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The Continuing Challenge of Nitrogen Loss to the Environment: Environmental Consequences and Mitigation Strategies

Christine H. Stark[1] • Karl G. Richards[2*]

[1] Department of Environmental & Geographical Sciences, Manchester Metropolitan University, Chester St, Manchester, M1 5GD, Great Britain
[2] Teagasc, Environmental Research Centre, Johnstown Castle, Wexford, Ireland

Corresponding author: * **karl.richards@teagasc.ie**

ABSTRACT

In this second part of our review on nitrogen loss to the environment, we discuss the increasing problem of air and water contamination with nitrogenous compounds with a focus on agricultural contributions, water quality standards and legislation and mitigation options available to land managers. Excess nitrogen in soil, aquatic and atmospheric environments is a growing global problem due to human activity increasingly dominating nutrient cycling. While sources of environmental nitrogen pollution are diverse, considerable concern exists regarding the impact of intensified agricultural systems and the effects of increasing rates of fertilisation on water and air quality, which negatively affect human and animal health, increase climate change, ozone depletion and atmospheric nitrogen deposition, and cause the deterioration of aquatic habitats. Over recent decades, regional, national and international legislation has been introduced to control and mitigate nitrogen loss from agricultural sources. While the effects of nitrogen on human and animal health have resulted in the introduction of limits that ensure drinking water quality, the role of nitrogen in aquatic eutrophication has greatly been underestimated in the past and the implementation of new standards and laws has been recommended. In addition, a wide range of mitigation measures, mostly practical farm management solutions, is available to agriculture to reduce nitrogen loss to the biosphere. The involvement of land managers in the decision making and implementation processes is, however, crucial to guarantee the success of measures that optimise agricultural production, minimise adverse effects on human and animal health and reduce environmental pollution. Profitable, environmentally sound and socially acceptable agricultural production has to be achieved through a joint approach by researchers, land managers and policy makers aimed at cost effective, targeted and innovative implementation of mitigation measures to reduce environmental N emissions.

Keywords: agricultural N emissions to water and air, health effects, eutrophication, N cycle, environmental quality legislation, mitigation options

Abbreviations: CWA, US Clean Water Act; **N_2**, di-nitrogen; **N_2O**, nitrous oxide; **NH_3**, ammonia; **NH_4^+**, ammonium; **NO**, nitric oxide; **NO_2**, nitrogen dioxide; **NO_2^-**, nitrite; **NO_3^-**, nitrate; **NO_x**, nitrogen oxides (NO + NO_2); **WFD**, EU Water Framework Directive

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INTRODUCTION

Today, a large proportion of human managed land and water use is dedicated to agriculture (FAO 2006) and the rise in global fertiliser production and use over the last century has enabled agricultural production to grow and satisfy ever-increasing demands for food, fibre and energy of a growing population. Producing higher yields per unit area has been accompanied by a significant intensification of production systems, which has created new problems for both the environment and society, ranging from soil degradation and erosion to loss of biodiversity and contamination of drinking water. In particular, the effects of nitrogen (N) in the environment are a cause for global concern.

Crop growth and development largely depend on N availability and N fertilisation greatly increases yields and changes crop quality. Crop uptake is therefore seen as the primary aim of fertiliser, and specifically N, management on farms (cf. Stark and Richards 2008). The greatest fertiliser use efficiency and the lowest risk of loss occur when adequate N is applied at the appropriate time, i.e. during crop growth, and rate. Since there is a fine line between fertiliser optimum and over-supply in terms of timing and rates, N fertilisation can quickly cause serious environmental problems. All excess N is prone to be lost from the soil system by leaching to groundwater, run-off to surface waters

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or in gaseous form to the atmosphere. Although most agricultural N inputs serve human needs, there are concerns about the risks posed to the environment, and human and animal health, as excess N can hold biological and ecotoxicological hazards (Jarvis *et al*. 1995). These include accumulation of toxic compounds in soils, e.g. ammonium (NH_4^+) or nitrite (NO_2^-), which negatively affect plant growth; excess nitrate (NO_3^-) and organic N compounds in waterways, which lead to eutrophication; accumulation of NO_3^- and NO_2^- in human and animal food; and the contribution to global warming and stratospheric ozone depletion through, for example, nitrous oxide (N_2O) (Stevenson and Cole 1999; Addiscott 2005).

It is vital to recognise that the production of mineral fertilisers itself has a substantial impact on the global climate. The Haber-Bosch process, used to synthesise N fertilisers, uses considerable amounts of methane (CH_4) or light petroleum fractions as its hydrogen source. The process is carried out at pressures of 250 atmospheres and 450-500°C, which requires considerable amounts of energy, currently obtained from fossil fuels, resulting in 1% of global energy supply being consumed by N fertiliser manufacture (Smith 2002). Both the production and transport of the final product give rise to emissions of CO_2 (carbon dioxide), N_2O and/or CH_4, which contribute to environmental problems, both globally (e.g. green house effect) and locally (e.g. acid rain, acidification of soils, eutrophication of waterways). Currently, there are no economically viable alternatives to fossil fuels for synthetic N fertiliser manufacture.

It is evident that a large number of different factors play a significant role in aggravating the problem of excess N in the environment by contributing to the contamination of ground- and surface waters and to anthropogenic emissions of greenhouse gases. While agriculture is not the only contributor to excess N in the environment, it is increasingly implicated in contributing to climate change through the emission of greenhouse gases and to the decline in water quality through N runoff and leaching. Soils are not only an important basis of agricultural production; they also form the boundary between agricultural activities, water and atmosphere. Monitoring and managing this interface, are two crucial factors in determining and manipulating the magnitude of N loss from agricultural systems.

CONSEQUENCES OF N LOSS TO THE ENVIRONMENT

N loss to water

Nitrogen loss from soils by leaching and runoff from agricultural land substantially contributes to ground- and surface water pollution, especially in temperate and humid climates where rainfall often exceeds evapotranspiration. Such a loss is of serious concern for water quality but also has to be considered in an economic context as any N lost from the farm system signifies a financial loss to the farmer. There are a number of known natural and anthropogenic sources contributing to N loss to water, and it is widely accepted that agriculture is not solely responsible for the problems associated with excess N in the environment. Urban sources of N leaching to ground- and surface waters include fertilisation of parks, gardens and golf courses, contaminated land, construction and landfill sites, sewerage systems and highway drainage. In comparison, N from agricultural sources originates from fertilisers, manures or sewage sludge applied to arable crops or pasture, from spills and leaks from slurry and fertiliser storage, or is released from soil or added organic matter under conditions favouring mineralisation (Lerner 1999). Leaching (drainage) of N occurs in soils with high N content when crop uptake is low and excess water percolates through the soil column by matrix or macropore flow transporting N compounds below the rooting zone, where plants can no longer utilise them. In particular, nitrate ions are readily mobile in the soil due to their negative charge, while most available ammonium ions are bound to organic matter and clay particles or rapidly transformed into NO_3^- by nitrification. In temperate regions, optimum conditions for leaching often occur during wetter, colder months, making NO_3^- leaching a common winter phenomenon. The amount of NO_3^- lost is determined by inherent soil and environmental factors, such as nitrification rate, soil texture and structure, soil permeability and water holding capacity, drainage volume, N fertilisation level (especially outside of the growing season), and crop type (i.e. N requirement and N use efficiency [see Stark and Richards (2008) for more detail on NUE]) as well as crop yield (lower yields can be a sign of low N uptake) and rainfall. In arid regions, N leaching from soil systems is usually lower, however, NO_3^- can still move down the soil profile and thus out of reach of younger plants with smaller root systems. In an agricultural context, N leaching largely depends on management techniques, and several studies have been carried out to examine effects of different management systems on environmental quality. These include comparisons of arable, horticultural and/or grassland systems (e.g. Goulding 2000; Matlou and Haynes 2006), conventional and organic farming systems (e.g. Aronsson *et al*. 2007) and reduced or no-till and conventional tillage (e.g. Mkhabela *et al*. 2008). Similarly, effects of various fertilisation and manuring strategies have been investigated (e.g. Andraski *et al*. 2000; Bergström and Kirchmann 2004; Dahlin *et al*. 2005), as has the use of catch crops (e.g. Hooker *et al*. 2008; Nemecek *et al*. 2008) and the application of nitrification and urease inhibitors (e.g. Yu *et al*. 2007; Zaman *et al*. 2008) (also see Stark and Richards 2008).

Overall, leaching losses between 20 and 300 kg total N ha^{-1} yr^{-1} have been observed in the UK (Goulding 2000), which were affected by crop and soil type with largest amounts observed under free draining set-aside sites (areas temporarily taken out of agricultural production). Similar levels were measured from individual urine patches under different soil types in Ireland (10-305 kg N ha^{-1} yr^{-1}) (Stark *et al*. 2007) and from a spring barley crop with and without undersown catch crops (3-327 kg N ha^{-1} yr^{-1}) (Hansen *et al*. 2007). Di and Cameron (2002) also reported average N leaching losses between 4 and 107 kg N ha^{-1} yr^{-1} and between 6 and 162 kg N ha^{-1} yr^{-1}, when reviewing data obtained from arable cropping and grazed grassland systems, respectively, under varying environmental conditions.

Generally, results are varied and show large seasonal and/or yearly fluctuations, which suggests that it is important to assess farming systems and crop rotations as a whole over a longer-term period instead of focussing on just one aspect or management technique. For example, NO_3^- losses from organic systems can be increased after ploughing of ley phases and incorporation of green manure crops, while overall losses are lower from organic compared to conventional farming systems, often resulting from lower N inputs and/or lower stocking densities (Dalgaard *et al*. 1998; Di and Cameron 2002). Most of these findings, however, depend on their assessment on a per area unit (e.g. hectare) or per unit of yield (e.g. tons) basis. In some instances, no differences or negative effects (i.e. higher N leaching) of organic management systems were observed when assessing the parameters per unit of yield (Aronsson *et al*. 2007). On the other hand, adjusting fertiliser levels, using undersown or winter catch crops and optimising irrigation strategies have been shown to be a consistently effective measures to reduce N leaching from arable and grassland systems (also cf. Mitigation Strategies, p. 49). Both strategies have been proven to achieve significant reductions in both total N leaching losses (kg ha^{-1}) and NO_3^--N concentrations in the drainage water (mg L^{-1}) (Hansen *et al*. 2007; Ulén *et al*. 2008b). It should be noted that the response of N leaching to N fertiliser supply is not linear and seems to remain low until N additions reach a threshold level, which might be closely linked to yield response (McSwiney and Robertson 2005). This will be of significance when determining optimum N supply to meet crop demand and limit environmental impact. With regards to tillage, NO_3^--N concentrations were generally lower in drainage water from no-till compared to conventional tillage systems, which can most likely be attributed to higher denitrification rates in the no-till soils (Mkhabela *et al*. 2008). However, while reduced tillage has a positive effect on N losses to waterways the effects on N gaseous emissions are ambiguous and possible interactions with or negative effects on other aspects of the N cycle have to be taken into consideration when assessing farming systems for their environmental impact.

Effects on human and animal health

High NO_3^- levels in drinking water are toxic to livestock and can result in methemoglobinemia and abortions in cattle, and were believed to be the primary cause for methemoglobinemia in humans and particularly in infants (blue-baby-syndrome). This widely accepted understanding has been challenged in recent years (Fewtrell 2004; Addiscott 2005), which has lead to drinking water standards being questioned, with a view to removing N restrictions in the agricultural sector (Addiscott 2005). However, NO_3^- may be just one of many interrelated factors causing methemoglobinemia. Thus, high NO_3^- concentrations in drinking water might still affect the severity and accelerate the condition by increasing the concentration and amounts of NO_3^- and NO_2^- in the body (Fewtrell 2004). Moreover, there is evidence that excess NO_3^- might be associated with the occurrence of non-Hodgkins lymphoma and Parkinson's disease (Ward *et al*. 1996).

In contrast, a number of investigations have demonstrated potential health benefits of NO_3^- in drinking water. Salivary NO_3^- is converted to NO_2^- in the mouths of adults and children of 6 months and over, which, under acidic conditions, is transformed to peroxynitrite via nitrous acid (HNO_2) and nitric oxide (NO). These products have been shown to have potent antimicrobial properties, which include, for example, control of fungal infections (Benjamin and Dykhuizen 1999), air tract inflammations (L'Hirondel and L'Hirondel 2002) and viral infections (Colosanti *et al*. 1999), as well as positive effects on the cardio vascular system through the inhibition of blood platelet aggregation (McNight *et al*. 1999). These studies on the potential positive effects of NO_3^- consumption challenge the EPA, WHO and EU standards regarding acceptable levels of NO_3^- in drinking water (L'Hirondel and L'Hirondel 2002) (Current standards for drinking water quality: EU Council Directive 98/83/EC[1]: < 11.3 mg NO_3^--N L^{-1}, 0.15 mg NO_2^--N L^{-1}; WHO[2]: 11.3 mg NO_3^--N L^{-1}, 0.91 mg NO_2^--N L^{-1}; US EPA[3]: 10 mg NO_3^--N L^{-1}; 1 mg NO_2^--N L^{-1}). Nonetheless, excess NO_3^- has been associated with other health problems for both humans and livestock, and questions remain in particular regarding the carcinogenic and teratogenic effects of other N compounds, such as N-nitrosamine (Ía GarcRoché *et al*. 1987). Even if NO_3^- is found to be purely beneficial for animal and human health, therefore removing the need for restrictions on drinking water, it must be kept in mind that its effects on the environment can be severe. To maintain environmental quality and prevent deterioration of freshwater and marine habitats, it would still be required to monitor and limit NO_3^- levels in water bodies.

Effects on aquatic systems

Eutrophication is the over-enrichment of aquatic environments with nutrients and organic matter, leading to changes in primary production, biological structure and turnover and resulting in a higher trophic state of water bodies. This mainly manifests itself as proliferation and subsequent death of algae, the decomposition of which leads to increased oxygen consumption by aerobic bacteria resulting in low dissolved oxygen concentrations in bottom waters and sediments of aquatic systems. This can result in hypoxic (low oxygen) or anoxic (oxygen depleted) conditions in stratified rivers, lakes, estuaries and coastal waters and is normally an annual summertime occurrence. Due to the low oxygen concentrations in hypoxic zones (<2 ppm), they often cannot support fish and other marine life and are commonly called dead zones. Hypoxic zones remain in the affected areas for as long as oxygen consumption exceeds replenishment. Currently, there are more than 200 known dead zones worldwide (Schrope 2006), examples are the Baltic sea, the northern Gulf of Mexico and Black Sea. The total hypoxic area in these three coastal zones is estimated to be 145,000 km^2.

The role of NO_3^- in surface water contamination by adding to eutrophication has been critically underestimated in the past. In freshwater environments, P (phosphorus) has often been identified as the foremost limiting nutrient for algal proliferation, thus misjudging the importance of N in phytoplankton growth and harmful algal blooms (Schindler 1977). Excess N has been shown to play an important role in the net primary production of aquatic systems, especially in lakes and streams with low N:P loading ratios (Elser *et al*. 1990) and N pollution has been increasingly linked with the occurrence of eutrophication. While other nutrients, such as silicon and iron, also significantly influence the growth and abundance of algae (e.g. diatoms), the extent of their impact is generally less than that of N and P (Anderson *et al*. 2002). There is evidence that large portions of the world's coastal and estuarine ecosystems are limited by inorganic N availability, which is mainly controlled by riparian and groundwater N inputs (Howarth and Marino 2006). Due to the complex interaction between nutrient loading and light limitation, individual estuaries respond differently to nutrient loading in time and space with some being more sensitive. This suggests that threshold values for water N concentrations will need to be lowered to prevent eutrophication (Cloern 2001), and that both P and N are equally important for preventing and managing eutrophication of aquatic ecosystems. Regulating (and limiting) N input into water bodies below the targets implemented to assure drinking water quality (10-11.3 mg NO_3^--N L^{-1} and 0.15-1 mg NO_2^--N L^{-1}; see above) may help reduce the threat of eutrophication. Dodds *et al*. (1998) suggested upper limits for total N of 1.26 mg N L^{-1} for eutrophic temperate lakes and 1.5 mg N L^{-1} for streams. In comparison, the Canadian Council of Ministers of the Environment (Anonymous 2003) recommends water quality standards of 2.9 and 3.6 mg NO_3^--N L^{-1} based on toxicity data to protect freshwater and marine ecosystems, respectively. Similarly, Camargo and co-workers recommended a maximum level of 2 mg NO_3^--N L^{-1} for the protection of sensitive aquatic animals (Ca-margo *et al*. 2005) and total N levels below the range of 0.5–1.0 mg N L^{-1} to protect aquatic ecosystems from acidification and eutrophication resulting from inorganic N pollution (Camargo and Alonso 2006). They also suggested focussing on total N levels, which includes organic N, rather than on dissolved inorganic N (i.e. $NO_3^- + NO_2^- + NH_4^+$) as total N may be related more closely with algal biomass and toxin production (Dodds *et al*. 1997; Graham *et al*. 2004). The relatively low limits could also protect aquatic animals against NO_2^- toxicity since most of the dissolved inorganic N is present as NO_3^- rather than NO_2^-. The threshold values for aquatic N concentrations to protect water habitats must be adjusted to be applicable to different water environments, systems and pathways. At present, detailed understanding of complex groundwater and surface water interactions, including information on hydrogeological factors such as travel time, dilution, dispersion and attenuation, is generally limited in most countries, which complicates deriving threshold levels for groundwater from reference values of dependent aquatic ecosystems (Hinsby *et al*. 2008). More importantly, detailed knowledge on biogeo- and biohydrochemistry is required, thus considering mineral N reducing processes, such as denitrification, dis-

[1] http://ec.europa.eu/environment/water/water-drink/index_en.html
[2] http://www.who.int/water_sanitation_health/dwq/en/
[3] http://www.epa.gov/safewater/contaminants/index.html

similatory nitrate reduction to ammonium (DNRA), chemodenitrification and anaerobic ammonium oxidation (anammox) in soils and water systems (cf. Stark and Richards 2008). By understanding and accounting for these processes, robust and scientifically defendable standards can be set at the catchment scale to ensure that threshold targets are not overly stringent.

N loss to air

Although effects of agricultural management on water quality have been the chief focus of environmental protection campaigns, air quality issues have become an increasing concern over recent years. Gaseous forms of N, including ammonia (NH_3), nitrogen dioxide (NO_2), NO and N_2O, can contribute substantially to global warming, the depletion of the stratospheric ozone layer and/or nutrient enrichment of ecosystems through atmospheric N deposition. They occur from both natural (including volcanoes, soils oceans, biological decay and lightning strikes) and anthropogenic sources. The anthropogenic sources are mostly of industrial and agricultural origin and can include industrial acid production, burning of biomass (including forest fires) and fossil fuels, emissions from animal and human wastes, mineral fertilisers, as well as biogenic emissions from managed soil systems (Bouwman *et al.* 1997). Over recent years, agriculture has been identified as the main anthropogenic source contributing about 50% towards total N emissions (including all four N types). In contrast, fossil fuel combustion accounts for about 30% and biomass burning for 10% of total emissions (Crutzen 1981; Krupa 2003; IPCC 2007). Although abiotic production of nitrogenous gases has been observed from soils (e.g. Venterea and Rolston 2000), N emissions are predominantly the result of microbial and N turnover processes, such as nitrification, denitrification and ammonia volatilisation from fertilisers and manures (Sanhueza 1982).

The Intergovernmental Panel on Climate Change (IPCC) has identified N_2O as a powerful, long-lived greenhouse gas (N_2O remains in the atmosphere for an average of 114 years) that contributes significantly to positive radiative forcing[4] and the depletion of the stratospheric ozone layer (IPCC 2001). Approximately 11 Tg N yr^{-1} of N_2O emissions occur from natural sources, including microbial processes in soils (65% of the total) and oceans (app. 30%) and chemical oxidation of NH_3 in the atmosphere, while emissions from anthropogenic sources amount to about 6.7 Tg N yr^{-1} (IPCC 2001; IPCC 2007). Mosier *et al.* (1998) divides agricultural sources of N_2O into direct and indirect emissions with direct emissions originating from agricultural soils and animal production, and indirect emissions being induced by agricultural activities. Indirect emissions stem from fertiliser production, volatilisation of NH_3, NO and NO_2 (combined referred to as nitrogen oxides or NO_x), leaching or run-off, and from N in litter or biomass that has been removed from the soil system totalling about 2.1 Tg N yr^{-1} (Mosier *et al.* 1998). Although the majority of all N_2O emissions emanate from natural sources, human activity has caused atmospheric N_2O concentrations to increase from 270 ppb in pre-industrial times to 319 ppb in 2005, and a further increase by approximately 50% by 2030 has been projected (FAO 2002). This increase has primarily been attributed to agricultural intensification, i.e. increased N fertilisation and new irrigation practices, land conversion from forests to agriculture, in particular in the tropics, increased biomass burning and growing atmospheric depositions of NO_x and NH_3 (Smith 1997b).

While atmospheric emissions of N_2O are almost entirely a result of microbial activity, the primary sources for NO_x are the burning of fossil fuels and biomass and lightning (Sanhueza 1982; Galloway *et al.* 2004). Nonetheless, agriculture has been identified as an important factor in influencing emissions of NO_x, and increases since the start of the industrial age linked to human activity also include rising fertiliser use and agricultural intensification (IPCC 2007). Worldwide, total gaseous losses of NO from soils have been estimated to be as high as 21 Tg N yr^{-1} and emissions from fertilised agricultural land are around 1.8 Tg N yr^{-1} (Davidson and Kingerlee 1997). Although the effects of NO_x gases are mainly localised due to their short lifetime in the atmosphere, they contribute to acidification and eutrophication of ecosystems through N deposition and play a central role in stratospheric ozone depletion and in the formation and accumulation of surface ozone, which can trigger serious respiratory problems (Vitousek *et al.* 1997).

In the early 1990s, anthropogenic NH_3 emissions were estimated between 43 Tg N yr^{-1} and 52 Tg N yr^{-1}, and are expected to rise to about 109 Tg N yr^{-1} by 2050 (Galloway *et al.* 2004). Ammonia is a common by-product of animal manures, where excess N is converted into NH_3 by microbial activity during the decomposition process. Livestock, especially cattle and poultry, farming has been identified as the largest NH_3 producer within agriculture and losses are greatest from animal housing and grazing and from manure spreading and storage. Currently, NH_3 emissions from agricultural sources (including animals, fertilisers and cropped soils) are estimated to account for over 80% of total global NH_3 emissions, with fertiliser use usually considered as the second most significant source after volatilisation from animal wastes (Galloway *et al.* 2004; Reidy *et al.* 2008). However, this proportion is anticipated to increase with agricultural emissions rising to over 100 Tg N yr^{-1}, while the quantities from other sources are expected to remain the same or even decrease in the future (Galloway *et al.* 2004). Nonetheless, large uncertainties remain regarding the magnitude of total annual NH_3 emissions and the monthly, daily and diurnal variations, which can be attributed to the fact that most agricultural NH_3 emissions originate from difficult to measure non-point sources such as livestock housing and manured or fertilised fields (Wu *et al.* 2008).

While there is evidence that NH_3 contributes indirectly to global warming, one major problem resulting from NH_3 volatilisation and emissions of other N gases are due to their role in N deposition (IPCC 2007). Especially volatilised NH_3 can be transported long distances from the site of origin before being deposited, thus creating the potential for far-reaching negative impacts. Nitrogen in the atmosphere is re-introduced into the soil N cycle by deposition, which occurs depending on the geographical location and the proximity to an NH_3 source by gaseous adsorption of N gases, such as NO_2, NH_3 or nitric acid (HNO_3), by wet deposition (through precipitation) of NH_4^+ and NO_x and by dry deposition (as dust) of NO_3^- (Stevenson and Cole 1999). In unpolluted areas, N deposition can range from <1kg N ha^{-1} yr^{-1} to a few kilograms, often the result of NO_x being formed naturally through lightning in the atmosphere. In regions with higher industrialisation, energy consumption and more intensive livestock farming, these levels are generally exceeded ten-fold (Smith 1997a). Atmospheric N deposition has increased over recent decades and ranges from 5 to 80 kg ha^{-1} yr^{-1} with a global average of 17 kg ha^{-1} yr^{-1} (Skiba *et al.* 2004) have been observed (also cf. Stevenson and Cole (1999): up to or > 50 kg ha^{-1} yr^{-1} and Addiscott *et al.* (1991): up to 40 kg ha^{-1} yr^{-1}). High N deposition may lead to acidification and over-enrichment of soils and water bodies with readily available N. The decline in pH may cause changes in microbial process rates, which, in turn, affect nutrient turnover and plant species composition within sensitive habitats. Other observed effects of increasing soil N concentrations include greater susceptibility in plants to pests, diseases and environmental perturbation (e.g. Fangmeier *et al.* 1994). It is also likely that further gaseous emissions occur as a consequence of N deposition, which could be re-released into the atmosphere, indicating the possibility of multiple cycles and feedback effects.

[4] Radiative forcing describes the relationship between radiation coming in and leaving the atmosphere. A positive radiative forcing results in warming of the Earth surface, while a negative radiative forcing causes cooling.

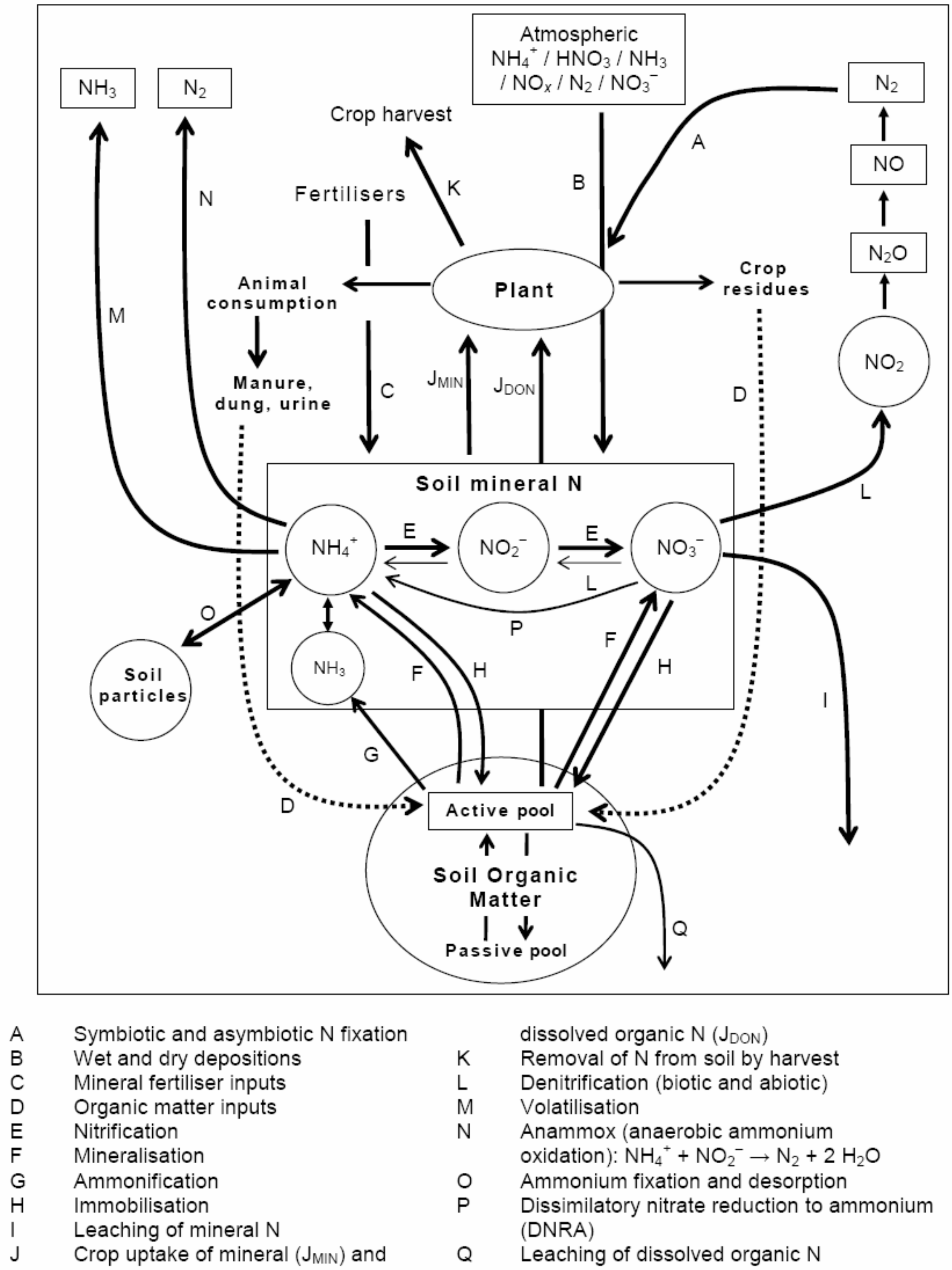

Fig. 1 Schematic representation of the soil nitrogen cycle.

The microbially mediated processes that lead to the evolution of N gases are driven by soil mineral N concentration, soil moisture content, oxygenation, temperature and availability of readily mineralisable organic matter. This indicates that agricultural management, e.g. fertilisation intensity and timing, irrigation practices, grazing intensity, tillage type and intensity, crop and residue type, can have direct and indirect effects on gaseous N emissions. Nitrous oxide and NO are produced under aerobic condition as a by-product of nitrification (the oxidation of NH_3 to NO_2^- and then NO_3^-) and under anaerobic circumstances as end-products of denitrification (Bremner and Blackmer 1978; Skiba *et al*. 1997) (**Fig. 1**). Nitrification usually occurs rapidly under warm, moist conditions and dominates as an emissions source at a water-filled pore space (WFPS) of up to 60%. In comparison, heterotrophic denitrification, the biological reduction of NO_2^- or NO_3^- to N_2, N_2O and/or NO, takes place mainly under anaerobic conditions in poorly drained soils and sediments (WFPS > 60%), although NO emissions are mostly a result of nitrification and are therefore often higher from well-drained compared to poorly-drained soils (Russow *et al*. 2008) (for more detail on N cycling processes, also refer to Stark and Richards (2008)).

Both processes are limited by the availability of an easily accessible C (carbon) source (Brettar and Höfle 2002; Russow *et al*. 2008), the presence of nitrifying or denitrifying organisms and, in the case of denitrification, the supply of NO_2^- or NO_3^-. It is important to note that the extent of NH_3 volatilisation in the soil also influences soil N availability, thus affecting nitrification and denitrification processes (Bouwman *et al*. 2001). While nitrification is a relatively constant process across ecosystems, denitrification rates are temporally and spatially extremely variable, which can be explained by varying N fertilisation rates and grazing or crop management practices throughout the year as well as seasonal fluctuations in soil temperature and, more importantly, soil moisture levels (Hyde *et al*. 2006). Moreover, high denitrification activity can occur within anaerobic microsites created by water-filled pores, the rhizosphere of crop plants, which supply organic C compounds and create anaerobic zones, or decomposing plant material. The process is carried out by over 33 genera of bacteria (incl. *Agrobacterium*, *Bacillus*, *Pseudomonas* and *Azospirillum*), most of which are capable of completely reducing NO_3^- to N_2 while several genera produce N_2O as final product of denitrification (Stevenson and Cole 1999). The ratio of N_2 to N_2O production strongly depends on soil properties such as oxygen levels (and thus soil moisture content and WFPS), pH, redox potential, temperature, NO_3^- content (high NO_3^- favours N_2O production) and C levels in the soil. Complete denitrification, i.e. N_2 production, which is environmentally benign, predominantly occurs under condition of >80% WFPS, high pH and low soil N levels (Stevenson and Cole 1999).

Production of N_2O and NO are driven by various interacting mechanisms and conditions. Nitrous oxide emissions are found to be closely linked with environmental factors, including climate, soil type and pH, and management-related factors, e.g. N application rate, fertiliser type, crop and residue type, plant cover, while NO emissions seem to be primarily controlled by form and rate of N fertilisation, organic matter content and drainage status of the soil (e.g. Breitenbeck and Bremner 1986; Abbasi and Adams 2000; Venterea and Rolston 2000; López-Fernández *et al*. 2007). However, especially the processes regulating NO emissions are not well understood and, to date, little agreement exists as to what the most important factors are that drive the extent of gaseous N emissions from soils (Venterea *et al*. 2005; Maljanen *et al*. 2007). In the past, the focus has been on studying and modelling N fluxes in response to N fertilisation and N availability, with results indicating a strong direct link between N fertilisation and emission levels (see, for example, Abbasi and Adams 2000; Venterea and Rolston 2000). More recent studies on arable and grassland systems, however, indicate that the response of N_2O evolution to the incremental rise in N fertilisation may not be linear (Chatskikh *et al*. 2005; McSwiney and Robertson 2005), and that N_2O emissions can be more accurately predicted by crop productivity than by the amount of N added to the system (see McSwiney and Robertson (2005) as an example for maize and Chen *et al*. (2008) for winter wheat).

There is also conflicting evidence regarding the effect of fertiliser type, including different forms of mineral N and organic fertilisers, such as slurry, manures and composts, on denitrification and gaseous N emissions from agricultural soils. Jones *et al*. (2007) found emissions to be greater from plots treated with poultry manure and sewage sludge pellets compared to fertilisation with ammonium nitrate (NH_4NO_3) and urea, likely due to the accumulation of inorganic N following organic matter additions, while López-Fernández *et al*. (2007) reported generally lower emission factors from organic fertilisers than from urea. Greater N_2O emissions have also been observed from anhydrous ammonia compared to other N fertilisers (Breitenbeck and Bremner 1986; Venterea *et al*. 2005). Bouwman *et al*. (2001) emphasised the significance of fertiliser type and application rate in affecting NO flux and emissions, while pointing out that it is more likely unused fertiliser N rather than total soil N driving the microbial processes that lead to gas production, this means timing fertiliser application with crop uptake is equally important. Generally, N_2O and NO emission peaks can be observed for a short period after fertilisation, which also indicates that the timing of fertiliser application is a crucial factor in regulating N_2O and NO flux and could explain the varying results obtained by different researchers.

Soil tillage intensity may affect both crop growth and soil nutrient turnover/balances, including emissions of greenhouse gases such as CO_2 and N_2O. While no-till or reduced tillage systems are widely promoted to reduce atmospheric CO_2 concentrations by increasing soil C storage, the impacts of different tillage practices on emissions of other greenhouse gases, such as N_2O and NO, also need to be considered in greenhouse gas balances. Findings comparing the effects of reduced to conventional tillage systems with regard to emissions of N gases are highly inconsistent (e.g. Venterea *et al*. 2005; Liu *et al*. 2007; Mkhabela *et al*. 2008). N_2O losses were shown to be significantly greater from minimum tillage fields (Beheydt *et al*. 2008) and from no till systems (Mkhabela *et al*. 2008) when compared with conventional tillage in a 1 and 2-year study, respectively. In the same study, Mkhabela *et al*. (2008) assessed the effect of tillage on NH_3 and found higher losses under no till compared to the conventional system. Jantalia *et al*. (2008) reported that reducing tillage did not significantly impact on N_2O emissions to the point of offsetting the benefits of C sequestration when comparing conventional tillage soybean-wheat systems with zero tillage agriculture including leguminous green manures on Brazilian Ferralsols. Similarly, Chatskikh *et al*. (2008) found in field measurements and model simulations conducted under Northern European conditions that N_2O emissions were lower from reduced compared to conventional tillage systems. The authors concluded that tillage effects on N_2O emissions are highly dependent on local climate, soil and management conditions. Malhi and co-workers (Malhi *et al*. 2006; Malhi and Lemke 2007) reported no significant effect of tillage or significantly lower N_2O emissions from an 8-year arable field experiment in Canada. With regards to tillage effects, the age of the system probably plays a significant role in determining N_2O losses as emissions tend to decline over time under long-term minimum or zero tillage, which can be attributed to changes in soil N availability (Six *et al*. 2004). Therefore, studies comparing differing management systems, including tillage, that are of less than three to five years duration may not be representative of the longer-term behaviour of these systems and results should be treated with caution. Similar care is advised when choosing N flux models to simulate gaseous emissions, which often have been shown not to correspond well with the measured data. Beheydt *et al*. (2008), for example, reported that the DNDC model overestimated emissions from conventionally tilled soils, while underestimating emissions from reduced tillage systems. Similarly, the concept of IPCC emission factors has been criticised with regard to N_2O and NO emissions, in particular under tropical conditions where environmental factors have been found to be of greater relevance than fertiliser type (Veldkamp *et al*. 1998; Jantalia *et al*. 2008).

Not surprisingly, most evidence demonstrates that the nature of N fluxes in agricultural soils is anything but straightforward and that the relationship between N_2O, NO and the external factors that influence gas evolution is not clear-cut. Overall, the literature suggests that both field measurements and experimental studies of gaseous N emissions from soils can be difficult to interpret and/or extrapolate as they are subject to high seasonal variability and because short-term experiments often produce misleading results. The frequency and quantity of measurements will also affect the validity of results and at least one measurement per day is recommended following events that trigger high emissions, while in periods of low emissions measurements can be taken less often (Bouwman *et al*. 2002).

One major factor that will affect future emissions of all greenhouse gases of anthropogenic origin, including N_2O and NO_x, is the potential for feedback effects from climate change. Global warming and rising CO_2 levels might stimulate microbial activity in soils and oceans leading to increased N_2O emissions from nitrification and denitrification processes. At the same time, melting of permafrost areas would result in the release of trapped N_2O and methane, which would further increase global warming and are examples of positive feedback. While it is feasible that rising temperatures cause a negative feedback effect by, for example, increasing evaporation and reducing wetland areas, Smith (1997b) states that the more rigorous models mostly predict positive feedback. There is evidence that the effect of global warming in N_2O emission is non-linear and that relatively small increases in temperature will bring about large increases in emissions (Smith 1997a). There clearly is a need for more experimental research and monitoring studies, especially in the longer-term, as well as N_2O and NO flux models that include parameters with higher predictive power in order to be able to more accurately predict emissions from different management scenarios and under wider environmental conditions.

LEGISLATION

As our understanding and appreciation of the environmental impact of N emissions from anthropogenic sources has improved, more effort is being put into combating both, the rise in N loss to the environment and the effects of increasing N emissions resulting from the intensification of agricultural production systems. Tackling the environmental N issue is not only of general concern due to possible negative health effects and nature conservation implications, and of interest to farmers wanting to minimise financial losses, it also poses a major legislative challenge. Over the last 45 years, legislation and guidelines have been introduced on a national and international scale to address excess N in the environment on various levels. These range from drinking water quality standards (for example, the National Primary Drinking Water Regulations in the United States[5] or the WHO guidelines for drinking water quality[6]) to legislation primarily focussed on environmental protection of water habitats (for example, the European Union Nitrates Directive (91/676/EEC)[7] and the US Clean Water Act[8]). Most early water quality legislation was principally aimed at safeguarding human health, while more recently, environmental protection has gained greater emphasis as water habitats are seen as ecosystems threatened by nutrient contamination and loss of biodiversity. The following provides examples of water quality legislation in Europe, USA and New Zealand, to show different approaches of societies attempting to balance agricultural and economic growth with environmental protection.

In the mid twentieth century, a number of health reports directly linked the occurrence of methemoglobinemia to elevated NO_3^- concentrations in drinking water (Comly 1945; Bosch *et al.* 1950) and advised keeping N levels below 45 mg or, at the most, 90 mg NO_3^- L^{-1}. This led to the introduction of 45 mg NO_3^- L^{-1} as a safe limit for drinking water by the Public Health Service in the United States in 1962 (McKee and Wolf 1963) and the WHO in 1971. Conversely, national legislative limits varied worldwide during the 1970s, ranging from 15 to 90 mg NO_3^- L^{-1}. To harmonise water quality standards within Europe, the EEC introduced the Council directive 80/778/EEC[9] in 1980, which set guidelines for the quality of drinking water for human consumption and established nitrate limits of 50 mg NO_3^- L^{-1} as the maximum admissible concentration, and 25 mg NO_3^- L^{-1} as the recommended, non-mandatory guide value. In the late 1990s, after a review by the WHO, which advocated the introduction of a single global standard to prevent methemoglobinemia, the guide level was abolished leaving 50 mg NO_3^- L^{-1} (= 11.3 mg NO_3^--N L^{-1}) as the EU-wide adopted limit in the new Drinking Water Directive (98/83/EC)[10]. With the introduction of the Nitrates Directive (91/676/EEC)[11] in 1991, the EEC for the first time addressed the wider implications of N in the environment and further restricted NO_3^- discharge in order to prevent ground- and surface water contamination from agricultural sources. The Directive requires Member States to designate vulnerable zones where NO_3^- leaching into already polluted waters causes additional contamination. Where necessary, Member States have to establish action programmes, which promote the application of codes of good agricultural practices. Moreover, the eutrophication status of freshwater, groundwater, estuaries and coastal waters has to be monitored every four years. The concurrent introduction of the Urban Waste Water Directive (91/271/EEC)[12] put in place emission standards for the discharge of urban wastewaters. These standards are based on the eutrophication sensitivity of receiving water bodies, and aim at reducing the N contribution of the urban sector to ground- and surface water contamination. Other European directives, e.g. the Habitats Directive (92/43/EEC)[13] and the Birds Directive (79/409/EEC)[14], require Member States to protect sensitive habitats and establish action programmes to restore ecosystems from anthropogenic impacts, including eutrophication, to achieve favourable conservation status. This has major implications for the nutrient status of water bodies, including estuarine and coastal areas, and is one of the reasons why the EU strengthened and amalgamated existing legislation on water protection with the Water Framework Directive (2000/60/EC)[15] (WFD) introduced in 2000. One of the main aims of the WFD is for Member States to achieve 'good' status for all surface, ground- and coastal waters by 2015. Member States must control chemical pollution, prevent further deterioration and enhance the condition of aquatic ecosystems, while simultaneously promoting the sustainable use of water. This directive, together with daughter directives and amendments, such as the Groundwater Directive (2006/118/EC)[16] and the Directive on Environmental Quality Standards in the Field of Water Policy (COM(2006) 397)[17], has increased legislative pressure on farmers and communities to reduce N loss from agricultural land with the objective of preventing eutrophication and other types of ecological deterioration of receiving water bodies.

Correspondingly, other countries have introduced legislation to protect drinking water and other water resources. In USA, the Clean Water Act[18] of 1972 (CWA) was introduced to restore and maintain the chemical, physical and biological integrity of waters in order to attain fishable and swimmable status. The CWA controls emissions from industrial and municipal facilities through the National Pollutant Discharge Elimination System, which is managed by authorised states and requires all point sources (i.e. discrete, identifiable places of discharge) that emit pollutants directly to surface waters to obtain a permit. Moreover, the CWA provides a demonstration grant program available to public and non-profit entities, which facilitates the implementation of control efforts of non-point source pollution from agricultural watersheds. These programs include development of local planning tools, public education and extension, assessments of non-point source pollution (i.e. discharge of

[5] http://www.epa.gov/safewater/contaminants/index.html
[6] http://www.who.int/water_sanitation_health/dwq/en/
[7] http://ec.europa.eu/environment/water/water-nitrates/index_en.html
[8] http://epw.senate.gov/water.pdf
[9] http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31980L0778:EN:HTML

[10] http://ec.europa.eu/environment/water/water-drink/index_en.html
[11] http://ec.europa.eu/environment/water/water-nitrates/index_en.html
[12] http://ec.europa.eu/environment/water/water-urbanwaste/directiv.html
[13] http://ec.europa.eu/environment/nature/nature_conservation/eu_nature_legislation/habitats_directive/index_en.htm
[14] http://ec.europa.eu/environment/nature/nature_conservation/eu_nature_legislation/birds_directive/index_en.htm
[15] http://ec.europa.eu/environment/water/water-framework/index_en.html
[16] http://ec.europa.eu/environment/water/water-framework/groundwater/policy/current_framework/new_directive_en.htm
[17] http://eur-lex.europa.eu/LexUriServ/site/en/com/2006/com2006_0397en01.pdf
[18] http://www.epa.gov/r5water/cwa.htm

pollutants from non-specific, unidentifiable sources such as runoff or leaching) at sub-watershed scale and information on best management practices and measures to reduce pollutant loadings. Water quality standards and monitoring criteria for receiving waters are well developed and widely used in the United States with regard to nutrients and pollutants, such as pesticides and suspended solids. Each state either adopts the national standards or develops its own legislation and / or standards based on US EPA (Environmental Protection Agency) criteria and guidelines, which in many cases are more stringent than the national regulations (EPA 1994). Maximum effluent limits and total maximum daily loads (TMDL) are applied to point sources ensuring water quality as part of the waste load allocation process (Swietlik *et al*. 1991). The US EPA defines TMDL as "a calculation of the maximum amount of a pollutant that a water body can receive and still meet water quality standards" (http://www.epa.gov/OWOW/TMDL/intro.html). It thus provides daily estimates of the maximum loading of any type of pollutant from all point, non-point and background sources, which does not exceed the threshold concentration in the receiving water body. More recently, the TMDL concept has been specifically applied to nutrient emissions from agricultural sources to water. In general, however, in the United States, the emphasis is put on non-regulatory and voluntary control of non-point source pollution (Foran *et al*. 1991), which is in contrast to the European approach.

In New Zealand, agriculture has experienced a major expansion over recent decades and dairy farming accounts for approximately 20% of the total export income (MfE 2003). The subsequently growing pressure on the environment and especially waterways led to the establishment of the Resource Management Act in 1991 (RMA), which required the sustainable management of natural and physical resources including agricultural lands and associated receiving waters. Approaches for the management of agricultural non-point source pollution have been developed in regional plans consistent with regional policy statements. Voluntary, economic and regulatory approaches have been promoted as part of the RMA. Regulatory means are used predominantly to control point source discharges; interest and stakeholder groups have also developed their own programmes or guidelines to manage the impacts of their activities on the environment. Some examples include the Federated Farmers' promotion of co-operative farmer landcare groups and the development of guidelines for sustainable agriculture (Rowan and Smith 1997).

Although the RMA has established water quality classes for surface waters and standards for temperature, pH and dissolved oxygen, no other national standards currently exist (Caruso 2000). While economic measures are rarely used, voluntary approaches are widely promoted in New Zealand. A framework with specific targets to promote sustainable dairy farming and protect water quality and water environments in dairying areas has been established in form of the Dairying and Clean Streams Accord (MfE 2003). To assist with the implementation of the Accord, regional actions plans have been developed jointly between Fonterra Co-operative Group (the main dairy co-operative in New Zealand) and regional councils. An assessment of the success of the Accord in June 2004 revealed that most of the set targets had been met and that nine regional action plans had been completed, while four were still being developed (MfE 2007). While the water quality in New Zealand is largely regulated by voluntary standards and best practice guidelines, the OECD strongly recommended the introduction of national environmental standards in a recent review to help regional authorities design regulatory and economic measures to reduce loss of N and other potential pollutants to ground and surface waters (OECD 2007). One step towards achieving this goal is the Health (Drinking Water) Amendment Act 2007, which came into force in July 2008[19].

Under this legislation, drinking water quality targets are set close to those in the US and EU. Suppliers of drinking water have to ensure compliance with New Zealand Drinking Water Standards[20] and implement public health risk management plans to protect drinking water from contamination. The Act further asks for information about compliance to be recorded and made available to the public, and imposes a series of penalties in the case of non-compliance.

Based on the perceived negative health effects, early legislation regulating NO_3^- in drinking water, such as the EEC Drinking Water Directive from 1980, used very simple measures of water quality as a gauge of success. Regulatory indicators such as residual soil N, nutrient inputs, farm nutrient balances and surpluses and livestock densities have been proposed and used for the evaluation of the potential impact of agriculture on water quality. The threshold or target values associated with these indicators often do not consider site specific environmental factors that can contribute to large variability. Schröder *et al*. (2004) emphasised the importance of global harmonisation of environmental legislation aiming at the introduction of a consistent set of rules rather than the standardisation of threshold values, which would consequently facilitate the selection of suitable indicators. In Europe, the WFD has changed from setting water quality standards by introducing the concept of water body specific targets, which now form the fundamental underpinning of the WFD. These targets are based on a water body's natural, unpolluted state, which serves as a reference condition, with the aim to achieve 'good' biological and chemical status. This will lead to considerable variation in threshold values for waters within countries and across the EU.

The challenge for all sectors releasing N into aquatic systems is to reduce emissions in order to decrease eutrophication of rivers, lakes, estuaries and coastal waters. In most countries, legislation is the primary driving mechanism to manage N pollution of waterways by introducing water quality standards. Legislation often also acts as a driver for both applied and fundamental N-related research. Nonetheless, the science of environmental monitoring requires further development to provide robust, accurate and timely information on water quality so that long- and short-term changes can be assessed and reported in an appropriate manner (Dworak *et al*. 2005). As achieving good status in water quality is a goal most countries aspire to, it is heartening to note that nutrient control measures developed through interdisciplinary research and introduced in Denmark over the last 20 years have led to a decline in N concentrations in coastal water by up to 44% (Carstensen *et al*. 2006). These encouraging observations are the first demonstration of the efficacy of nutrient reduction measures to improve water quality. However, simple water quality standards fail when applied to water bodies with differing morphologies and residence times. Based on a monitoring study, Ekholm *et al*. (2007) reported that water protection measures introduced as part of the EU Agri-Environmental Program in Finland in 1995 did not result in a noticeable reduction in nutrient loading in agricultural catchments, rivers, lakes and estuaries and did not lead to an improvement in water quality. The authors attributed the ineffectiveness of the actions primarily to the soils' inherent nutrient loading but also hypothesised that negative effects of environmental factors, such as changes in agricultural production or climate, on nutrient reduction may outweigh the benefit of the agri-environmental measures adopted. They suggested that the initiative might have been more successful if there had been a more direct focus on the principal areas and practices that contribute to high nutrient loading.

It is equally important for policy makers and stakeholders to understand the concept of lag time, which is defined

[19] http://www.moh.govt.nz/moh.nsf/0/7b0ed97145c8f73dcc256e27006e1053?OpenDocument

[20] http://www.moh.govt.nz/moh.nsf/by+unid/12F2D7FFADC900A4CC256FAF0007E8A0?Open

as the period between introducing protection measures and the first improvements in water quality at the catchment scale. This issue is particularly evident for diffuse N sources as there are generally long and complex pathways between soil, groundwater and rivers (Collins and McGonigle 2008). The British Geological Survey recently modelled aquifer response times for England and Wales and the results indicated a timescale of decades (8 and 46 years to reach 50% breakthrough in alluvial and chalk aquifers, respectively) for most aquifer classes during which time NO_3^- pollution will continue to reach discharge points despite reductions in contemporary surface loadings (Lord *et al*. 2007). Factors affecting the response time of groundwater bodies to mitigation measures include the amount of recharge (i.e. rainfall – [evapotranspiration + overland flow + lateral flow]), as higher recharge results in faster response times; the hydrology of the unsaturated zone (including matrix and preferential flow paths); the depth of the unsaturated zone; hydrogeological factors, such as porosity, storage and permeability; and the length of the pathway between recharge and discharge. In many countries, estimated response times of groundwater quality to changes in N inputs are not realistic for baseflow-dominated systems or chalk catchments, where significant retardation of chemicals can occur. Model simulations suggest that N groundwater concentrations in such systems will not decline for several decades after input has been reduced or stopped, and that increases resulting from historical nutrient loading are inevitable within the short term (Jackson *et al*. 2007). In general, the timelines envisioned for legislative changes to take effect are often too short and set unrealistic targets for land managers to reduce N loss to the environment. The WFD, for example, aims to achieve 'good' status of water bodies by 2015, which is 3 years following the implementation of the measures. Achieving this objective not only depends on the efficacy of the actions taken but also on the response time of the respective water body.

Evidently, the responsibility of improving water quality status does not solely lie with land managers. It is important for policy makers to be aware and understand the complexities of nutrient transfer from soil to water and not simply to use legislation to introduce and/or reinforce compliance measures. If nutrient reduction measures are found to be ineffective as a result of system effects, such as the response time of receiving waters and long hydrological pathways, it would be unjustified to burden land managers with more regulations and related cost increases. A number of pieces of legislation, including the WFD, now involve public participation with the aim of improving water quality. Community groups, such as farmers, private homeowners, academic researchers and local government representatives, participate in discussion groups and the decision making process on water management. Policy makers hope that encouraging the contribution of public groups and private individuals with different viewpoints and priorities will make it easier to meet the needs of all stakeholders and facilitate achieving the targets set by national and international legislation. In the Netherlands, it has been shown that high levels of cooperation and clear incentives are required when aiming to reduce agricultural impacts on the environment. Outcomes of pilot projects indicate that knowledge transfer as well as financial incentives are important means by which to encourage land managers to adopt new strategies and introduce change (van den Berg and van Lamoen 2008). The more-inclusive approach, which takes into account all pollution sources and puts emphasis on input from stakeholders, has resulted in active participation of farmers in the Dutch pollution programmes. The stakeholder focus of these projects has enabled the exchange of views, ideas and approaches in order to implement additional N reduction measures and improve the efficiency of existing means. Another important step towards solving the N problem is for land managers to appreciate the impact of agricultural practices on water bodies and atmosphere beyond the farm boundary. A study in Scotland highlighted that many farmers did not accept their responsibility for water quality deterioration and often considered non-terrestrial environments as 'out of sight, out of mind' (Macgregor and Warren 2006). Intense consultation and collaboration between policy makers and land managers may not only improve the understanding of environmental impacts beyond the farm gate, it may also yield more practical and economical approaches to nutrient reduction measures. Through day-to-day management, land managers have an intimate knowledge of their land, enabling them not only to implement nutrient reduction measures effectively but also to identify additional measures that are suitable for a particular farm environment. While this greatly increases the prospect of meeting legislative targets to protect and improve water quality, it is also essential for land managers to understand and accept the impact of farming systems and practices on environmental quality and for legislators to acknowledge the expertise and know-how of land managers (Cunningham and Sinclair 2005).

Gaseous emissions of N compounds are also attracting increasing international attention, as evident in recent introduction of legislation on air quality and green house gas emissions. However, covering these issues in detail is outside the scope of the review. In brief, these legislations are primarily implemented at the international scale due to the transboundary effect of gaseous compounds (e.g. N_2O contributes to climate change and NH_3 contributes to soil acidification and eutrophication a long distance from the source of origin) and include the 1999 Gothenburg Protocol[21], the 1997 Kyoto Protocol[22], the 2008 Bali road map[23] and the Kyoto2 framework[24], which is intended to replace the Kyoto Protocol in 2012. The Gothenburg Protocol addresses the issue of acidifying gases with the aim of reducing NO_x and NH_3 emissions by 41 and 17%, respectively, compared to levels of 1990, while the Kyoto protocol sets a reduction target for greenhouse gases (including N_2O) of 5.2% for industrialised countries (Annex 1 coutries[25]). More recently, further reductions in greenhouse gas emissions have been agreed by the international community as part of the Bali road map, which also has the support from larger nations including USA. Correspondingly, the EU outlined its new approach to further limit greenhouse gas emissions by at least 20% by 2020, or 30%, if other developed countries are prepared to commit to similar targets[26]. Overall, gaseous N emissions especially from agricultural sources can be expected to be subject to increasing restrictions and regulations, which will be implemented on a national scale to meet reduction targets and help combat climate change.

MITIGATION STRATEGIES

Over the past 30 years, research has focused on quantifying N loss to the environment, improving our understanding of N cycling processes and developing strategies to reduce N loss to the environment. Increased knowledge of terrestrial and aquatic N cycling processes has identified pathways and processes that can be manipulated to reduce N emissions and maximise nutrient use efficiency. Similarly, there are a wide range of agricultural and agronomic practices, which can be employed to reduce environmental N pollution, generally referred to as mitigation methods, options or measures or best management practices. A range of approaches can be applied to reduce N loss to air and water from agriculture at different levels within the farm system. In **Table 1**, we identify mitigation options based on land management, integrated nutrient (or fertiliser) management,

[21] http://www.unece.org/env/lrtap/full%20text/1999%20Multi.E.Amended.2005.pdf
[22] http://unfccc.int/resource/docs/convkp/kpeng.pdf
[23] http://unfccc.int/resource/docs/2007/cop13/eng/06a01.pdf
[24] http://www.kyoto2.org/
[25] http://unfccc.int/parties_and_observers/items/2704.php
[26] http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=COM:2008:0030:FIN:EN:HTML

Table 1 Selection of practices and measures to mitigate N loss from agriculture to air and water, effects on different N species and consequences for farmer and farm system.

Category	Mitigation measures	Effect on N species	Impact on farm system	Reference
Land management				
Tillage	a) Timing of tillage (e.g. spring ploughing)	$\downarrow NO_3^-$; $\downarrow N_2O$	$\uparrow$herbicide use; winter green cover required; possible $\uparrow$crop N deficiency	Dahlin *et al.* 2005
	b) Reduced tillage / no till	$\downarrow NO_3^-$; $\updownarrow N_2O$	$\downarrow$cultivation costs; $\uparrow$soil C content	Jantalia *et al.* 2008; Mkhabela *et al.* 2008
	c) Management of physical soil properties (limit compaction; increase aeration)	$\downarrow N_2O$	$\uparrow$cultivation costs	Oenema *et al.* 1998
Landscape planning / engineering	a) In-field and field edge vegetated buffer strips	$\downarrow NO_3^-$; $\updownarrow N_2O$	no extra cost; $\uparrow$pests	Ulén *et al.* 2008
	b) Field margin hedgerows	$\downarrow NO_3^-$; $\uparrow N_2O$; $\downarrow NH_3$	$\uparrow$planting cost; $\uparrow$biodiversity	Livesley *et al.* 2008
	c) Prevention of animal access to water ways	$\downarrow NO_3^-$	$\uparrow$fencing; bridging and drinking water costs	Line *et al.* 2000
Farming system / Land use	a) Conversion from arable to extensive grassland farming	$\downarrow NO_3^-$; $\downarrow N_2O$	$\downarrow$yields	Lord *et al.* 1999
	b) Conversion from arable/grassland farming to forestry	$\downarrow NO_3^-$; $\downarrow N_2O$	$\downarrow$yields; change of farming system	Saggar *et al.* 2008
	c) Conversion to organic farming	possible $\downarrow NO_3^-$; $\downarrow N_2O$	possible $\downarrow$yields; change of farming system	Aronsson *et al.* 2007
Denitrification enhancement	a) C amendment of groundwater and drains	$\downarrow NO_3^-$; $\uparrow N_2O$	$\uparrow$costs; $\uparrow N_2O$	Fenton *et al.* 2008
	b) Deterioration of field drainage	$\downarrow NO_3^-$; $\uparrow N_2O$	$\downarrow$yields	Shepherd and Chambers 2007
	c) Re-instating wetlands and farm ponds	$\downarrow NO_3^-$; $\uparrow N_2O$	$\uparrow$establishment cost	Hoffmann and Baattrup-Pedersen 2007
Other	Crop rotation management / Cover cropping	$\downarrow NO_3^-$; $\downarrow N_2O$	$\uparrow$cost; $\downarrow$weeds	Hansen *et al.* 2007; Hooker *et al.* 2008
	GPS guided precision farming, e.g. for fertilisation	$\downarrow NO_3^-$; $\downarrow N_2O$	$\downarrow$fertiliser costs; $\uparrow$technology + costs	Power *et al.* 2000
	Controlled irrigation on dry soils	$\downarrow NO_3^-$; $\uparrow N_2O$	$\uparrow$costs; $\uparrow$yields	Power *et al.* 2000
	Selection of high yielding sward and crop species	$\downarrow NO_3^-$; $\downarrow N_2O$	$\uparrow$costs; $\uparrow$yields	Velthof *et al.* 1998
	Control of soil pH	$\downarrow NO_3^-$; $\downarrow N_2O$	possible $\uparrow$costs	Oenema *et al.* 1998
Integrated nutrient management				
Quantity	a) Matching fertiliser quantity with crop demand	$\downarrow NO_3^-$; $\downarrow N_2O$; $\downarrow NH_3$	$\downarrow$fertiliser costs; $\uparrow$technology	Andraski *et al.* 2000; van Groenigen *et al.* 2008
	b) Accounting for N applied as animal manure	$\downarrow NO_3^-$; $\downarrow N_2O$	$\downarrow$fertiliser costs	van Groenigen *et al.* 2008
	c) Minimum crop requirements/ fertilisation below crop requirements	$\downarrow NO_3^-$; $\downarrow N_2O$	$\downarrow$yields; $\downarrow$fertiliser costs	Velthof *et al.* 1998; Shepherd and Chambers 2007
	d) Increasing N use efficiency	$\downarrow NO_3^-$; $\downarrow N_2O$	$\downarrow$fertiliser costs; $\uparrow$yields; possible $\uparrow$technology	Skiba *et al.* 1997
	e) Reduction of pasture N content by reducing fertilisation	$\downarrow NO_3^-$; $\downarrow N_2O$ $\downarrow NH_3$	$\downarrow$fertiliser costs; $\uparrow$feeding costs	Oenema *et al.* 1998
Timing	a) Autumn vs. winter application of manures	$\downarrow NO_3^-$; $\uparrow N_2O$	$\uparrow$storage costs; crop N deficiency possible	Smith *et al.* 2002; Dahlin *et al.* 2005
	b) Fertilisation according to crop requirements	$\downarrow NO_3^-$; $\downarrow N_2O$	$\downarrow$fertiliser costs	Velthof *et al.* 1998
	c) No fertilisation at high-risk times	$\downarrow NO_3^-$; $\downarrow N_2O$	no extra cost; possible $\uparrow$storage cost; $\uparrow$spread lands (manure)	Chambers *et al.* 2000
Placement	No fertilisation of high-risk areas	$\downarrow NO_3^-$; $\updownarrow N_2O$	$\downarrow$fertiliser costs; $\uparrow$technology; $\uparrow$spread lands required (manure)	Chambers *et al.* 2000; Cuttle *et al.* 2006
Fertiliser type / additives	a) Non-nitrate based fertilisers (e.g. urea over NH_4NO_3)	$\downarrow NO_3^-$; $\downarrow N_2O$; $\uparrow NH_3$	$\downarrow$fertiliser costs	Oenema *et al.* 1998; Sommer *et al.* 2004
	b) Organic vs. mineral fertilisers	$\updownarrow NO_3^-$; $\updownarrow N_2O$	fertilisation timing and rate crucial factors	Jones *et al.* 2007; López-Fernández *et al.* 2007
	c) Use of slow release fertilisers	$\downarrow NO_3^-$; $\downarrow N_2O$; $\downarrow NH_3$	$\uparrow$technology; possible $\uparrow$costs	Boeckx *et al.* 2005
	d) Use of urease and/or nitrification inhibitors	$\downarrow NO_3^-$; $\downarrow N_2O$; $\downarrow NH_3$	$\uparrow$costs; $\uparrow$yield	Zaman *et al.* 2008
	e) Use of biochar (biomass derived black carbon)	possible $\downarrow NO_3^-$; $\downarrow N_2O$	$\uparrow$soil physical properties; possible $\uparrow$yield; $\uparrow$technology; more research needed	Lehmann *et al.* 2003; Yanai *et al.* 2007
	f) Biological N fixation	$\updownarrow NO_3^-$; $\downarrow N_2O$; $\downarrow NH_3$	$\downarrow$fertiliser costs	Velthof *et al.* 1998; Berg-ström and Kirchmann 2004
Application method	Fertiliser / manure application method (injection, trailing shoe, band spread, incorporation into soil)	$\downarrow NH_3$; $\updownarrow NO_3^-$; $\updownarrow N_2O$	$\uparrow$technology; $\uparrow$spreading cost; $\downarrow$fertiliser cost	Malgeryd 1998; Chambers *et al.* 2000
Livestock management				
Grazing density	a) Reduction of stocking rates	$\downarrow NO_3^-$; $\downarrow N_2O$; $\downarrow NH_3$	$\downarrow$yields	Di and Cameron 2002

Table 1 (Cont.)

Category	Mitigation measures	Effect on N species	Impact on farm system	Reference
	b) Increase of productivity per animal + decrease in number of animals	↓NO_3^-; ↓N_2O ↓NH_3	↑technology; possible ↑cost	Oenema *et al.* 1998
Timing	Reduction in grazing day and/or season duration	↓NO_3^-; ↓N_2O; ↑NH_3	↑storage and housing costs	Velthof *et al.* 1998; van Groenigen *et al.* 2008
Other	Positioning of feed and water troughs away from water ways	slight ↓NO_3^-; ↓N_2O	↑cost; ?practicality	Cuttle *et al.* 2006
	Manipulation of animal diet to increase N use	↓NO_3^-; ↓N_2O; ↓NH_3	↑technology; possible ↑feed cost	Velthof *et al.* 1998; van Groenigen *et al.* 2008
	Choice of grazing system (beef / dairy / sheep)	possible ↓NO_3^-; ↓N_2O	change in farm system; possible ↑cost	Di and Cameron 2002
Manure management				
Storage	a) Increase in manure storage capacity	↓NO_3^-; ↓N_2O; ↕NH_3	↑cost	Shepherd and Chambers 2007
	b) Low NH_3 emission housing	↓NH_3; ↓N_2O	↑housing cost	Mosquera *et al.* 2005
	c) Reduction of effluent waste volume	↓NO_3^-, ↓N_2O; ↓NH_3	↑housing / structural costs	Shepherd and Chambers 2007
	d) Composting or batch storage of solid manure	↓NO_3^-; ↑N_2O; ↑NH_3	↑housing cost	Cuttle *et al.* 2006
	e) Export of manure to neighbouring farms	↓NO_3^-; ↓N_2O	↑transport and storage cost	Shepherd and Chambers 2007
	f) Incinerating poultry litter	↓NH_3; ↓NO_3^-; ↓N_2O	economic feasibility	Shepherd and Chambers 2007
Manure type	Use of solid manure over slurry	↓NO_3^-; ↓N_2O; ↑NH_3	↑housing cost	Cuttle *et al.* 2006
Location of manure heaps	a) Away from waterways and field drains	↓NO_3^-; ↓N_2O	slight ↑transport cost	Cuttle *et al.* 2006
	b) On concrete and collect the effluent	↓NO_3^-; ↓N_2O	↑storage cost	Cuttle *et al.* 2006

livestock and manure management practices, the effect of each measure on emissions of different N species, and the consequences their introduction might have for farmers and farm systems. Often the identification of adequate mitigation options is not sufficient as implementation can be difficult. With regard to the implementation, two approaches have been identified: (i) the establishment and introduction of generic, national minimum standards and measures, and (ii) the implementation of targeted measures, which are based on risk assessment exercises. Often, high-risk areas can be identified within a catchment, which contribute proportionately more to emissions than their spatial extent would suggest. Focusing primarily on these high-risk areas can increase the success of mitigation and improve the cost/benefit ratio of implementation. In addition, it is difficult to assess the impacts and effectiveness of mitigation measures solely by measuring water or air quality due to difficulties associated with detecting small changes against inherently large background variability, distinguishing the effects of external factors and the long response times (ranging from decades to centuries in groundwater or groundwater-fed surface waters). The large variety of mitigation measures, their interdependency, which exists in many cases, different efficacies and difficulties with regard to implementation and assessment of efficacy clearly illustrate the complexity of mitigation in general and of selecting a strategy appropriate for a specific environment or site in particular.

At the land management scale, the main means to reduce N loss to the environment are improving N uptake, enhancing denitrification and land use change. Increasing N uptake from agricultural soils will capture N before it is lost by leaching or run-off, which can be achieved through, for example, the use winter or undersown cover crops, establishment of buffer zones and/or hedgerows, or improvement of irrigation management to more accurately meet crop demand and control soil moisture fluctuations. Moreover, good management practice, such as establishing and maintaining productive swards on grassland farms and reducing soil compaction, will contribute towards maximising crop N uptake and create unfavourable conditions for denitrification in soils thus increasing soil mineral N content and reducing the likelihood of gaseous emissions. Another important strategy to improve N uptake and reduce both N_2O and NO_3^- emissions from agricultural systems is nutrient (or fertiliser) management, as part of which crop N demand is predicted with regard to quantity, timing and spatial variation and matched by appropriate fertilisation. Successful and sustainable nutrient management should also include the selection of appropriate fertiliser types. Changing from nitrate-based fertilisers to urea, for example, has been shown to reduce N_2O and NO_3^- emissions but also has the potential to increase NH_3 emissions depending on application method and timing (Oenema *et al.* 1998). Moreover, life cycle assessment revealed that putting more emphasis on biological as opposed to industrial N fixation greatly reduces N loss potential by eliminating or limiting emissions during manufacture and transport of inorganic fertilisers (Velthof *et al.* 1998). Nutrient management strategies must also account for N content in soils and organic sources, such as crop residues, animal and green manures, and soil mineralisation potential, which is related to inherent soil properties and past land management. The development of nutrient budgeting along with econometric decision support tools has aided farmers with the implementation of whole-farm nutrient management systems. Nonetheless, it is necessary to enhance existing tools and models by including other, non-nutrient, management measures to mitigate nutrient loss and address cost/benefit issues of the mitigation options.

Increasing denitrification activity in waterways aims to reduce NO_3^- levels by allowing drainage systems to deteriorate (Cuttle *et al.* 2006); reinstating wetlands and riparian zones (Hoffmann and Baattrup-Pedersen 2007); and adding C substrates to drains or groundwater permeable reactive barriers (Fenton *et al.* 2008). However, enhancing denitrification also increases development and evolution of N_2O; consequently, this mitigation option may still lead to pollution (i.e. in gaseous form) under conditions that do not allow for complete denitrification to N_2. Changing agricultural and land use practices from high to low emission systems by, for example, converting from arable to extensive grassland farming has been shown to significantly reduce N loss to the environment (Lord *et al.* 1999). The implementation of this option can be costly, as it often results in lower financial returns. Government subsidies or international financial support may well be required to make it an attractive and feasible option for land managers. Nonetheless, land use change should be considered as a mitigation strategy for selected fields in high-risk areas, such as critical source areas for N_2O emissions or NO_3^- leaching. Within livestock enterprises, N loss can be mitigated by adjusting livestock management, manure storage and manure application strategies. The most straightforward approach to reduce N emissions is by decreasing stocking rates as proposed in the EU Nitrates Directive but this comes at an economic cost for farmers. When lower stocking rates are coupled with increased productivity, on the other hand, per animal emissions can be reduced successfully per unit product (e.g. L of milk or kg of meat), which could be a more sustainable and acceptable approach (Oenema *et al*. 1998). Livestock dietary manipulation can also improve N use efficiency by animals and alter or reduce N excretion lowering N losses to the environment (van Groenigen *et al*. 2008). Low-protein forage has been shown by Luo *et al*. (2008) to be an effective mitigation option, when comparing low-protein maize with pasture silage with regards to N_2O emissions in milk production systems. Reducing the duration of grazing per day and/or season also has a significant effect on the quantity of N excreted and can limit N input at high-risk times. Extending housing periods for livestock means that more animal waste is collected and stored prior to land application. This enables more accurate and appropriate timing of land spreading manures in accordance with crop requirements and during low risk periods. In addition, livestock housing offers the potential to minimise NH_3 loss from manures by manipulating housing and storage facilities, which can, however, lead to high inorganic N levels in the manure and thus pose the risk of N leaching when applied to the field (Mosquera *et al*. 2005). In contrast, composting solid manures decreases mineral N content and NH_3 emissions during spreading; however, more NH_3 is produced during compost production. Such systems also require greater use of buildings and machinery, which will have their own environmental effects. Certain manure application methods are known to reduce NH_3 emissions, e.g. shallow injection or trailing shoe (Malgeryd 1998). While these techniques reduce NH_3 loss and increase soil N availability, thus reducing synthetic fertiliser requirements, they may lead to an increase in N_2O emissions (van Groenigen *et al*. 2008).

In the Netherlands, an experimental dairy farm (De Marke, Hengelo; http://www.projectdemarke.nl) was established during the 1990s to combine environmentally sound farming with production agriculture. The De Marke system uses a wide range of mitigation strategies, including fertilisation management, grazing day length and season, crop rotation, manure management, animal diet and stocking rates. In a simulation study, this system has been shown to reduce N surplus from 293 to 138 kg ha^{-1} and increase N utilisation from 23 to 36% compared with standard commercial practice (Rotz *et al*. 2005). This approach indicates that by combining mitigation measures, it is possible to increase N use efficiency and reduce N loss to the environment.

While the mitigation of N loss to water has received plenty of research and practical attention, the implementation of gaseous N mitigation measures is still in its infancy even though a wide range of options has been identified (**Table 1**). The main constraints for implementation are (i) the complex and often poorly understood control of gaseous N sources in animal production systems; (ii) the limited knowledge on the economic and social costs involved; and (iii) the past research and current policy bias towards mitigating NO_3^- and NH_3 losses. This indicates the importance of identifying cost effective, integrated measures that simultaneously reduce N loss to water and emissions of gaseous N compounds to the atmosphere.

The N cycle has been described as a "leaky pipe" (Davidson and Mosier 2004) whereby controlling one leak can cause or increase leaks in other parts of the cycle. The effect of mitigation measures on all N turnover processes and pathways must be considered as well as their effect on other environmentally sensitive compounds, such as P, C, pesticides, pathogenic microorganisms and soil erosion. It has been shown, for example, that measures that decrease NO_3^- leaching may increase NH_3 emissions or soluble P losses (Harris and Catt 1999). Another less discussed aspect in the debate about N loss is the farmer's economic interest to comply with the regulations and reduce N losses from agricultural systems. Any excess fertiliser- or manure-N that is lost by either leaching or volatilisation represents a financial loss and shows a flawed fertilisation strategy as well as poor N use efficiency. As well as being a burden on the environment, these losses potentially represent an expensive waste to the farmer when fertiliser costs are high. Unfortunately, the introduction of mitigation measures is often associated with an increase in costs or a reduction in returns. The De Marke study mentioned a decline in income per cow by about 20% after implementation of the mitigation plan (Rotz *et al*. 2005). It is worth noting, however, that financial returns are directly tied to the costs of inputs, which means that increasing N fertiliser prices might make currently uneconomic mitigation measures profitable. This shows the critical importance of having a fully integrated approach across all scientific disciplines working with agriculture, from the most fundamental aspects of plant science and biogeochemistry, through agronomics, economics and sociology.

SUMMARY

Increased human activity over the last century, particularly industrialisation, modernisation and intensification of farming practices, have directly, e.g. through fertiliser and manure applications, or indirectly, e.g. through atmospheric N deposition, increased anthropogenic N inputs into aquatic and soil habitats. Nitrogen availability in water and soil environments has increased to damaging levels, leading to negative effects on climate, human health and the environment. Of all human-derived N emissions, agriculture is the most significant single cause for current high N levels in the biosphere. The environmental consequences of excess N are considerable and long-term and, in particular in the case of highly mobile N gases, their potential to create positive feedback effects may further exacerbate their environmental and societal impacts. The introduction of legislation has increased awareness and pressured societies to reduce N losses to acceptable levels. Initially, national and international legislation was introduced to monitor and maintain standards for drinking water quality, but over recent decades the damage excess N compounds cause to the environment has been increasingly recognised. This is reflected in new thresholds for ground- and surface water N concentrations, which have evolved from simple parameter limits to more complex integrated management targets, as enshrined in the US Clean Water Act and the EU Water Framework Directive. Environmental N legislation now integrates multiple water quality parameters, which include measures of hydrological, hydrogeological, hydromorphological and biological properties as well as their interactions. In Europe, the implementation of N legislation has led to significant decreases in aquatic N levels; however, the timeframe envisioned for changes to take effect is often too short and the focus on single environments, such as water or air, is too simplistic. In addition, the overall impact and interaction of mitigation measures must be considered as they may simultaneously have positive and negative effects on different N species and even impact on the release of other nutrients, such as C or P, or pathogenic microorganisms. There is increasing need for integrative legislation addressing N loss as a holistic, global problem in order to decrease the impact of all polluting nutrients on above and below ground systems.

The introduction of environmental legislation, however, often treats the symptoms rather than the cause. Although not the sole source for excess N in the environment, agriculture is one of the largest anthropogenic contributors to climate change, atmospheric N deposition and to the deterioration of waterways through emissions of N gases and

NO_3^- leaching, respectively. The complex nature of agricultural systems, which are dynamic, interdependent and have a strong socio-economic component, shows the intricacy and multi-dimensionality of the N problem. Tackling this issue needs a multifaceted approach and the active involvement of land managers, especially, farmers, who the legislation is aimed at. Land managers need to understand the purpose and origin of environmental quality targets to be able to accept them, especially if they have to implement changes that negatively affect them. For N reduction strategies to be successful, it is essential for researchers and policy maker to recognise and appreciate this fact and not to disregard the inherent knowledge that land managers have of their land. It is important to custom-tailor packages of mitigation measures for each farm or even field, and integrate land managers in the legislative decision making progress. Flexibility is crucial when dealing with an environment as complex as a farm system and a problem as dynamic and intricate as N loss to the environment, where a 'one size fits all' approach will not achieve much. Nor should the fact be discounted that land managers might be equally interested in reducing N losses from the farm system, once they understand the issues, as any N lost is not available to the crop. Thus, N loss not only constitutes a legislative and environmental problem but can also be a financial loss.

The large number of long-standing and new problems caused by N loss to the environment in solution or gaseous form, as well as the development and recent introduction of water and environmental quality legislation clearly show the continued importance and urgency of managing N loss from agricultural systems. Sustainable agricultural production must meet the growing worldwide demand for agricultural produce, while also commensally reducing environmental pollution, i.e. this is not a Cartesian dichotomy. It is vital to achieve both sustainable food production and environmental protection; failure to achieve one task, or to achieve one at the expense of the other, would be a failure. Economically, socially and ecologically sustainable and acceptable management requires researchers, policy makers and land managers to jointly identify and develop practical and feasible solutions to reduce N loss and limit the environmental impact of N emissions. The wide range of mitigation measures currently available coupled with innovative application of techniques and interdisciplinary approaches will help to achieve this objective.

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