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A review of nitrous oxide mitigation by farm nitrogen management in temperate grassland-based agriculture

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Abstract

Nitrous oxide (N_2O) emission from grassland-based agriculture is an important source of atmospheric N_2O . It is hence crucial to explore various solutions including farm nitrogen (N) management to mitigate N_2O emissions without sacrificing farm profitability and food supply. This paper reviews major N management practices to lower N_2O emission from grassland-based agriculture. Restricted grazing by reducing grazing time is an effective way to decrease N_2O emissions from excreta patches. Balancing the protein-to-energy ratios in the diets of ruminants can also decrease N_2O emissions from excreta patches. Among the managements of synthetic fertilizer N application, only adjusting fertilizer N rate and slow-released fertilizers are proven to be effective in lowering N_2O emissions. Use of bedding materials may increase N_2O emissions from animal houses. Manure storage as slurry, manipulating slurry pH to values lower than 6 and storage as solid manure under anaerobic conditions help to reduce N_2O emissions during manure storage stage. For manure land application, N_2O emissions can be mitigated by reducing manure N inputs to levels that satisfy grass needs. Use of nitrification inhibitors can substantially lower N_2O emissions associated with applications of fertilizers and manures and from urine patches. N_2O emissions from legume based grasslands are generally lower than fertilizer-based systems. In conclusion, effective measures should be taken at each step during N flow or combined options should be used in order to mitigate N_2O emission at the farm level.

Keywords: Nitrous oxide; mitigation options; temperate grassland; N management

1. Introduction

Nitrous oxide (N₂O) is a potent greenhouse gas (GHG) with a global warming potential 298 times higher than carbon dioxide (CO₂) over a 100-year time horizon (Solomon et al., 2007). It is the third most important anthropogenic GHG and contributed about 6.0% to the overall global radiative forcing in 2011 (WMO, 2012). In addition, N₂O currently is the single most important stratospheric ozone-depleting substance and is expected to remain the largest throughout the 21st century (Ravishankara et al., 2009). Global average mixing ratio of N₂O has been increasing with a rate of 0.78 ppb yr⁻¹ over the past 10 years (WMO, 2012). The mitigation of N₂O emissions has been regarded as one of the major choices to combat climate change and has received much attention (Reay et al., 2012; Smith et al., 2012).

The challenges for mitigating N₂O emissions are substantially different from those for CO₂ and methane (CH₄) because on one hand about 90% of anthropogenic N₂O emissions are from the agricultural sector while on the other hand nitrogen (N) is essential for food production (IPCC, 2007; Davidson, 2012). The increase in N₂O emissions from agriculture is largely induced by the elevated N inputs via synthetic fertilizer N or manure (Davidson, 2009). However, to meet the nutritional needs of a growing human population more N inputs to agriculture are likely needed (Davidson, 2012). N₂O is produced mainly by two biological processes during N cycling, i.e., nitrification and denitrification, which is stimulated by N surplus between N input and crop demand (Smith et al., 2008). N₂O emissions are supposed to be reduced by increasing N use efficiency (NUE, percentage of applied N taken up by the crop), which seldom exceeds 50% (Davidson, 2012). N management to increase NUE has been recognized as an effective way to mitigate N₂O emissions from agriculture (Smith et al., 2008).

Globally, grassland-based agriculture is the major part in agriculture sector with permanent pastures responsible for 68% of all the agricultural land (FAO, 2009). Synthetic fertilizer N and manure are widely used to sustain farm productivity in intensively or semi-intensively managed grassland systems. In extensively managed grasslands a large proportion of N₂O emissions are from excreta deposited by grazing livestock, mostly from urine patches. In New Zealand and Australia, for example, where extensive grassland management is characterised as year-round grazing of grass-clover pastures and very low input of fertilizer N, direct N₂O emissions from excreta recycled to the soil surface by grazing livestock contributed between 50% and 60% of the direct N₂O emissions and up to 80% when indirect emissions (from NH₃ volatilization and NO₃⁻ leaching) are included (de Klein et al., 2001; deKlein et al., 2005). The second largest source, fertilizer N, contributed no more than 15% (de Klein et al., 2008). In more intensive managed systems with greater reliance on inputs of fertilizer N, the contribution of excreta recycled by grazing livestock can also be considerable. For example, in the Netherlands, Schils *et al.* (2005) reported that N recycled by grazing livestock accounted for 44% of total N₂O emissions compared to 22% from fertilizer N, 14% from soil and 11% from manure management in an intensive grassland-based dairy production system receiving total annual inorganic N inputs of 275 kg ha⁻¹. Indirect emissions of N₂O from leached nitrate and from volatilized NH₃ accounted for 9% of total emissions (Schils et al., 2005).

During the past two decades, a few reviews about N₂O mitigation or N losses related to grassland-based agriculture systems have been conducted, including N₂O mitigation from herbivore production systems (Schils et al., 2011), GHG and NH₃ emissions from organic mixed crop-dairy systems (Novak et al., 2011), GHG emissions from manure management (Chadwick et al., 2011), NH₃ and N₂O emissions with different manure application methods (Webb et al., 2010). In this

review, major choices of N management on grassland farms were evaluated with respect to their effectiveness to mitigate N₂O emissions. The knowledge synthesized in the review will be useful for identifying potential cost-effective and sustainable ways to mitigate N₂O emissions from grassland-based agriculture under temperate conditions.

2. Mechanisms underlying nitrous oxide emissions from grassland

The emission of N₂O arises from microbial nitrification and denitrification of inorganic N in the soil, which in turn is derived from excreta deposited by grazing livestock, application of synthetic fertilizers and manures, and biological N fixation (BNF) (Fig. 1). Nitrification consists of two steps, NH₄⁺ oxidation to NO₂⁻ and NO₂⁻ oxidation to NO₃⁻, carried out by ammonium-oxidizers and nitrite-oxidizers, respectively (Ward, 2000). Denitrification is the anaerobic microbial reduction of NO₃⁻ to dinitrogen (N₂). During the denitrification process, NO₃⁻ is successively reduced to NO₂⁻, NO, N₂O and finally dinitrogen (N₂). Since N₂O is an intermediate during denitrification, it can be both produced and consumed. Nitrification and denitrification are tightly coupled since NO₂⁻ or NO₃⁻ produced during nitrification can be utilized by denitrifiers and this coupling can take place in soils where favourable conditions for both nitrification and denitrification are present in neighbouring microhabitats (Wrage et al., 2001). However, under oxygen (O₂) limiting conditions, NH₄⁺ may be oxidized to NO₂⁻ and then sequentially reduced to NO, N₂O and N₂. This process, which is carried out by autotrophic ammonium-oxidizers is termed nitrifier denitrification (Wrage et al., 2001). The relationships between nitrification, denitrification and nitrifier denitrification are shown in Fig. 1. In addition to the above micro-organism mediated processes, some abiotic processes (mostly chemodenitrification) may also contribute to the production of N₂O under certain

conditions (Williams et al., 1992). Current evidence indicates that most of the N₂O evolved from soils is produced by biological processes and that little is produced by chemodenitrification (Bremner, 1997).

3. Potential N₂O mitigation options by farm N management

3.1 Options to lower N₂O emissions from excreta patches

Urine and dung patches on grasslands represent high (up to more than 1000 kg N ha⁻¹), random and very local additions of N and readily available carbon (C) that can create optimal conditions for N₂O production (van Groenigen et al., 2005). It was estimated that between 0.1 and 3.8% of urine-N and between 0.1 and 0.7% of the dung-N is emitted to the atmosphere as N₂O (Oenema et al., 1997). In countries that depend economically to a large extent on livestock farming, these fluxes are major contribution to the national GHG budget. It is therefore imperative to seek measures lower N₂O emissions from excreta patches.

3.1.1 Restricted grazing

Restricting grazing has been proposed as an option to reduce N₂O and other GHG emissions (Oenema et al., 2001; Schils *et al.*, 2006; de Klein *et al.* 2006; Luo *et al.* 2008). This management tactic involves a reduction in grazing time or livestock number, each of which results in decreased dung and urine deposition. Therefore there is a great potential to lower N₂O emissions via decrease in excreta deposition to pasture. In Ireland, there has been a 14% reduction in GHG emissions

associated with dung and urine deposited by grazing livestock since 1998, mainly due to decreased livestock numbers, particularly sheep, arising from the policy of extensification driven by the Common Agricultural Policy (CAP) (Humphreys, 2008). Although both N₂O and GHG emissions could be mitigated by decreased livestock numbers, this option would mean a profound decrease in the farm profitability. In addition, the growing human population requires continuing increase in animal protein consumption, as it was projected that animal numbers will increase by 40% between 2000 and 2030 (Oenema et al., 2005). So reducing numbers of animals to lower N₂O emissions is unlikely to be acceptable.

A compromise way to restrict grazing is to reduce grazing time within a year. With this practice, animals are kept off grazing paddocks and kept on stand-off/feed pads or in animal houses at a period when it leads to greatest N losses, so excreta deposition is reduced (Luo et al., 2010). This practice provides opportunity for controlling N losses, as the animal excreta is collected and can be applied evenly to the pasture at targeted rates and optimum time when the risk for N losses is minimal (van der Meer, 2008). Recent research by Kennedy *et al.* (2009) has shown that dairy cows given restricted access time (9 h per day) to pasture under unfavourable conditions for grazing had similar pasture intake and milk production to that of unrestricted dairy cows. In the study, restricted cows were kept indoors and did not receive alternative feed to replace the grazed pasture. Excreta generated indoors were collected, stored and applied mechanically under favourable conditions for herbage production. When slurry is applied mechanically it is generally more evenly distributed than when it is deposited by grazing animals; hence, the N contained within the slurry is used with greater efficiency and accordingly with less N₂O loss. There is an economic incentive to impose this management practice during periods when soils are vulnerable to poaching damage and there is a spin-off benefit in terms of N₂O emissions. Reductions in total direct and indirect N₂O emissions of

7 to 11% have been reported when conventional grazing management was partially replaced by restricted grazing during the wetter months of the year in New Zealand (de Klein et al., 2006; Luo et al., 2008b).

3.1.2 Dietary manipulation or additives

There is an exponential relationship between N intake and N excretion in urine (Ledgard et al., 2009). Ruminants on lush spring pasture commonly ingest protein in excess of their requirements, but are usually energy limited, resulting in higher ruminal ammonia concentrations being excreted in the urine as urea (Whitehead, 1995). Therefore, balancing the protein-to-energy ratios in the diets of ruminants is important in minimizing the N₂O emissions resulting from excess urinary N excretion (Eckard et al., 2010). Dairy cows fed a 14% crude protein (CP) diet excreted 45% less urinary N than did dairy cows fed a 19% CP diet (Misselbrook et al., 2005). To reduce the N content in the diet has been proposed as the most efficient way to reduce the amount of excreta N, whilst maintaining animal production (Ledgard, 2001; de Klein et al., 2005). The premise of this practice is that pasture typically contains an excess of protein relative to animal requirements and supplements with a low protein concentration (e.g. maize silage) can increase efficiency of N utilization (Kebreab et al., 2001). With this practice, low-protein forage such as maize silage is normally used to reduce dietary N concentration so that to increase N use efficiency and reduce environmental N emissions (Luo et al., 2008a). By reducing the protein content of supplement feed, a 20% reduction in estimated N excretion to urine and faeces was achieved (Nielsen et al., 2003). The limited information available supports the efficiency of such management practices on total N₂O emissions from whole farm systems. For example, total N₂O emission from the dairy farm

systems consisted of a maize supplementation system with a stocking rate of 3.8 cows ha⁻¹ of grazed pasture with maize silage brought in was slightly lower than a control system with a stocking rate of 3.0 cows ha⁻¹ of grazed pasture; but the N₂O emission per kg of milk production from the maize supplementation was 22% lower than that from the control system (Luo et al., 2008a).

A controversial option of dietary manipulation is to use hippuric acid, which is a ruminant urine constituent. Ruminants fed with protein rich feeds have been shown to have higher urinary hippuric acid concentrations than ruminants with lower protein diets, so that the concentration of hippuric acid in urine may be changed by manipulating pasture species composition (Clough et al., 2009). Several laboratory-based studies reported significant decline (48-54%) in cumulative N₂O emissions from synthetic urine by hippuric acid addition (van Groenigen et al., 2005; Kool et al., 2006). However, according to a field study, increasing hippuric acid addition in bovine urine had no effect on N₂O mitigation (Clough et al., 2009). Further studies are needed to determine the potential of hippuric acid as an option for N₂O mitigation.

Supplementing a diuretic in the form of salt (NaCl) to a cow's diet has been suggested as a potential mitigation option to reduce N loss as N₂O and nitrate due to increase in the spread of urine, thereby diluting the rate of urinary-N and increasing plant uptake of N (Ledgard et al., 2007). However the results are not consistent between laboratory and field experiments. N₂O emission was reduced only in laboratory study but not in field study (van Groenigen et al., 2006). More assessments are needed for this option.

3.2. Management of synthetic fertilizer N application

Synthetic fertilizer N application in agriculture is one of the important factors leading to the rapid increase of atmospheric N₂O concentration in recent decades (Davidson, 2009). Fertilizer N rate, type, timing, and placement have all been proposed to influence N₂O flux from agricultural soils (Millar et al., 2010) and provide potential for emission abatement (Smith et al., 2007). Nevertheless, only fertilizer N rate is used to calculate national N₂O emissions regardless of fertilizer N type, timing and placement for the IPCC guidelines (IPCC, 2006).

3.2.1 Fertilizer N type

Major fertilizer N types include anhydrous ammonia, ammonium-based fertilizers, ammonium nitrate, calcium ammonium nitrate, nitrate-based fertilizers, ammonium phosphates, urea, urea-ammonium nitrate (Bouwman et al., 2002). Fertilizer N type has been reported to affect soil N₂O emissions (Bouwman et al., 2002; Venterea et al., 2005). N₂O emissions were 2–4 times greater from treatment amended with anhydrous ammonia than from those amended with urea ammonium nitrate and broadcast urea in a cropland study (Venterea et al., 2005). Bouwman et al. (2002) reported that nitrate based fertilizers resulted in significantly lower emissions of N₂O than ammonium based fertilizers based on analysis of a global data set. In another data synthesis, Stehfest and Bouwman (2006) concluded there was no significant difference among most fertilizer types in terms of N₂O emissions. Since the effect of fertilizer type on N₂O emission may be confounded, and since very limited studies have been conducted to compare the responses of N₂O emission to fertilizer type while keeping the same for other factors, more studies are needed before any solid conclusions are drawn.

A new form of fertilizer N type has emerged recently to enhance the efficiency of fertilizer N, i.e., slow-released fertilizers which are defined as a fertilizer that release (convert to a plant-available form) their plant nutrients at a slower rate relative to a “reference soluble” product (Hall, 2005). From the perspective of N₂O mitigation, only the effectiveness of polymer-coated fertilizers (PCFs) have been tested in several studies among the slow-release fertilizers (Akiyama et al., 2010). A comprehensive analysis showed that PCFs significantly reduced N₂O emissions by 14% to 58% with an average of 35% (Akiyama et al., 2010). However, most of the existing studies were conducted in cropland, whether PCFs have similar efficiency in reducing N₂O emissions needed further evaluation.

3.2.2. Timing of fertilization

Synchronous timing of fertilizer N application with plant N demand is an important measure to improve N use efficiency (NUE) and thus reduce N loss to the environment (Crews et al., 2005). However, measures for synchronizing N supply are often aimed to reduce N loss via NH₃ volatilization and nitrate leaching, which in total account for up to 50% of N inputs. Limited data are available to show whether synchrony of nitrogen supply declines soil N₂O emission from grassland. Based on studies in cropland, it seems that improving NUE cannot consistently reduce N₂O emissions (Phillips et al., 2009), probably because the practices that improve NUE by reducing NH₃ and/or NO₃⁻ losses may make more N available in the soil for both N uptake in crops and soil N₂O production (Venterea et al., 2012). In conclusion, current evidence does not support synchronous timing of fertilizer N application is an effective way to decrease soil N₂O emission.

3.2.3. Fertilizer N placement

Very few studies have been conducted to investigate the effect of fertilizer placement on soil N₂O emission for grassland. The studies in cropland show that fertilizer placement affects soil N₂O emission (Liu et al., 2006; Venterea et al., 2008; van Kessel et al., 2013). A meta-analysis shows that placement of fertilizer N (>5 cm depth) can be an effective strategy for mitigating N₂O emissions in no-tillage and reduced tillage systems, especially in humid climatic conditions (van Kessel et al., 2013). The underlying mechanism is that both nitrification and denitrification potential tend to decrease rapidly with depth in no-tillage soils, and hence deep N placement may simply decrease the supply of inorganic N substrates within the most biologically active zone where they can be converted to N₂O via nitrification and/or denitrification (van Kessel et al., 2013). However, whether this practice is effective in grassland needs further investigation.

3.2.4. Fertilizer N rate

Among the many practices of fertilizer N management, fertilizer N rate is likely most straightforward in affecting soil N₂O emission. Reducing the N rate is perhaps the only practice that has broad consensus as being generally reliable for reducing N₂O emissions (Venterea et al., 2012). The Intergovernmental Panel on Climate Change (IPCC) suggests a default linear emission factor of 1% for fertilizer N application, regardless of the rate of application (IPCC, 2006). For the few N₂O response experiments in which more than two levels of N were applied, N₂O flux in response to increasing N rates has been described by both linear (Halvorson et al., 2008) and nonlinear functions (Bouwman et al., 2002; McSwiney et al., 2005; Kim et al., 2010; Hoben et al., 2011; Rafique et al., 2011). These studies suggested that N₂O fluxes were found to increase exponentially with increasing N rate for both grassland and cropland when multiple N rates were used. N₂O emissions changed slightly when fertilizer N inputs were below a certain level, but increased

rapidly above that level, or high rates of N fertilization led to increasing rates of N₂O loss without economic gains in yield. For example, in a study of cropland, N₂O fluxes were moderately low (ca. 20 g N₂O-N ha⁻¹ day⁻¹) at levels of N rates up to 101 kg N ha⁻¹, where grain yields were maximized, after which fluxes more than doubled (to >50 g N₂O-N ha⁻¹ day⁻¹) (McSwiney et al., 2005). Similarly, in another study with multiple sites and N rates, the two N fertilizer rates above those recommended for maximum economic return (135 kg N ha⁻¹), average N₂O fluxes were 43% (18 g N₂O-N ha⁻¹ day⁻¹) and 115% (26 g N₂O-N ha⁻¹ day⁻¹) higher than were fluxes at the recommended rate, respectively, but the corn grain yield only increased by 2% and 6%, respectively (Hoben et al., 2011). Although these examples were for cropland, the responses of N₂O emission to N rates for grassland are most likely similar. Overall, the above examples highlight the potential to lower agricultural N₂O fluxes with no or little yield penalty by reducing N fertilizer inputs to levels that just satisfy crop needs.

3.3. Manure management

Globally, manure production and use contribute more N₂O to the atmosphere than synthetic fertilizer N (Davidson, 2009). The input of synthetic fertilizers on farms can be minimized, in part, by manure recycle within the farm (Fig. 1), i.e., by keeping a lower farm-scale N balance (or farm-gate N surplus), the difference between whole farm N inputs and outputs (Schröder et al., 2003). In general, a lower farm-gate surplus will result in lower losses of N₂O from a farm. For example, Schils *et al.* (2006) reported that a decrease in surplus N of 1 g kg⁻¹ milk lowered GHG emissions by approximately 29 g CO₂-eq kg⁻¹ milk. Similarly, Olesen *et al.* (2006) reported that GHG

emissions were found to decrease linearly with increasing farm N efficiency (-16.5 kg CO₂-eq MJ⁻¹ per % increase in efficiency).

N₂O and other greenhouse gases can be produced and emitted at each stage of the 'manure management continuum', i.e., the livestock building, manure storage, manure treatment and manure spreading to land (Chadwick et al., 2011). Although all N losses represent potential N₂O emissions, we will focus on direct N₂O emission from the 'manure management continuum'. Other N losses and GHG emission related to manure management are referred to some comprehensive reviews (Rotz, 2004; Webb et al., 2010; Chadwick et al., 2011; Novak et al., 2011).

3.3.1. N₂O emissions during manure collection stage

Manure is normally collected from animal houses and standing yards. Few studies have been conducted to measure N₂O emissions from hard standing areas. The limited evidence shows very low or no N₂O emissions at these areas due to predominantly anaerobic conditions (Ellis et al., 2001; Misselbrook et al., 2001).

N₂O emissions from animal houses depend on whether bedding materials are used. The slurry/faeces/urine remains in a predominantly anaerobic state with little opportunity for the NH₄⁺ to be nitrified, therefore little or no N₂O emissions are likely to occur from such systems (Zhang et al., 2005). By comparing N₂O and NH₃ emissions from cattle and pigs housed in slurry-based and straw bedded buildings, Thorman et al. (2003) reported that there were little or no N₂O emissions from slurry-based cattle or pig buildings, whilst there were N₂O emissions from cattle housed with straw bedding (4-5 mg N m⁻² d⁻¹). Much higher emissions may occur from deep litter systems, especially mechanical mixing of deep litter is involved (Groenestein et al., 1996). However, uncertainties remain. For example, Amon et al. (2001) reported there was no significant difference

between slurry-based and straw-based stall systems for dairy cows in N₂O emissions.

3.3.2 N₂O emissions during manure storage

There is evidence showing that N₂O and other GHG emissions during manure storage stage are greater than emissions after manure application (Amon et al., 2006; Clemens et al., 2006). Manure can either be stored as slurry (liquid) in lagoons or aboveground tanks or stored as solid manure in heaps, but manure is normally stored as slurry for intensive systems. For example, in Europe, livestock slurry is stored in 20–40 m diameter storage tanks with or without coverage, or in lagoons; in the United States and Canada, slurry is handled in stores or alternatively the slurry is diluted with water to encourage decomposition of the solids and pumped into anaerobic lagoons (Petersen et al., 2006). N₂O emission from slurry/liquid manure is normally lower than from solid manure during storage (Sommer et al., 2000; Berg et al., 2006; IPCC, 2006). For example, the IPCC default emission factors for slurry/liquid manure and solid manure are 0.1% and 2%, respectively (IPCC, 2006). However, the potential for methane emission from liquid manure is larger than from solid manure. For example, the emission factors proposed in the IPCC methodology for conversion of digestible organic carbon to methane are 1–2% for solid storage, 39–72% for slurry storage or extended pit storage, and up to 100% for manure stored in anaerobic lagoons (IPCC, 2000).

3.3.2.1 Slurry storage and treatment

N₂O emission from slurry stored without surface cover is very low due to limited transformation of ammonium to nitrate under anaerobic conditions so that the subsequent denitrification is also very low. Stored slurry with a surface layer of straw or natural crust may be a

source of N₂O emissions (Sommer et al., 2000; Berg et al., 2006; VanderZaag et al., 2009). This is because the presence of a surface crust can provide aerobic conditions in the crust and hence nitrification can occur with N₂O generation (Sommer et al., 2000). Manipulating (via addition of inorganic or organic acid) the balance between ammonia and ammonium by lowering the pH value of slurry below 6.0 is an effective measure to reduce emissions of N₂O and CH₄ and NH₃ as well (Berg et al., 2006).

Several slurry treatment options, including slurry dilution, slurry additives, slurry aeration, slurry separation and anaerobic digestion are usually used to reduce slurry dry matter concentration in order to insure evenly distribution of nutrient after application (Amon et al., 2006). Although NH₃ emissions may be lowered from the treated slurry, their efficiency in decreasing N₂O emission has not been well investigated. Amon et al. (2006) compared N₂O emissions from slurry which was treated with different methods (Table 1). N₂O emissions were increased by separation, digestion, straw cover and aeration (Table 1). Fangueiro et al. (2008) studied N₂O emission from five cattle slurry fractions with distinct characteristics obtained using a combined separation process (screw press + chemically enhanced settling using polyacrylamide (PAM)). In contrast, significant N₂O emissions were observed only in the untreated slurry and liquid fractions of the separated slurry. Separation of slurry produces a solid fraction that is stored in manure heaps before being used as a fertilizer in crop production (Hansen et al., 2006).

Biogas treatment of animal manures is an upcoming technology because it is a way of producing renewable energy (biogas). However, little is known about effects of this management strategy on GHG emissions during fermentation (anaerobic digestion), storage, and field application of the substrates compared to untreated slurries (Clemens et al., 2006). The results are not consistent in terms of whether anaerobic digestion can reduce N₂O emission during slurry storage or

application. In a study, N₂O emission from storage of anaerobically digested slurry was lower in winter but higher in summer relative to untreated slurry; and there was no difference between slurry types after application in their N₂O emissions (Clemens et al., 2006). Another study reported higher N₂O emission from anaerobically digested slurry (Amon et al., 2006).

Biological aerobic treatment is widely and is regarded as a necessary way to remove N from the slurry as N₂ in order to protect water courses (Beline et al., 1999). The treatment results in a concentrated sludge (15% of original volume) suitable for land spreading and an effluent (85% of original volume) that can be used as irrigation or discharged to the sewer system (Willers et al., 1996; Loyon et al., 2007). In addition to N₂, higher emission rates of N₂O (up to 13%) and NH₃ (up to 40% of total N) may also be emitted from aerobically treated slurry dependent on the aeration rate and temperature (Willers et al., 1996; Beline et al., 1999). In contrast, Loyon et al. (2007) reported that aerobic treatment (using an intermittent aeration) decreased NH₃ emission by 30-68% and N₂O emission by 55% relative to untreated manure.

3.3.2.2 Solid manure storage and treatment

Solid manure heaps may be significant sources of N₂O emissions (Chadwick et al., 1999). Maintaining anaerobic manure conditions can significantly reduce N₂O emissions from solid manure storage heaps (Chadwick et al., 2011). For example, emissions of NH₃, N₂O, and CH₄ were reduced by 12%, 99%, and 88%, respectively, when the manure heap was covered with an airtight material relative to the uncovered heap (Hansen et al., 2006). N₂O losses were lower from the composted farmyard manure than from the anaerobically stacked manure (Amon et al., 2001). Compaction of manure in storage may also reduce N₂O emissions by creating anaerobic conditions thus inhibiting nitrification. The inhibition of nitrification will prevent the formation of NO₃⁻ and

hence there will be no losses of N₂O via denitrification (Chadwick, 2005). Chadwick (2005) observed that compaction and covering also significantly reduced N₂O emissions from cattle manure by ca. 30%. However, the compaction may increase the potential for CH₄ emissions (Chadwick, 2005).

Straw is often used as additive during solid manure storage. The effect of straw addition in lowering N₂O emissions is not consistent (Brown et al., 2000; Sommer et al., 2000; Yamulki, 2006). Yamulki (2006) showed that straw addition to farm yard manure reduced the mean cumulative N₂O emission by ca. 40% relative to conventional manure heaps with the emissions of N₂O accounting for up to 0.3% and 0.7% of the total initial N in the straw-used and conventional heaps, respectively. In addition, straw addition reduced CH₄ emissions (Yamulki, 2006). In contrast, N₂O emissions increased by 2 times from solid dairy manure amended with chopped straw to water contents of 70, 75, and 80% relative to the unamended samples (Brown et al., 2000).

3.3.3 Manure application

N₂O emissions following manure application to grassland have been reported by many studies (Chadwick et al., 2000; Velthof et al., 2003; Fangueiro et al., 2008; van der Meer, 2008; Luo et al., 2010; Chadwick et al., 2011). These studies indicated that N₂O emissions from manure application may also be affected by manure type, timing, rates or placements of application. The efficiency of timing and rates in mitigating N₂O emissions following manure application should be similar to fertilizer N application.

The effects of manure type on N₂O emissions are not consistent and dependent on soil conditions, N and C content in manure, and application period (Chadwick et al., 2011). Clemens et al. (2006) reported that there was no difference between slurry types after application in their N₂O

emissions. Chadwick et al. (2000) compared N₂O emissions following application of pig slurry and dairy cow slurry at equal volume in April, July and October. They found that cumulative N₂O emissions were greater following application of dairy cow slurry than pig slurry in April, but significantly greater N₂O emissions resulted from pig slurry-treated plots than dairy cow slurry-treated plots in July and October. These results indicate that manure type may not be regarded as a major factor affecting N₂O emissions following manure application.

Solid manures can only be broadcast on the surface of grassland (Chadwick et al., 2011), but there are four main types of slurry application systems (Misselbrook et al., 2002), i.e., (a) surface broadcast - uniform covering of slurry across the crop; (b) band spreading - slurry placed in discrete bands on the crop by trailing hoses; (c) trailing shoe - slurry placed in discrete bands on the soil surface below the crop canopy; (d) injection - slurry placed in shallow, open slots or deep, closed slots within the soil. These manure application techniques were originally developed to reduce N losses via NH₃ (Webb et al., 2010). Their efficiency in reducing emissions of N₂O and other GHG emissions have not been well studied. According to a recent review (Webb et al., 2010), reduced-NH₃ application techniques may increase emissions of N₂O, while placement of effluent below the soil surface by injection or aeration resulted in elevated CH₄ emissions (Sistani et al., 2010). Therefore, the direct and indirect emissions of N₂O and other GHG should be assessed. In addition, the results are not consistent even in the same study. For example, Sistani et al. (2010) compared N₂O and other GHG emissions from a control, an inorganic fertilizer treatment receiving 179 kg N ha⁻¹ as urea-NH₄NO₃ (UAN), and three swine effluent application methods (surface application, direct injection, and application in combination with soil aeration) that received a target rate of 200 kg N ha⁻¹ in two consecutive years. In year one, N₂O emissions were similar for the UAN, surface effluent, and aeration effluent treatments (with an average of 0.72 g N₂O m⁻²) but

higher than the injection treatment ($0.47 \text{ g N}_2\text{O m}^{-2}$). However the pattern was reversed in year two, with the injection treatment emitting $0.82 \text{ g N}_2\text{O m}^{-2}$ and the remaining N source treatments emitting an average of $0.36 \text{ g N}_2\text{O m}^{-2}$. These results demonstrate that climatic conditions and application method need consideration when evaluating the impact of liquid manure management on GHG emissions (Sistani et al., 2010).

3.4 Use of nitrification inhibitors to lower N_2O emission from urine patch, application of synthetic fertilizer or manure

Nitrification inhibitors (NIs) are compounds that delay the bacterial oxidation of ammonium to nitrite by depressing the activity of *Nitrosomonas* bacteria in the soil (Macadam et al., 2003; Watson et al., 2009). The most extensively studied NIs are dicyandiamide (DCD), 3,4-dimethyl pyrazole phosphate (DMPP) and nitrapyrin (Watson et al., 2009). As nitrate is the substrate for denitrification, the use of NIs along with ammonium based fertilizers, have the potential to reduce N_2O emissions from both nitrification and denitrification.

A series of studies have shown that both N_2O emissions and NO_3^- leaching from urine patches and application of fertilizer or manure can be significantly mitigated by treating grazed pasture soil with NIs including DCD and DMPP (Di et al., 2008; Qiu et al., 2010; de Klein et al., 2011; Di et al., 2012). In a study covering four different soils under different climatic and management conditions, N_2O emissions from urine patches were declined by 61-73% with an average of 70% (Di et al., 2007). Recent studies in Ireland have shown that DCD can reduce N_2O emissions by between 49 to 70% from urine patches (Dennis et al., 2008; Selbie et al., 2010) and from slurry applied to grassland soils (Cahalan et al., 2010). In addition, DCD could reduce NO_3^- leaching from urine

patches by 68-76% (Di et al., 2004; Di et al., 2005). Dennis *et al.* (2010) reported that nitrate leaching from urine patches was reduced by up to 45% through the use of DCD on free draining soils in Ireland. In New Zealand, Monaghan *et al.* (2009) showed that application of granular DCD lowered NO_3^- leaching by 21 to 56% from hydrologically isolated dairy pastures over a 4-year study. Ledgard *et al.* (2008) tested a novel approach to supplement animals with the NIs during the main N loss period in such a way that the NIs would be excreted by the animals, principally in the urine. Their study highlights the potential for using direct administration of N process inhibitors to grazing animals to reduce environmental N emissions from urine patches in pasture systems. However, despite the high recovery of DCD in urine (>85%), there is a potential of metabolic residues of the inhibitors in the animals. It is still not clear whether the metabolic residues in animal products are detrimental to human health.

Akiyama *et al.* (2010) evaluated the effectiveness of NIs on N_2O emissions using meta-analysis, and found that (i) NIs significantly reduced N_2O emissions (mean: -38%, 95% confidence interval: -44% to -31%) compared with those of conventional fertilizers and their effectiveness was relatively consistent across the various types of inhibitors and land uses; (ii) NIs were effective in reducing N_2O emission from both synthetic fertilizer N and organic fertilizers; and (iii) the efficacy was higher for grassland (-54%, -60% to -43%) than for upland fields (-34%, -43% to -26%) and paddy fields (-30%, -39% to -18%). In 2008, DCD was reported to have decreased New Zealand's N_2O emissions by 40.8 Gg CO_2 -eq, a 0.1% decrease in total agricultural N_2O emissions (Anon, 2010). Due to the effectiveness of NIs in the decrease of both direct and indirect N_2O emissions in New Zealand, they were considered an effective measure to lower N_2O emissions from agricultural soils by the IPCC Fourth Assessment Report (IPCC, 2007).

Despite the effectiveness in lowering N_2O emissions, some side effects have been reported for

NIs. Use of NIs extended retention of N in the NH_4^+ form in the soils, and hence they have the potential to increase NH_3 volatilization, despite some studies reported DCD reduced NH_3 volatilization by 28-38% from urine patches (Zaman et al., 2012) A meta-analysis reveals that there is significance increase in NH_3 loss (by 0.3-25.0%) in both pasture and cropping soils after NIs application, especially for the soils with higher pH and lower cation exchange capacity (Kim et al., 2012).

There are some additional costs associated with using NIs. Subbarao et al. (2006) estimated that using nitrapyrin or DCD added about 25 to 30% to the cost of fertilizer N. Hence, Monaghan *et al.* (2009) suggested that in the absence of substantial herbage yield benefits, the cost of using inhibitors such as DCD was not economical unless farmers received carbon credit payments for the reduction in N_2O emissions. Under current circumstances the cost of NIs is the main factor limiting their widespread use (Subbarao et al., 2006). For NIs to be more widely adopted, they will need to be competitively priced.

3.5 Use of biological N fixation in association with forage legumes as an alternative to N fertilizer

3.5.1 N supply by biological N fixation

Biological N fixation (BNF) in association with forage legumes provides an alternative N source for grasslands. White clover is the main legume in pastures and meadows of temperate regions and is adapted to survive in a range of grassland environments, while other legumes are of less importance (Rochon et al., 2004). Most often, white clover is grown with companion grasses.

In pasture-based farming systems in Australia and New Zealand, N₂ fixation by white clover was the main source of N input (Ledgard et al., 2001). Average biological N₂ fixation in grazed permanent clover/grass pastures in temperate regions of the world has been reported to be 80 to 100 kg N ha⁻¹ yr⁻¹ (range 10 to 270 kg N ha⁻¹yr⁻¹) (Davies et al., 1996; Ledgard, 2001). This fixed N becomes available slowly over time to the grass in pastures after it is released into soil *via* exudates from living legume roots, by mineralization of senesced legume tissues and in excreta after consumption by grazing animals (Ledgard et al., 2009).

Rising costs of fertilizer N and environmental regulations governing stocking densities and fertilizer N use on farms is increasing interest in the use of white clover in grassland. In a recent review, Andrews et al. (2007) concluded that herbage and milk production from white clover-based pastures (perennial ryegrass with 20% white clover in herbage DM on an annual basis) are likely to be similar to that from a perennial ryegrass pasture receiving annual input of 200 kg ha⁻¹ of fertilizer N and around 70% of that obtained with perennial ryegrass receiving annual input of 350 to 400 kg ha⁻¹ of fertilizer N. In many countries in the northwest of Europe, these very high rates of fertilizer N input are no longer permissible due to regulations associated with the Nitrates Directive (European-Council, 1991). For example, in Ireland under Statutory Instruments (SI) No. 101 (Anon, 2009) the stocking density on dairy farms is limited to 2 dairy cows per hectare and, under these circumstances fertilizer N input should not exceed approximately 200 kg ha⁻¹. Subject to certain conditions farmers can seek derogation from these limits up to a maximum permissible fertilizer N input of approximately 279 kg ha⁻¹ (Anon, 2009). Similar or lower maximum allowable rates pertain in Northern Ireland, the Netherlands, Denmark and Germany (Humphreys, 2008).

3.5.2 N₂O emissions from legume based grasslands

Although some early studies show that several *Rhizobium* species, in the free-living form, in legume root nodules or as isolated bacteroids can denitrify nitrate and release N₂O (Rochette et al., 2005), recent studies suggest that direct N₂O emission from BNF *per se* is negligible (Rochette et al., 2005; Carter et al., 2006; Zhong et al., 2009). The N₂O emissions induced by the growth of legume crops/forages may be estimated solely as a function of the above-ground and below-ground N inputs from crop residues (Rochette et al., 2005; Carter et al., 2006; IPCC, 2006; Li et al., 2011). Accordingly N₂O emissions from legume-based grasslands are much lower than fertilized grasslands. For example, Some study reported up to 5-fold of more N₂O emission from heavily N fertilized grasslands than from their legume-based counterparts (Ruzjerez et al., 1994). A data synthesis indicates that the average soil N₂O emissions from field-grown legumes, N fertilized grass pastures and crops, and unfertilized soils are 1.29, 3.22 and 1.20 kg N ha⁻¹ yr⁻¹, respectively (Jensen et al., 2012). Therefore N₂O emissions from grass legume mixes are only slightly greater than background emissions (Rochette et al., 2005; Jensen et al., 2012). However soil N₂O emissions for legume-based pastures grazed by animals will be significantly increased due to excreta patches. For example, soil N₂O emissions from white clover/ryegrass systems receiving no fertilizer N or 58 kg fertilizer-N ha⁻¹ yr⁻¹ were found to be only 16-19% lower relative to a grass/fertilizer system (226 kg N ha⁻¹ yr⁻¹) (Li et al., 2011).

Furthermore, N₂O emissions may be indirectly lowered by legume-based systems. It allows dairy farmers to achieve the same net income at lower stocking densities compared with a higher stocked system reliant on high inputs of fertilizer N (Table 2). A lower stocking density means less N cycling within the system and lower urine deposition, which is a major source of N₂O emission as aforementioned. Although limited studies have conducted to compare nitrate leaching or NH₃

volatilization in legume and fertilizer-based systems, the current evidence suggests that N losses via these two pathways may be lowered in legume-based systems (Crews et al., 2004). Therefore indirect N₂O emission from these N losses can be reduced. A study shows that nitrate leaching was reduced by 48% to 76% when fertilizer N was replaced by alfalfa (*Medicago sativa*) (Owens et al., 1994). Similarly, NH₃ volatilization from legume residues was found to be much lower (Larsson et al., 1998).

Total GHG emissions have been reported to be substantially lowered by legume-based systems. An LCA analysis shows that GHG emissions from white clover-based dairy production systems (80-99 kg N ha⁻¹ yr⁻¹ with stocking densities of 1.8-2.2) are 11% to 23% lower than fertilizer-based systems (180-353 kg N ha⁻¹ yr⁻¹ with stocking densities of 2.0-2.5) in Ireland (Yan et al., 2013) Similarly, GHG emissions per kg of milk from a grass/fertilizer system (160 kg N ha⁻¹) were found to be 12% higher than from a clover/ryegrass system receiving no fertilizer N in New Zealand (Ledgard et al., 2009). Schils *et al.* (2005) used a farm level approach to compare GHG emissions from grass/fertilizer and grass/clover farms. Compared to a grass/fertilizer-N system, the GHG emissions (excluding carbon sequestration) from a grass/clover farm were 23% lower per ha and 11% lower per kg milk.

There is substantial economic benefit to replacing synthetic fertilizer N by BFN in grasslands (Rochon et al., 2004; Andrews et al., 2007; Humphreys et al., 2012). It is likely that fertilizer N costs will continue to increase relative to farm gate product prices (Humphreys et al., 2012). Under these circumstances the economics of using white clover become increasingly favourable, offering substantial potential to lower GHG emissions, if its use were to become widespread on farms.

3.6 Combined options to mitigate N₂O emission

Efficiencies of each option in lowering N₂O emissions are presented in Table 3. At each stage of farm N flow, there are options to lower N₂O emissions. Although single option of N management has the potential to lower N₂O emission, the mitigation capacity is relatively small. In some cases, the decrease of N₂O emission in one step of the N flow (Figure 1) may be offset by increased N₂O losses in another step. For example, restricted grazing by keeping animals off grazing paddocks and on stand-off/feed pads or in animal houses can efficiently lower N₂O emission from the grazing paddocks due to excreta deposition. However, this decrease may be offset if manure is not effectively managed during in-house or storage stage. In other words, effective measures should be taken at each step during N flow or combined options should be used in order to mitigate N₂O emission at the farm level.

So far few studies have been conducted to investigate the effectiveness of combined options in lowering N₂O emission. Nevertheless, according to the limited available evidence, the effectiveness of the combined options to mitigate N₂O emission may not be simply additive of individual options. For example, N₂O emission was lowered by 39, 41 and 55% from restricted grazing, DCD application and their combination in a pasture of New Zealand (Luo et al., 2010). Quantitative analysis of N flows and N₂O losses in dairy farming systems reveal that the implementation of a package of measures to improve N management may reduce N₂O emissions from dairy farming systems by up to 70% (Velthof et al., 1997). Vallejo et al. (2005) showed that addition of dicyandiamide reduced N₂O emissions from 2.95% to 0.50% of applied N when injected into soil in a Mediterranean climate. The above studies show that the combined options were more effective in mitigating N₂O emission than individual option but the decrease in N₂O emission by the combined options was lower than the total decrease of N₂O emission by the individual options. More studies are undoubtedly needed to investigate the effectiveness of combined options in

mitigate N₂O emission.

4. Conclusions

The efficiencies of major N management options in mitigating N₂O emissions from grassland-based agriculture are reviewed:

- (1) Restricted grazing by reducing grazing time is an effective way to decrease N₂O emissions from excreta patches. Balancing the protein-to-energy ratios in the diets of ruminants can also decrease N₂O emissions from excreta patches.
- (2) Among the managements of synthetic fertilizer N application, only adjusting fertilizer N rate and use of slow-released fertilizers are proven to be effective in lowering N₂O emissions from grasslands.
- (3) Use of bedding materials may increase N₂O emissions from animal houses. Manure storage as slurry, manipulating slurry pH to values lower than 6 and storage as solid manure under anaerobic conditions help to reduce N₂O emissions during manure storage stage. For manure land application, N₂O emissions can be mitigated after reducing manure N inputs to levels that just satisfy crop needs.
- (4) Nitrification inhibitors have been shown to substantially lower N₂O emissions associated with applications of fertilizers and manures and from urine deposited during grazing.
- (5) N₂O emissions from legume based grasslands are generally lower than fertilizer-based systems. In addition, legume-based systems can be as profitable as conventional fertilizer-N-based systems.

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Table 1. Net total N₂O emissions during storage and after field application of differently treated dairy cattle slurry (given in g N₂O per m³ slurry) (Amon et al., 2006)

	Untreated	Separated	Digested	Straw cover	Aerated
Storage	20.2 ^a	9.0 ^b	28.5 ^a	42.2 [*]	49.3 [*]
Storage-solid fraction		13.2			
Field application	3.8 ^a	6.4 ^b	2.7 ^a	10.3 ^c	4.9 ^b
Total emission	24.0	28.6	31.2	52.5	54.2
% Storage	84.2	77.6	91.3	80.4	91.0
% Application	15.8	22.4	8.7	19.6	9.0

Different superscripts indicate significant differences at $p < 0.05$ (separately for emissions during storage and after field application).

*No statistical comparison with untreated slurry was possible as the shape of the cumulated emissions curve was different from untreated slurry and the t-test for differences in regression parameters could not be applied.

Table 2. The economic performance of systems of dairy production based on N-fertilized grass (FN) and grass-white clover (WC) grassland including sales, variable and fixed costs, and gross and net margin per ha (Humphreys et al., 2012).

System	FN	WC	P value
Stocking density (LU ha ⁻¹)	2.8	2.04	<0.05
Fertilizer N (kg ha ⁻¹)	246	90	<0.001
Milk sales (€ ha ⁻¹)	3168	2875	<0.05
Total sales (€ ha ⁻¹)	3530	3205	<0.05
Fertilizer N (€ ha ⁻¹)	223	75	<0.001
Concentrate (€ ha ⁻¹)	312	275	NS
Contractor charges (€ ha ⁻¹)	299	253	<0.01
Total variable costs (€ ha ⁻¹)	1400	1146	<0.01
Gross margin (€ ha ⁻¹)	2131	2058	NS
Fixed costs (€ ha ⁻¹)	860	781	<0.05
Net margin (€ ha ⁻¹)	1271	1278	NS

Table 3. Efficiency of farm N management options in lowering N₂O emissions.

Options	Efficiency	Comments	Refs
Excreta patch management			
Reduced grazing animal	+	Economically not acceptable	Humphreys 2008
Reduced grazing time	+	Depend on grazing time	de Klein et al., 2006; Luo et al., 2008b
Dietary manipulation with low-protein forage	+		Luo et al., 2008a; Nielsen et al., 2003
Dietary manipulation with salt	?	Not supported by field study	Van Groenigen et al., 2005; Kool et al., 2006; Clough et al., 2009
Dietary manipulation with hippuric acid	?	Not supported by field study	Ledgard et al., 2007; van Groenigen et al., 2006
Apply with NIs	+		Di et al., 2007; Dennis et al., 2008; Selbie et al., 2010
Fertilizer N management			
Fertilizer N type - PCFs	+		Akiyama et al., 2010
Fertilizer N type - others	?	Depending on timing and site conditions	Bouwman et al., 2002; Stehfest and Bouwman, 2006; Venterea et al., 2005
Timing of fertilization	?		Phillips et al., 2009; Venterea et al., 2012
Fertilizer N placement	?	Effective in cropland, but needs test in grassland	Liu et al., 2006; Venterea et al., 2008; van kessel et al., 2013
Fertilizer N rate	+	N ₂ O emissions can be mitigated after reducing N fertilizer inputs to levels that just satisfy crop needs	Bouwman et al., 2002; Halvorson et al., 2008; Hoben et al., 2011; McSwiney et al., 2005; Rafique et al., 2011
Apply with NIs	+	Need additional cost	Akiyama et al., 2010
Manure management			
Use of bedding materials	-		Groenestein et al., 1996; Thorman et al., 2003; Zhang et al., 2005
Storage as slurry	+	Need more space	Sommer et al., 2000; Berg et al., 2006; IPCC, 2006
Slurry storage - cover	-		Sommer et al., 2000; Berg et al., 2006; VanderZaag et al., 2009
Slurry storage - lower pH	+		Berg et al., 2006
Slurry storage - treatment	?		Amon et al., 2006; Beline et al., 1999; Fangueiro et al., 2008; Clemens et al., 2006; Loyon et al., 2007; Willers et al., 1996

Solid manure - anaerobic conditions	+		Chadwick et al., 2011; Chadwick et al., 2005; Hansen et al., 2006
Solid manure - Straw addition	?		Brown et al., 2000; Sommer et al., 2000; Yamulki, 2006
Application - manure type	?		Clemens et al., 2006; Chadwick et al., 2000
Application - rate	+	Similar to fertilizer N	
Application - timing	?	Similar to fertilizer N	
Application - methods	?		Webb et al., 2010; Sistani et al., 2010; Chadwick et al., 2011
Application - with NIs	+	Need additional cost	Akiyama et al., 2010
Use of biological N fixation			
BNF	+		Li et al., 2011; Ruzjerez et al., 1994; Rochette et al., 2005; Jensen et al., 2012; Schils et al., 2005; Yan et al., 2013

* PCFs denotes polymer-coated fertilizers

** “+”, “-” and “?” correspond positive, negative, and positive/negative efficiencies in lowering N₂O emissions, respectively.

Figure legends:

Fig. 1. Nitrogen cycling in grassland based systems showing N₂O production. Open arrows represent soil N cycling processes (nitrification (1), denitrification (2) and nitrifier denitrification (3)). Solid arrows denote the relative size and direction of the N flows. Percentages indicate the estimated transfer of N from one compartment to the other compartment (modified from Oenema et al. (2005) and Wrage et al. (2001)). N₂O production in soil also applies to the manure environment. N losses other than N₂O are not shown. BNF — Biological N fixation.

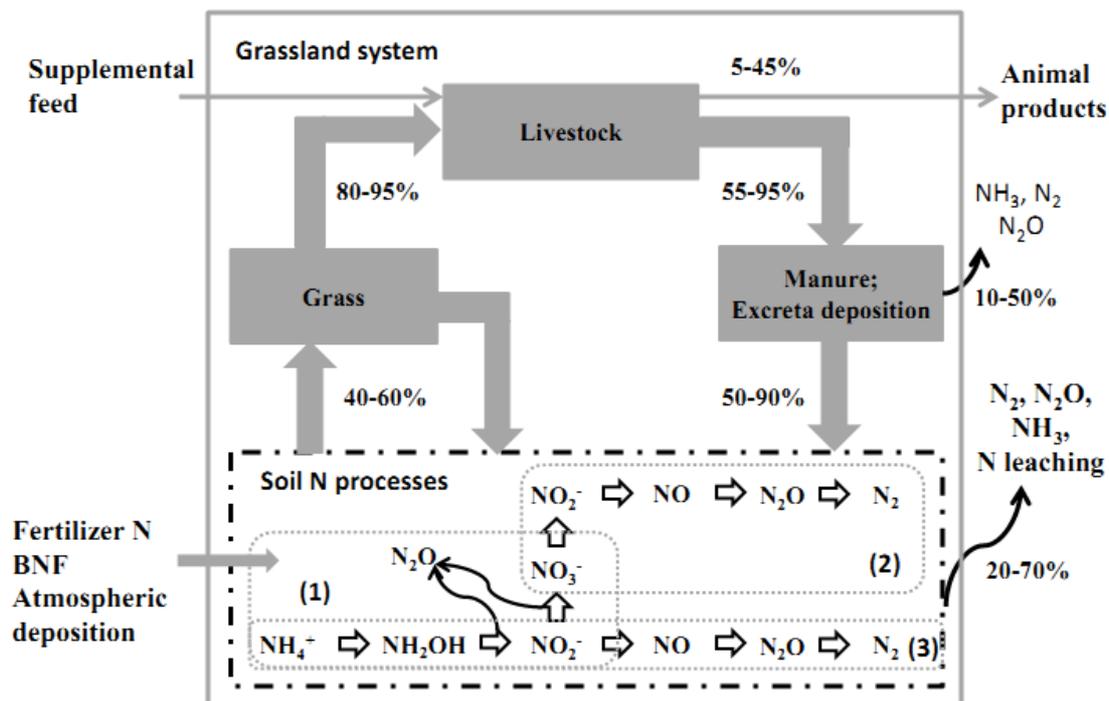


Fig. 1. Nitrogen cycling in grassland based systems showing N_2O production. Open arrows represent soil N cycling processes (nitrification (1), denitrification (2) and nitrifier denitrification (3)). Solid arrows denote the relative size and direction of the N flows. Percentages indicate the estimated transfer of N from one compartment to the other compartment (modified from Oenema et al. (2005) and Wrage et al. (2001)). N_2O production in soil also applies to the manure environment. N losses other than N_2O are not shown. BNF — Biological N fixation.