanced background N_2O emissions after long-term input of organic matter through manuring and application of crop residues should be considered. Krauss et al. (2017b) found that fertilization with slurry and manure compost increased annual N_2O emissions during winter wheat after more than ten years of differentiated management

25   compared to sole slurry fertilization (mean values in the period (369 days), were 2.2 and 2.9 kg N_2O-N ha^{-1}, respectively). They related this to higher microbial biomass and SOC. Mean values for the upper 10 cm in soil were 28 and 30 Mg C ha^{-1} for fertilization with slurry and manure compost and sole slurry fertilization, respectively.

4.5 Contribution of total N-input and high emission events to N_2O emissions

Skinner et al. (2014) concluded in a review that soil characteristics (soil N content) had a greater impact on N_2O emissions

30   from organic production than the total-N input by fertilization. Lack of correlation between N_2O emissions and N fertilization in organic production corresponds to the more recent findings of Krauss et al. (2017b) and Pugesgaard et al. (2017). Pugesgaard et al. (2017) observed no significant correlation between N_2O emissions and N input in fertilizer/manure, for either annual emissions or spring emissions, but N_2O emissions were correlated with N input in residues from the previous main crop and CC. This agrees with the findings of Bouwman et al. (2002), van Groenigen et al. (2010), Peyrard et al. (2016) and Shcherbak

35   et al. (2014), who observed low EF's when N fertilization was below optimum as commonly found in organic production systems.

As discussed earlier N_2O emission in organic crop rotations are driven by enhanced background emissions from long term input of organic matter, and episodes with enhanced N_2O emission after application of crop residues or organic fertilizer, but are N_2O emissions primarily driven by background emissions or by episodes with high N_2O fluxes? We used data from mainly

40   organic field trials (Table S2) and one non-organic trial fertilized with organic fertilizers (Baral et al., 2017) to calculate the impact of the highest N_2O fluxes on the total N_2O emissions. In Frick (CH), Edinburgh (UK), Aberdeen (UK) and Ås (NO), the highest daily flux rates were 605, 211, 297 and 94 g N_2O-N ha^{-1}day^{-1}, respectively (Table S2). Because of the high flux

rates, we hypothesised that high emission were responsible for a major part of the N_2O emissions from these systems. The single days with the highest fluxes correspond to 18% (65 days measurement period) in Frick, 2% in Edinburgh (161 days), 17 % in Aberdeen (38 days), and 2% in Ås (218 days) of the cumulated N_2O emissions in the measurement periods. The five highest daily N_2O fluxes corresponded to 22, 7, 55 and 5% of the N_2O emission in the measurement periods in these

5   investigations, respectively. In field trials conducted on well-structured sandy loams at either Foulum or Flakkebjerg in Denmark (Table S2, 105 to 365 days), peak N_2O fluxes from one or five days, however, only constituted from <1-8% and 5-14% of the total emission in the periods, respectively. The highest daily flux rate in these trials was only 78 g N_2O-N $ha^{-1}day^{-1}$. This was in a non-organic treatment heavily fertilized with cattle slurry and digested sewage sludge (476 kg total N ha^{-1}, Table S2 (Baral et al., 2017)). From this we reject the hypothesis that high emission events were responsible for a major part

10   of the N_2O emissions from these systems, rather background emissions seemed to be the major N_2O source. However, a simple regression model (Eq. 1) showed that the average daily N_2O flux correlated positively with episodes with high N_2O fluxes. From this we can conclude that when the conditions for high N_2O fluxes are met for one or more days, there is a large chance for high total N_2O emissions in the period. The small peaks in the Danish field trials reveals that well-drained sandy soils promote rapid water infiltration and good gas-diffusivity and in turn low N_2O emissions.

15   We wanted to explore the impact of soil conditions on the highest N_2O flux peak. In a stepwise regression (Eq. 2), the content of clay, SOC, NO_3-N in soil and soil temperature had significant positive impacts on peak N_2O fluxes in the selected investigations (Table S2). (P_{clay}, $P_{SOC} < 0.001$, P_{NO3}, $P_{temperature} < 0.01$, $P_{WFPS} < 0.05$). The content of NH_4-N in soil did not affect peak N_2O fluxes. These findings indicate that denitrification is the main cause for high N_2O-flux rates in these studies. To visualize the impact of different factors on the mean and the highest N_2O flux, and the contribution of the 5 highest daily

20   fluxes to the cumulated N_2O emissions in the measurement period we grouped the investigations according to % clay in soil, N added with organic fertilizers, SOC, soil pH, experimental period, type of crop and mean daily precipitation in the period (Fig. 2). Because of lack of information, we were neither able to include total-N input nor soil porosity in the regression analyses and the box-plots.

In line with the findings of Skinner et al. (2014) we did not find a clear impact of N added with organic fertilizers, but the soils

25   with the lowest content of SOC, showed the lowest mean and lowest peak N_2O emission. This supports a hypothesis that the SOC content of the soil, and thus the content of SOM, are important drivers for N_2O emission. At high SOC contents other factors seem to be more important. The impact of type of crop on N_2O emissions do mainly follow the trend we have seen in other investigations (Sect.s 5.2, 5.3) with higher N_2O emissions when cereals succeed a short-term ley (grass-clover or clover) and that mixtures with monocotyledons and legumes do not have higher N_2O emissions than cultures with only

30   monocotyledons. The high residual effects of previous leys supports the idea that background emissions are the main driving force for N_2O emission in organic crop rotations. The high N_2O emissions (up to 600 g N_2O-N ha^{-1} day^{-1}) from one occasion in Switzerland with incorporation of weeds in wet soil have a large impact on all box-plots (Fig. 2). None of the other groupings in Fig. 2 showed any clear causal effect on N_2O emissions.

The low effect we observed of pH on N_2O emissions (Eq. 2, Fig. 2) is contradictory to enhanced N_2O emission often observed

35   in acid soil caused by hampered N_2O reductase enzyme (Bakken et al., 2012). A reason for this could be that in these soils and with these managements other factors meant more for N_2O emissions than pH. Although denitrification was likely to have been the main cause of N_2O fluxes at the highest N_2O flux rates, the soil NO_3 concentration are often low between the episodes with high N_2O flux rates, as observed by Brozyna et al. (2013), Chirinda et al. (2010), Li et al. (2015a), Nadeem et al. (2012) and Pappa et al. (2008).

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Fig.2. Box-plot (mean -, median x, upper and lower quartile) for (a) mean N₂O flux in the period (g N₂O-N ha⁻¹day⁻¹) and the highest daily flux rate in the same period (g N₂O-N ha⁻¹day⁻¹) and (b) % contribution of the 5 highest daily fluxes to cumulated N₂O emissions in the period. N=number of sites, n=number of observations

4.6 Impacts of earthworms

Abundant earthworm populations in organic crop rotations (Sect. 3.3) are likely to influence N_2O fluxes as they significantly affect mineralization and reduction of N compounds to N_2O and N_2 (Prieto, 2011). N_2O is emitted from intestinal microbes but is also released from nitrates emitted in body fluids in the earthworm gut as well as from casts, middens and burrows (Prieto, 2011). On the other hand, earthworms improve soil porosity and aggregate stability (Bronick and Lal, 2005) and thus gas-diffusivity and water infiltration in soils, which will reduce N_2O emission. Epigeic species (living near surface and feeding on surface litter) and anecic species (deep burrowing) are well known to enhance N_2O production, because they feed directly on decomposing herbage (Evers et al., 2010; Nebert et al., 2011; Lubbers et al., 2011). Endogeic earthworms that feed on SOM particles are most common in cultivated arable soils (Hansen and Engelstad, 1999), and they do not increase denitrification (Postma-Blaauw et al., 2006). There are too few published results to robustly predict the impact of earthworms in arable organic crop rotations on N_2O emissions as this will depend on local climatic and edaphic conditions.

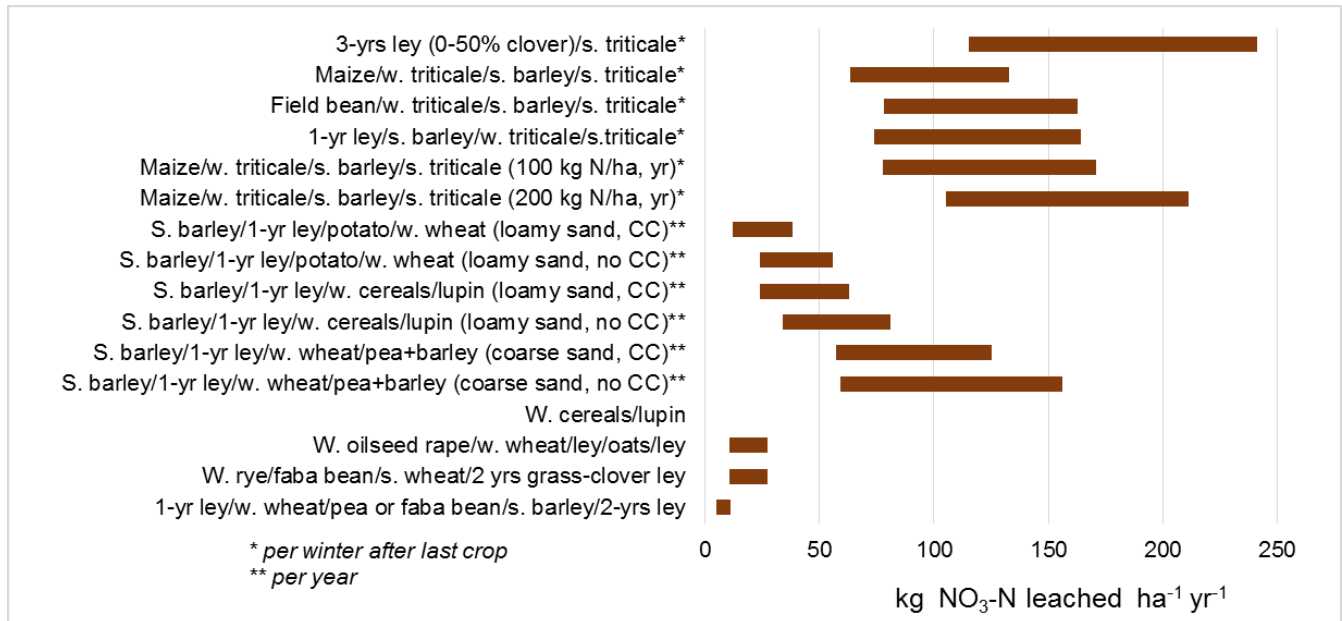
5 Drivers of NO_3 leaching in organic arable crop rotations

5.1 Mechanisms for NO_3 leaching

NO_3 leaching is an abiotic process driven by diffusion and convection (e.g. Johnsson et al., 1987), where NO_3 is transported out of the root zone along with the downward water flow. In addition to soil water content, soil texture and structure are important in determining leaching rates. Fine textured soils have slower infiltration rates than coarse textured soils, and porous sandy soils are most vulnerable to leaching, also because these soils often have more shallow rooting depths than loamy soils (Askegaard et al., 2005). The impact of soil type was clearly demonstrated by Askegaard et al. (2011) who found that, depending on soil type (coarse sand>loamy sand>sandy loam) and precipitation, 20-100 kg N ha^{-1} yr^{-1} was leached on average for the crop rotations. In their study, the location on coarse sand had 200-300 more mm rainfall per year than the other locations. The leaching was considerably higher than for Swedish clay soils: 20 kg N ha^{-1} yr^{-1} (Stenberg et al., 2012).

Due to its high mobility in soil, NO_3 can easily be lost from the agroecosystem by leaching during periods of high drainage rates. A well-developed active root system enhances NO_3 uptake, while a poor root system will not utilize all the NO_3 within the soil profile (Dunbabin et al., 2003). NO_3 remaining in the soil after the growing season of crops, or mineralised subsequently, will greatly increase the risk of leaching loss. This could happen outside the growing season, but also when there is poor crop establishment caused by unfavourable seedbed structure or from crop failure caused by diseases or pests (Stenberg et al., 2012). If crop failure coincides with rainy weather, the risk of severe NO_3 leaching is large. This was observed by Torstensson et al. (2006) and De Notaris et al. (2018) in organic farming systems, where potato growth was restricted due to early crop termination following disease outbreaks. Torstensson et al. (2006) determined the annual leaching in the potato year to be 75 kg N ha^{-1} after green manure and 98 kg N ha^{-1} after pea/barley (Table S3), whereas De Notaris et al. (2018) observed substantially higher leaching rates. They measured 213 kg N ha^{-1}yr^{-1} leached when the potato followed green manure and 133 kg N ha^{-1}yr^{-1} after grain legumes in a year with early occurring potato late blight. This was substantially higher than in previous years (140 and 78 kg N ha^{-1}, respectively).

Extreme rainfall events and/or periods with drought can significantly affect leaching for a variety of reasons. A field experiment over 13 years in the UK showed that N leaching in winter from fertilized grass (non-organic) was highly correlated with the preceding summer's soil moisture deficit, with the highest losses following dry summers (Tyson et al., 1997). In this case, poor grass growth due to drought led to a buildup of NO_3 from unused fertilizer present in the autumn. Prolonged mineralization of organic fertilizers or crop residues due to drought may also lead a similar situation in organic farming systems. Tosti et al. (2016) found, under Mediterranean rainfed conditions, that the risk of NO_3 leaching was mainly at the onset of drainage due to rainfall, i.e. at the initial stage of growth, and being typically variable among years depending on timing of heavy rains. Thus, amendments applied at pre-crop stage would be a risky practice for NO_3 leaching. Most N leaching studies in organic

farming in Mediterranean environments focused on row and vegetables crops (e.g. Campanelli and Campali, 2012), because these systems are most demanding in N inputs and thus have higher N applications and potential leaching than in common arable crops.

5 Fig. 3. Lower and upper level of NO$_3$ leached (kg NO$_3$-N ha^{-1}yr^{-1}) in various field investigations: with or without cover crops (CC), different N amount applied, and with or without grass-clover ley. The data used are given in Table S3.

5.2 Legumes

In a crop rotation with a large contribution through BNF, some of the N inputs will be retained in crop residues and in particular in mulched green manures (Frøseth et al., 2014). In their review, Crews and Peoples (2005) found that when the N input was
10 based on BNF, the proportion of the N retained in the soil was higher (58% of legume N) than in the fertilized systems (31% of fertilizer N). From this, it is likely to assume that the risk of N release outside the crop growing period are high in rotations without legumes. In their meta-analyses of crop yield and N dynamics as influenced by CCs, Tonitto et al. (2006) concluded, however, that on average, NO$_3$ leaching was reduced by 40% in legume-based systems relative to conventional fertilizer-based systems. The reason for this is probably the large difference in N input between legume-based systems relative to conventional
15 fertilizer-based systems. The response of NO$_3$ leaching to N input in fertilizer, manure and residues may also differ between sites due to soil type and precipitation (Pandey et al., 2018).

5.2.1 Grain legumes

Nitrate leaching reported from crop residues of grain legumes vary. Highest values are found when grain legumes are grown in monoculture rather than in mixtures with e.g. cereals, and when CCs are not used (Plaza-Bonilla et al., 2015). Stenberg et
20 al. (2012) observed higher NO$_3$ leaching after faba bean compared to after non-leguminous crops. On a clay soil in Sweden, they observed an average leaching of 20 kg N ha^{-1} yr^{-1}, which was twice that for spring cereals. On average over three years on loamy sand in Denmark, De Notaris et al. (2018) reported about twice as high NO$_3$ leaching following a barley/pea intercrop compared with spring wheat or spring barley. In a sandy soil in northwest Germany, Kayser et al. (2010) observed that 83 kg N ha^{-1} leached in triticale following field bean. In a worst-case scenario, Askegaard et al. (2011) observed annual NO$_3$ leaching
25 of 270 kg N ha^{-1} during and after a lupin crop on a course sandy soil in a situation where the lupin crop did not ripen, leaving a large amount of N in crop residues (same experiment as De Notaris et al. (2018)). Pappa et al. (2008) observed very low N-leaching during and after a barley/pea intercrop, but they observed a significant effect of the pea cultivar on N leaching in the autumn and winter period.

5.2.2 Forage legumes

Many authors (Kayser et al., 2010; Neumann et al., 2011; Stalenga and Jończyk, 2008) emphasize that one of the most critical times for NO_3 leaching in organic crop rotations occur after soil incorporation of a grass-clover ley. N leaching is low during the growing period of grass-clover leys (Kayser et al., 2010), but because of the large amounts of mineralized N after termination of a grass-clover ley (Sect. 3.3), the risk of NO_3 leaching is large for one to two years after termination of these crops (Berntsen et al., 2005). The leaching may occur shortly after ley termination, during winter, or during the succeeding seasons, depending on time of incorporation, quality of the herbage, the weather and the crop sequence.

Stenberg et al. (2012) observed higher NO_3 leaching following termination of a grass-clover ley than following faba bean, but values were still low (4 kg N ha^{-1} higher in average). They found the highest leaching when the grass-clover ley lasted for two years (up to 40 kg N leached ha^{-1} yr^{-1}). This corresponds to the finding of Kayser et al. (2010), who observed greater NO_3 leaching during the winter after spring incorporation of a three-year ley than after a one-year ley (121 versus 83 kg N ha^{-1}, Fig. 3). However, the crop yield of triticale yielded much better after the three-year ley than after the one-year ley. The percentage share of clover (0-5, 30 and 50%) did not influence the amount leached after ley termination, neither the crop yield. Eriksen et al. (2008) measured NO_3 leaching after 1 to 8 year old grass/clover leys, but found that the length of the ley had no effect on NO_3 leaching. Stenberg et al. (2012) observed that cereals succeeding grass-clover ley had nearly double yearly N leaching compared to cereals with no legume pre-crop. The highest NO_3 leaching occurred after cultivation of a winter rye (48 kg N ha^{-1} yr^{-1}).

De Notaris et al. (2018) observed that NO_3 leaching during cultivation of spring wheat was about 50 kg N ha^{-1} higher when the spring wheat succeeded a two-year green manure crop (alfalfa or grass-clover) than when it succeeded a grain legume (107 versus 50 kg N ha^{-1}). Similarly, Askegaard et al. (2011) observed peaks in NO_3 leaching in autumn and winter after ploughing-in grass-clover ley. At crop rotation level, inclusion of grass-clover or alfalfa on 25% of the area increased NO_3 leaching rate by 6-12 kg N ha^{-1} (De Notaris et al., 2018). Forage legumes may also be undersown as intercrops to increase soil fertility in organic crop rotations. Pappa et al. (2008) found that clover intercropped in spring barley only increased annual NO_3 leaching by 1-2 kg NO_3-N ha^{-1}.

5.3 Cover crops

CCs are grown between main crops to minimize NO_3 leaching. Many field trials in non-organic systems have shown reduced leaching using CC (e.g., Rasse et al., 2000; Torstensson and Aronsson, 2000; Constantin et al., 2010; Valkama et al., 2015). This is also the case for organic crop rotations (Tonitto et al., 2006; Askegaard et al., 2011; Tosti et al., 2014; Tosti et al., 2016; De Notaris et al., 2018). The reduction in NO_3 leaching can be substantial. Studies in Nordic countries report reductions of 50-60% in N leaching (Askegaard et al., 2011; De Notaris et al., 2018; Torstensson and Aronsson, 2000, Fig. 3). If the cash crop fails, the effect of CCs on reduced leaching can be even higher. In a year where potato late blight caused crop failure in potato, the CCs reduced N-leaching by 95% when the potato succeeded a grain legume (from 133 to 6 kg N ha^{-1} leached), and by 92% when the potato succeeded a green manure ley (from 213 to 17 kg N ha^{-1} leached) (calculated from Table S3, De Notaris et al., 2018).

De Notaris et al. (2018) concluded that the use of CCs had a larger impact on leaching than a substantial variation in N surplus between alternative cropping systems. In three long-term field trials (13-17 years) in Northern France, Constantin et al. (2010) observed that CCs were the most efficient management option for reducing NO_3 leaching (from 36 to 62%). Good establishment and growth of the CC is essential to obtain sufficient uptake of SMN and thus reduce NO_3 leaching. Stenberg et al. (1999) found no significant reduction in NO_3 leaching during winter from a ryegrass CC that was undersown in spring. They explained this by poor CC establishment. De Notaris et al. (2018) also observed occasions with very small effect of CCs on NO_3 leaching. They related this to CC growth and identified threshold values in CC above-ground biomass determined in November, above which N leaching was reduced to a stable low level. NO_3 leaching from spring wheat averaged 15 kg N ha^{-1}

yr^{-1} with CC biomass above 0.9 Mg ha^{-1}, and 41 kg N ha^{-1} yr^{-1} with CC biomass below 0.9 Mg ha^{-1}. In potatoes, the average N leaching was 11 and 80 kg N ha^{-1} yr^{-1} with CC biomass above and below 1.5 Mg ha^{-1}, respectively.

Including legumes in CC mixtures does not seem to reduce the ability of CCs to reduce NO$_3$ leaching (Tonitto et al., 2006; Tosti et al., 2014; De Notaris et al., 2018; Shelton et al., 2018). In a field trial with barley, hairy vetch and a 50:50 mixture of

5 both species as CC, Tosti et al. (2014) found that, in all years, the barley/vetch mixture decreased N leaching to the same level of pure barley, both during its own growing cycle and after CC incorporation into the soil. De Notaris et al. (2018) concluded that the same degree of reduced NO$_3$ leaching was obtained with legume-based CCs as with non-legume CCs. The CC was either undersown in spring or after harvest of the main crop. The undersown legumes were white clover and red clover, and winter vetch was used in the mixture sown after harvest. Shelton et al. (2018) found greater NO$_3$ leaching from intercropped

10 hairy vetch than from simultaneously grown wheat and wheat/hairy vetch mixture. When the CC is a pure stand of legumes, the CC does not necessarily reduce N leaching (Tosti et al., 2014; Valkama et al., 2015; Shelton et al., 2018). Tosti et al. (2014) concluded that hairy vetch sown as a pure crop in autumn showed high BNF, but no NO$_3$ leaching mitigation effect as compared to bare soil. Valkama et al. (2015) found in their meta-analysis of Nordic studies of undersown CCs that legumes (white and red clovers) in pure stand did not diminish the risk of NO$_3$ leaching.

15 **5.4 Tillage**

Tillage stimulates soil N mineralization, at least in the short term. Tillage is also often associated with soil incorporation of plant residues that may lead to net N immobilisation or mineralisation, depending on quality of the residues. Timing of tillage is therefore crucial for the fate of the mineralised N, whether SMN becomes available for crop uptake or subject to leaching. In general, incorporation should consider soil type, climate conditions and type of herbage (C/N ratio). Thorup-Kristensen and

20 Dresbøll (2010) suggested late incorporation of CCs in high rainfall areas on sandy soils, and earlier in low rainfall areas on NO$_3$ retentive soils. Field studies have shown rapid N mineralization from N-rich plant material, even at low temperatures (Breland, 1994; Thorup-Kristensen and Dresbøll, 2010). Spring incorporation has therefore been recommended to increase N recovery by subsequent crops. However, under Scandinavian conditions, there may still be a deficit in crop-available N, even after spring incorporation of a green manure ley (Frøseth et al., 2014; Känkänen et al., 1998). Under such conditions,

25 Torstensson and Aronsson (2000) suggest that late autumn incorporation of CCs, instead of spring incorporation, will be preferable with respect to N availability for the subsequent crop and will not substantially increase NO$_3$ leaching. Under Mediterranean climatic conditions, characterized by mild rainy winters and warm to hot dry summers, there is a risk of NO$_3$ leaching if residues are incorporated prior to the wet season. No studies were found that measured NO$_3$ leaching in relation to timing of tillage in organic arable farming under these conditions.

30 In organic crop production, the timing of cultivation for mechanical control of perennial weeds may conflict with the aims of high N use efficiency (Melander et al., 2016). Askegaard et al. (2011) found, on sandy soils in Denmark, that the management of crop and soil during autumn was the main determinant of N leaching. Stubble harrowing in autumn for controlling perennial weeds, followed by bare soil during winter, led to an average of 25 kg N ha^{-1} more leached than for soils left untouched with a cover of weeds/volunteers. NO$_3$ leaching increased with increasing number of autumn soil cultivations.

35 Reducing tillage intensity may also enhance the need for weed management, and thereby the risk of NO$_3$ leaching. In their meta-analysis comparing different reduced tillage intensities in organic farming, Cooper et al. (2016) found that the weed incidence was consistently higher, by about 50 %, when tillage intensity was reduced, although this did not always lower the yields. Compared to conventional tillage, reduced tillage may reduce NO$_3$ leaching, but this depends on the establishment and growth of the succeeding crop (Känkänen et al., 1998).

40 Bare fallow has traditionally been a method to control perennial weeds by repeated tillage of superficial or deeper top soil layers. In practical organic farming, a bare fallow can sometimes be used before or after the main crop if conditions have promoted perennial weeds. However, if carried out in the growing season, soil temperature and moisture conditions favour

soil microbial activity and therefore the build-up of SMN, which greatly increases the risk of NO_3 leaching (Borgen et al., 2012).

6. Key drivers of N_2O emissions and NO_3 leaching and suggested mitigation strategies

Easily available N and degradable C added through organic inputs enhances risks of high N_2O emissions in the short term because of enhanced biological activity (Sect. 3.3) and increased denitrification potential (Sect. 4.1), and in the longer term because of higher soil content of N and C in labile organic matter (Sect. 3.1) contributing to N_2O emissions from mineralized SOM (background emission, Sect. 4.1, 4.3, 4.4, 4.5). There is no strong correlation between total N-input and N_2O emissions in organic arable crop rotations (Sect. 4.5), which is another indication of the strong impact of background emissions in these systems (Sect. 4.5). The same is also true of the relatively low impact of episodes with high N_2O fluxes on the total N_2O emission in various investigations (Sect. 4.5, Fig. 2).

From this, we postulate that background emissions in most organic crop rotations are a more important driver for N_2O emissions than episodes with high N_2O flux rate (hot moments). Nevertheless, reducing periods with hot moments of denitrification and thus high N_2O fluxes rates are important for reducing total emissions. One mitigation measure is to avoid large applications of residues from crops comprising easily available N and degradable C like clover or Brassica (Sect. 4.3). Mulching of grass-clover on top of a ley seems to only slightly increase N_2O-emissiom compared with no mulch (section 4.2). Incorporating organic material in the soil surface does not reduce the risk for high N_2O fluxes and N_2O emissions are often higher after surface application than after ploughing (Sect. 4.3.2). Mixing by rotary harrow can enhance emissions, particularly in moist soil. If the crop residues are removed, and composted, used in a biogas plant or treated by other methods before targeted soil application, such events can be avoided. To avoid that these measures result in that GHG emissions only are moved to another place, this requires measures that minimise GHG emissions during and after treatment of the plant residues (see sect 7). The largest stimulation of denitrification by application of plant residues or other organic material with a high content of degradable C seems to be in soils with a low content of SOC, particularly if the soil has a high NO_3 content of or easily available N is applied (Sect. 4.4). If crop residues consist of less degradable C and easily available N like straw from cereals and grain legumes, they do not stimulate rapid denitrification and enhanced N_2O flux rate (Sect. 4.3), and the N_2O flux is likely reduced (Xia et al., 2018).

Background emissions are highly influenced by content and mineralization patterns of SOM and release of SMN and degradable C (Sect.s 3 and 4.1). Thus, weather conditions will have a large impact on the background emissions, which are likely to be higher in warm and moist years, than in cold or dry years. These conditions cannot be influenced by farmers, but it is important to develop strategies to decrease the content of SMN to reduce the risk of N_2O emissions and NO_3 leaching in periods with risk of SMN accumulation. The choice and sequence of crops and the use of CC is a central strategy for simultaneously mitigating N_2O emissions and NO_3 leaching in organic systems. Poor timing of N released from crop residues from preceding crops and crop N uptake (Sect. 3.3 and 5.2) are major challenges. There is a need to reduce periods with bare soil, although this may conflict with the need for mechanical weed control (Melander et al., 2016).

CCs take up surplus SMN and protect soil from erosion, they reduce NO_3 leaching, and during growth CCs do not contribute to N_2O emissions (Sect. 4.2). However, in areas with frost, frost sensitive crops like fodder radish, will release easily available N and degradable C during thawing, and thus stimulate denitrification (Sect. 4.3.1). The choice of CC must be adapted to local conditions for ensuring good establishment and appropriate tolerance to frost and droughts. A mixture of legumes and non-legumes (for instance grasses or cereals) are just as efficient for reducing N leaching as sole non-legume CCs, whereas sole legumes are not as efficient (Sect. 5.3). CCs containing legumes have a lower C:N ratio than CC without legumes, which enhances the N fertilizer value for the following crop (Li et al., 2015b).

The knowledge about the rooting pattern of different crop species can be used as a tool for designing crop rotations that achieve higher N use efficiency and thereby reduces the risk of NO_3 leaching. Deep-rooted crops, especially tap rooted ones, can

recover NO₃ from deeper soil layers before and after more shallow-rooted cash crops, such as leek (Thorup-Kristensen, 2006). As shown by Fan et al. (2016), the root distribution and rooting depth may differ between varieties, although plant breeders do not normally select crops based on the root system.

For the choice of crop, species and varieties should be well-adapted to the climate conditions on the farm and the soil fertility level. Crops in good conditions also compete better against weeds. This decreases the need for soil management to achieve weed control, and thereby reduces the risk of NO₃ leaching. Timing of release of N from residues and amendments and crop uptake are crucial for minimizing the risk of NO₃ leaching. This can be achieved by timing of soil tillage and incorporation of residues. In any case, the effect of mitigation strategy is highly dependent on soil type and precipitation.

Because of the large impact of poorly aerated soil on N₂O emissions (Sect. 4.1) and plant growth, measures should be taken to improve and maintain a good soil structure. In organic production, soil fauna, microorganisms and the development and maintenance of soil structure, are supported by crop rotations that include legumes or grass-clover leys, use of CCs and application of organic fertilizers. Even so, traffic and tillage under wet soil conditions are damaging to soil structure and should therefore be avoided.

A way to reduce N₂O emissions and NO₃ leaching per unit produced is to increase yields in organic production as more land is commonly needed per unit product in organic than non-organic production (De Ponti et al., 2012, Meier et al., 2015). However, as discussed by Röös et al. (2018) this is not straight forward as many of the available measures have negative side effects. More targeted and thus efficient use of N applied through crop residues and organic fertilizers seems to have co-benefits in terms for higher productivity as well as reduced N₂O emissions and NO₃ leaching.

7. Research and innovation needs

The knowledge of N cycling and losses in arable organic farming is constrained by few investigations that have adequately quantified the magnitude and timing of N₂O emissions and NO₃ leaching. Many of the reported results are from a few experiments, including a long-term experiment in Denmark (Olesen et al., 2000). Such long-term experiments are important for quantifying N transformation and loss processes in organic cropping systems, where the legacy effects of N in plant residues and other organic amendments are often considerably larger than in non-organic systems. Lessons from these experiments can help to develop methods and technologies to improve synchronisation of N released from crop residues and soil organic matter and the N demand of cash crops. However, there are few long-term experiments with organic arable crop rotations, and even fewer have been systematically used for quantifying the N cycling processes. Therefore, research on existing and new long-term experiments covering relevant soil and climatic conditions should be coordinated to make data and facilities available for research on N-dynamics, including N₂O emissions, NO₃ leaching, NH₃ volatilization and the fate of added N. In this way, useful lessons can be learned on the impact of long-term management on the N-dynamics. The availability of historical data on N-input and crop management is a prerequisite to achieve this, such data could be made available and structured through a coordination of long-term experiments.

CCs have been shown to efficiently reduce NO₃ leaching (Sect. 5.3), whereas they can reduce or increase N₂O emissions depending on the conditions (Sect. 4.2, 4.3). To optimise the use of CCs for crop productivity, environment and climate, improved knowledge is needed of locally adapted crop rotations and CC that maintain living plants that can take up available N and simultaneously reduce weed pressure.

Simulation models can be used to quantify N transformation and loss processes and how different management practices affect N storage in SOM, as well and short-term effects of crop residues and organic fertilizers on N₂O emissions and NO₃ leaching. However, process-based models fail to accurately simulate the impact of grass/clover leys on turnover of N and C (Doltra et al., 2019, Frøseth, 2016). To our knowledge, no model of field scale does include all C and N turnover processes, such as phyllodeposition and rhizodeposition. These processes may be of greater importance in organic than non-organic systems, because of the greater emphasis on fertility building measures. This may lead to underestimation of the amount of N and C

returned to soil. Improving models on these aspects requires more comprehensive data on all C and N inputs and flows in the cropping systems. This can only be achieved through studies using isotopic labelling of C and N, and consideration should be given to conducting such studies also in the long-term experiments. There is further uncertainty on the release of N from legumes and thus the impact of legume residues on N_2O emission and NO_3 leaching (see Sect.s 4.2, 4.3 and 5.2). More research

5 on the mineralization pattern of various legume residues would therefore be useful.

Heterogeneity is another aspect that make it difficult to predict availability of N and C in organic systems and thus N_2O emission. In addition to the soil heterogeneity, variability is created from incorporated crop residues, CC, short term leys and organic fertilizer that are seldom evenly distributed. Uneven impact of frost thawing / drying rewetting will add to this variability and make it difficult to predict hot spots and hot moments of N_2O emissions. There is also a need to develop

10 technologies to better measure this heterogeneity and to take account for the heterogeneity in N dynamic models adapted to organic crop rotations.

Composting of crop residues and manures is common on organic farms and is a way to make the organic matter more resistant and useful as a soil amendment. However, composting manure without emissions of N_2O and other greenhouse gases is challenging (Chadwick et al., 2011) and needs to be addressed to develop a more comprehensive picture of greenhouse gas

15 emissions from organic arable production and how to mitigate them. Neither composting nor biogas fermentation are always feasible. Improved understanding of different pathways for microbial degradation of crop residues is needed as well, to better manage crop residues and organic waste for C and N retention properties.

8. Conclusions

Organic arable production is based on diverse crop rotations, N inputs through BNF in legumes, external inputs of manure and

20 compost, and recovery of excess N in grassland and cover crops. This results in considerable inputs on N in organic matter from plant residues, green manures and application of organic fertilisers and soil amendments that enhance the content of total and labile SOC in organic arable production compared to non-organic production. When the conditions for mineralization of SOC are met this will lead to a high availability of easily available N and degradable C, and an enhanced content of SMN, if available N is not taken up by growing plants. Conditions with high content of SMN and degradable C provide hot moments

25 for high N_2O fluxes. Degradable C will increase microbial growth and thus O_2 consumption in soil leading to anaerobe conditions and provide suitable conditions for denitrification of available NO_3. In organic arable crop rotations background emissions are more important in most situations than episodes with high N_2O fluxes for the total N_2O emission from organic arable crop rotations. SMN from mineralization of SOM and plant residues in combination with periods of bare soil or sparse plant growth and precipitation surplus provide drivers for NO_3 leaching.

30 Main mitigation strategies are targeted use of growing plants to take up surplus soil mineral N from root zone, and targeted use of crop residues and organic fertilizers to synchronise the availability of N with the crop demand. Continued use of CCs has a proven ability to reduce NO_3 leaching from organic arable crop rotations but does increase N_2O emissions in some situations. A CC mixture of legumes and non-legumes (for instance grasses or cereals) is just as efficient as sole non-legumes and has a better impact on soil fertility than non-legumes. More research is needed to develop locally adapted crop rotations

35 and CC mixtures that at the same time reduce N_2O emissions and NO_3 leaching.

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40 leaching, Supplementary material Table 3 and complementary data from Swedish field trials to this Table; Jarosław Stalenga: Content of the MS, mainly SMN, Supplementary material Table S1 and Table 1; Jørgen E. Olesen: Overall content and

structure of the MS, Fig. 1, complementary data from Danish field trials in Table S2; Maike Krauss: Content of the MS, complementary data from Swiss field trial in Table S2; Paweł Radzikowski: Content of the MS, mainly earthworms, main editing of references and reference list; Jordi Doltra: Content of the MS, mainly from Mediterranean perspective; Shahid Nadeem: Content of the MS, mainly N_2O, complementary data from Norwegian field trial in Table S2; Torfinn Torp: Statistical analyses on cause of N_2O fluxes based on Table S2; Valentini Pappa: Complementary data from field trials in Edinburgh and Aberdeen in Table S2; Christine Watson: Overall development and content of the MS.

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Captions supplementary material

S1. Soil mineral nitrogen (SMN) contents in soil profiles in organic field trials in Norway and Poland as influenced by crop rotation, soil tillage and N fertilization.

S2. N$_2$O emission and the five highest daily N$_2$O flux rates in the given measurement periods for organic field trials in Switzerland, Denmark, Scotland and Norway. WFPS, soil temperature and soil mineral-N at 0-20 cm depth are given for the day with highest flux rate. Abbreviations: CS = Cattle Slurry, CCM = composted cattle manure, PS = Pig slurry, P=ploughing, H = Harrowing, CCinc= Cover crop incorporated, CCs= Cover Crop under sown, CCh = cover crop harvested

S3. Annual N leaching (total N or nitrate N) reported from organic field trials in Europe, as influenced by crops, soil type and N applied as organic fertilizers.

S4. Statistical analyses Eq. (1).

S5. Statistical analyses Eq. (2).