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Emission of nitrous oxide in Denmark

Sources related to agriculture and natural ecosystems

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Abstract

The present increase in concentration of atmospheric nitrous oxide (N_2O) of 0.2-0.35% yr^{-1} has caused much concern, since nitrous oxide contributes to the greenhouse effect and to the destruction of stratospheric ozone (O_3). The emissions in Denmark of nitrous oxide from natural and agriculture-related sources have been estimated.

It was found that present emissions from these sources are high above unperturbed natural emissions. Cultivated land is the most significant contributor of nitrous oxide. But the widespread use of N-fertilizers has profound effects on total emission through elevated emission from cultivated land and elevated emission from aquatic systems due to the fertilizer induced eutrophication.

It seems unrealistic to reduce the present emission to natural levels, but a significant reduction is possible if a reduction, and perhaps also a change, in N-fertilizer use is implemented. Further research in the potential of natural and agriculture-related sources is needed, if we are to understand the size and the dynamics of these sources.

Preface

Scope

The much-debated greenhouse effect has caused concern among policy-makers as well as the public, since atmospheric concentrations of the so-called "greenhouse gases" are increasing and are expected to have profound effects on future global climate.

Although less known for its influence on the greenhouse effect than e.g. carbon dioxide (CO_2) nitrous oxide (N_2O) is indeed a very potent greenhouse gas which can not be ignored. In order to judge its effects it is essential not only to determine the direct anthropogenic emissions, but also the emission from natural ecosystems and human influence on the emission from these systems.

Funding

The Ozone and Climate Committee of the Nordic Council of Ministers has initiated a study to establish budgets of greenhouse gases in the Nordic Countries. The present report is a background report for the Danish contribution to this study, which has been coordinated by the National Environmental Research Institute. The work on this report has been financed by the National Environmental Research Institute.

*Structure of
the report*

The report reviews the studies on emissions of nitrous oxide from agriculture and from natural ecosystems and it attempts to establish emission rates from these sources in Denmark. The emission rates are discussed briefly. As far as possible it has been attempted to estimate the human influence on the emission rates from natural ecosystems.

Also included in the report is introductory chapters on atmospheric and microbial dynamics of nitrous oxide (Chapters 2 and 3).

*Acknowledge-
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Linda West turned the manuscript into a report and made it ready for printing. Jes Fenger inspired and encouraged. Many persons and institutions kindly assisted with valuable information and are quoted in the text.

1. Introduction

Increases in atmospheric concentration

At present the atmospheric concentration of nitrous oxide (N_2O) is increasing at 0.55-1.05 parts per billion volume (ppbv) or 0.20-0.35% per year (Weiss, 1981; Khalil, Rasmussen, 1983, 1986, 1988; Bolle *et al.*, 1986; Brunke *et al.*, 1990; Prinn *et al.*, 1990; Rohde, 1990).

Analysis of air bubbles in polar ice cores suggest that the concentration of nitrous oxide over the past few hundred years must have increased from a pre-industrial level of 270-290 ppbv to the current concentration of about 310 ppbv (Bolle *et al.*, 1986; Pearman *et al.*, 1986; Etheridge *et al.*, 1988; Khalil, Rasmussen, 1988; Zardini *et al.*, 1989).

It is essential to note that the predominant rise of the atmospheric concentration of nitrous oxide is observed during this century (Pearman *et al.*, 1986; Khalil, Rasmussen, 1988), which is indicative for the influence of anthropogenic activities as sources for the increased concentration.

The presently observed rate of increase of atmospheric nitrous oxide may seem small. However, the lifetime (or residence time) of nitrous oxide in the atmosphere is relatively long (100-200 years) (Khalil, Rasmussen, 1983; Bolle *et al.*, 1986; Cicerone, 1987; Khalil, Rasmussen, 1988; Zardini *et al.*, 1989; Prinn *et al.*, 1990; Rohde, 1990) and the observed 0.2-0.35% increment imply that total global sources of nitrous oxide are now elevated by some 20 to 30% over unperturbed steady-state sources of about 13-14 Tg $\text{N}_2\text{O-N yr}^{-1}$ (Kaplan, 1984; Dickinson, Cicerone, 1986; Khalil, Rasmussen, 1988; Bouwman, 1990).

The increasing flux from industrial combustion along with land use changes especially in the tropics, increased use of N-fertilizers, and increased biomass burning probably account for most of the observed increase in atmospheric concentration (Weiss, 1981; Bolle *et al.*, 1986; Hao *et al.*, 1987; Bouwman, 1989; Prinn *et al.*, 1990).

Major global sources

The emission from cultivated soils (2-4 Tg $\text{N}_2\text{O-N yr}^{-1}$, which includes an estimated increase in emission of 0.6-2.3 Tg $\text{N}_2\text{O-N yr}^{-1}$ resulting from the application of mineral fertilizers in agriculture), natural soils (3-9 Tg $\text{N}_2\text{O-N yr}^{-1}$), fossil fuel burning (1 Tg $\text{N}_2\text{O-N yr}^{-1}$), oceans (including estuaries) (1-4.4 Tg $\text{N}_2\text{O-N yr}^{-1}$) and biomass burning (1-2 Tg $\text{N}_2\text{O-N yr}^{-1}$) at present form the major sources in the global nitrous oxide budget (Kaplan, 1984; Schmidt *et al.*, 1988; Zardini *et al.*, 1989; Bouwman, 1990).

These estimates are still highly uncertain. E.g. a 7-8 Tg $\text{N}_2\text{O-N yr}^{-1}$ has been suggested for tropical wet and moist forests alone (Kaplan, 1984); the measurements made so far on fossil fuel emissions might be incorrect (Muzio, Kramlich, 1988; Bouwman, 1990). Further Bouwman (1989) gives much wider ranges when estimating global nitrous oxide emissions from cultivated soils (0.3-28 Tg $\text{N}_2\text{O-N yr}^{-1}$) and natural soils (11-27 Tg $\text{N}_2\text{O-N yr}^{-1}$).

Variations in fluxes

Estimates of regional and global emissions of nitrous oxide are impeded by the extreme spatial and temporal variations in measured fluxes and the currently used measurement techniques, often involving small flux-chambers, which make the extrapolation of these fluxes a difficult task. Moreover, emission rates from natural ecosystems are severely underrepresented in existing data.

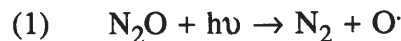
This study will only consider natural and agriculture-related sources of nitrous oxide in Denmark. All nitrous oxide emission rates are given as weight of nitrogen-nitrous oxide (N-N₂O) and not as total weight of nitrous oxide (N₂O).

2. Atmospheric dynamics of nitrous oxide

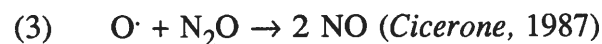
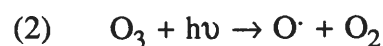
Nitrous oxide plays important roles in the chemistry of the atmosphere and in the radiation budget of the Earth. A continued increase in the atmospheric burden can be expected to have a significant impact on stratospheric ozone (O₃) and global climate.

Atmospheric reactions

Because nitrous oxide is inert in the troposphere (*Banin, 1986; Cicerone, 1987*), it can be transported into the stratosphere where most of it is decomposed through:

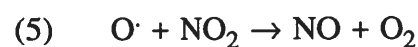
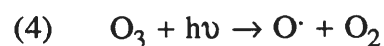


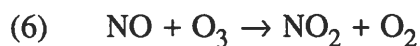
About 5% of the nitrous oxide is photolytically oxidized to nitric oxide (NO) via:



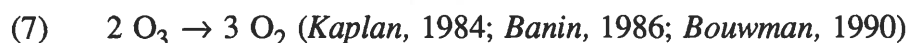
The total atmospheric destruction of nitrous oxide has been estimated to be about 10.5 Tg N₂O-N yr⁻¹ (*Bouwman, 1990*).

Nitric oxide is a much more reactive gas and catalyses the conversion of ozone to molecular oxygen (O₂) in the stratosphere according to the reactions:





With the net result:



Impact on ozone

The stratospheric ozone is located between 10 and 40 km altitude. Below 25 km ozone is formed under the influence of different nitrogenous oxides (NO_x) and in this 10-25 km region of the stratosphere NO_x thus counteracts the above-mentioned set of reactions (4) - (7). Above 25 km the NO_x additions from nitrous oxide decrease the ozone concentration, whereas below 25 km NO_x protects ozone from destruction (Bouwman, 1990).

Ozone is an important component of the stratosphere. It serves as a shield against biologically harmful solar ultraviolet (UV) radiation, initiates key stratospheric chemical reactions, and transforms solar radiation into the mechanical energy of atmospheric winds and heat (Cicerone, 1987). Banin et al. (1984) have estimated an average decrease of 1.5% in ozone column per 10% increase in nitrous oxide concentrations.

Infrared absorption bands

Besides its important roles in stratospheric chemistry, nitrous oxide has strong infrared (IR) absorption bands at 4.5, 7.78, 8.56 and 17 micrometer (Wang, Sze, 1980; Banin, 1986; Mitchell, 1989) and thus contributes to the greenhouse effect. Nitrous oxide is also the precursor to e.g. stratospheric nitric acid (HNO_3) which has IR absorption bands at 5.9, 7.5, 11.3, and 21.8 micrometer (Wang, Sze, 1980). Consequently, changes in nitric acid may also affect the global radiation budget.

Warming potential

Because greenhouse gases have different lifetimes, the relative cumulative impact of each mole of gas may be quite different from its relative initial impact. Therefore, the relative warming potential of given greenhouse gas emissions must be considered, rather than the relative radiative forcing of changes in atmospheric concentrations. Nitrous oxide has an instantaneous impact, 250 times greater than carbon dioxide (CO_2) (2.8 and $0.015 \text{ Wm}^{-2} \text{ ppm}^{-1}$, respectively, Lashof, Ahuja, 1990). The decay of nitrous oxide in the atmosphere roughly follows a simple exponential relation. However, this does not apply to carbon dioxide. Thus, due to their differing decay-functions the cumulative effect of nitrous oxide compared to carbon dioxide depends on the time period. Assuming a hundred years time period the atmospheric lifetime is 120 years, compared to the lifetime of nitrous oxide of 100-200 years (Rohde, 1990). At this time period the cumulative impact (and global warming potential) of nitrous oxide is about 300 times greater than for carbon dioxide.

In the future, the atmospheric lifetime of nitrous oxide could decrease if the ozone concentration decreases above 30 km altitude because increased UV radiation just below 30 km would increase the rate of nitrous oxide photolysis (*Ramanathan et al.*, 1985). This would lower the relative impact of nitrous oxide. The present contribution of nitrous oxide to global warming relative to other greenhouse gases has been estimated to 3.1% (*Lashof, Ahuja*, 1990), 4% (*Bouwman*, 1990), and 5% (*Rohde*, 1990).

Future increases

The papers consulted during this study leave no doubt that the concentration of atmospheric nitrous oxide will undergo further increases with time because of increasing rates of fossil fuel burning and application of mineral fertilizers. This is also due to the fact that atmospheric nitrous oxide concentrations are not yet in equilibrium with the strength of the global sources, even if these are constant because of the long lifetime of nitrous oxide.

The concentration has been estimated to increase to 320-330 ppbv by year 2000 (*Dickinson, Cicerone*, 1986), to 360-390 ppbv by year 2030-35 (*Khalil, Rasmussen*, 1983; *Ramanathan et al.*, 1985, 1987; *Khalil, Rasmussen*, 1988; *Mitchell*, 1989), 350-450 ppbv by year 2050 (*Dickinson, Cicerone*, 1986; *Hao et al.*, 1987), and 440-1430 ppbv by year 2100 (*Ramanathan et al.*, 1987).

Effects of future increases

Some authors have tried to estimate the effect on global climate by future increased nitrous oxide levels. *Wang and Sze* (1980) suggests that a doubling of present levels might contribute 0.44K to global warming plus an additional 0.23K resulting from the associated climatic feedbacks from ozone and nitric acid perturbations. *Khalil and Rasmussen* (1983) and *Ramanathan et al.* (1985, 1987) have estimated that an increase of 25% in atmospheric nitrous oxide (which is reached in 50 years at present rates of increase) will cause a global average temperature rise of 0.1K-0.6K.

The total effect of the different greenhouse gases is expected to cause an increase in the average global temperature of 0.8 to 4.5K some time around the middle of the next century (*Dickinson, Cicerone*, 1986; *Ramanathan et al.*, 1987). When global warming occurs as a result of the greenhouse effect there is a potential for additional increases of nitrous oxide emissions from biogenic sources due to the effects of changes in soil and water temperature and of soil moisture status on microbial metabolism.

A warmer temperature regime would increase biological activities leading to greater production of nitrous oxide from soils and water systems. Using a general circulation model (GCM) *Turner et al.* (1990) found that for the Pacific Northwest region of the United States denitrification and thus potential nitrous oxide emissions were 1.7 times greater when simulating a climate with a doubled CO₂ concentration which was 4.9K warmer and 3.5% wetter than present climate.

Melting of the upper layers of permafrost in the high arctic could add more nitrous oxide to the atmosphere. The combined response of wetlands and permafrost to increased temperature may add as much or more nitrous oxide to the atmosphere than expected from the increasing anthropogenic sources (*Khalil, Rasmussen, 1989*).

A potential positive feedback mechanism between emission rates of biogenic nitrous oxide and global warming is supported by analysis of air bubbles in polar ice cores which have shown that the nitrous oxide concentration was lower during periods of global cooling (*Khalil, Rasmussen, 1989*).

The global warming that has apparently occurred may already have produced about 20% of the increase of nitrous oxide between pre-industrial times and the present (*Khalil, Rasmussen, 1989*). As increases in nitrous oxide flux from soils and water systems will usually be small relative to the total pool of soil nitrogen, the increase in flux of nitrous oxide could probably last for decades or centuries.

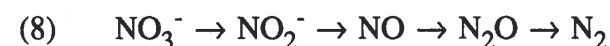
3. Microbial processes and nitrous oxide

Several enzymatic processes involve nitrous oxide. Two microbial processes, biological denitrification and nitrification, are regarded as the main sources for nitrous oxide. Biological denitrification is also a sink for nitrous oxide since nitrous oxide is a free intermediate of the reaction whereas the role of nitrification as sink for nitrous oxide sink is unknown.

3.1. Biological denitrification

Biological denitrification is the dissimilatory reduction of nitrate (NO₃) and nitrite (NO₂) to gaseous forms of nitrogen by facultatively anaerobic bacteria producing molecular nitrogen (N₂) or oxides of nitrogen, like nitrous oxide, when oxygen is limiting.

The general pathway of the reduction of nitrate (and nitrite) during denitrification is:



The energy for these reactions is supplied by the decomposition of carbohydrates.

No universal relationship between gases produced

Denitrifying bacteria are aerobic, but in the absence of oxygen they use nitrate as an electron acceptor for growth. Denitrification occurs only at low oxygen pressures. Most denitrifying bacteria can reduce nitrous oxide to molecular nitrogen. The $N_2:N_2O$ and $N_2O:NO$ ratios in the gas evolved during denitrification is highly variable, and there is no universal relationship between these ratios and total denitrification. Low pH, high nitrate concentration, low availability of organic matter, low soil moisture content are the major prerequisites for a high fraction of nitrous oxide to total denitrification gases, but the relative importance of these parameters are difficult to assess (Sahrawat, Keeney, 1986; Keller et al., 1988; Nommik, Larsson, 1989; Arah, Smith, 1990).

The shift in the $N_2O:N_2$ ratio within soil environments can be very dramatic, and nitrous oxide producing systems can thereby switch into nitrous oxide sinks.

There is also evidence that *Rhizobia* bacteria may induce significant losses of nitrous oxide from leguminous crops and natural ecosystems with leguminous plants (Bertelsen, 1990; Bouwman, 1990). But more work needs to be done to clarify the importance of rhizobial denitrification.

3.2. Chemical denitrification

Chemical denitrification is the reduction of nitrite or nitrate by chemical reductants producing molecular nitrogen or oxides of nitrogen, like nitrous oxide.

Controversial proces

However, there is much controversy as to the importance of chemical denitrification. Chemical denitrification is supposed to occur especially following the addition of ammonium fertilizers and at high nitrite concentrations which have been attributed to the inhibition of bacterial oxidation of nitrite presumably as a result of ammonia toxicity.

Chemical denitrification may be of importance at times in neutral to alkaline soils where nitrite-accumulation is a problem (Sahrawat, Keeney, 1986). However, it has been found that the gas formed consists predominantly of nitric oxide which has led to the conclusion that chemical denitrification usually do not significantly contribute to nitrous oxide production.

Unless otherwise stated the term denitrification will imply biological denitrification in this report.

3.3. Nitrification

Nitrification is the biological oxidation of ammonium (NH_4) to nitrite or nitrate.

It is carried out by highly specialized autotrophic bacteria and by a multitude of heterotrophic microorganisms.

Except for oxygen-deficient systems like poorly drained and submerged soils, ammonia is used as an energy source and is readily converted to nitrate by nitrification:



Occurs under aerobic conditions

Contrary to denitrification, nitrification occurs under aerobic conditions. However, it is often found that the formation rate of nitrous oxide during nitrification increases as the oxygen concentration decreases (*Klemetsson et al.*, 1988).

The chemical pathways leading to nitrous oxide production during nitrification are not clearly understood. Nitrite is most likely used as an alternative electron acceptor at low oxygen concentrations, resulting in the evolution of nitrous oxide (*Poth, Focht*, 1985; *Sahrawat, Keeney*, 1986).

Minami, Fukushi (1986) suggest that in well-aerated soils hydroxylamine may react with nitrite forming nitrous oxide. However, this reaction is not as important as nitrite reduction for nitrous oxide production during nitrification (*Bouwman*, 1990).

In well-aerated soils emissions of nitrous oxide are greatly increased by the addition of nitrifiable forms of nitrogen such as ammonia and urea (*Breitenbeck et al.*, 1980; *Breitenbeck, Bremner*, 1986b).

Traditionally, denitrification has been considered the main source of nitrous oxide emission from different ecosystems and it is generally assumed that nitrification is of limited importance in permanent grasslands and forest soils receiving no fertilizers (*Richards*, 1987; *Robertson, Tiedje*, 1987). However, studies in agricultural ecosystems indicate that nitrification can contribute significantly (e.g. *Bremner, Blackmer*, 1981; *Robertson, Tiedje*, 1984; *Cates, Keeney*, 1987) and might even be dominant over denitrification until soils become very wet (*Parton*, 1988). It may also represent a major source of nitrous oxide in marine environments (*Elkins et al.*, 1978).

3.4. Environmental factors controlling nitrous oxide forming processes

It is extremely difficult to predict the amount of nitrous oxide produced in an environment for two reasons: 1) nitrous oxide is a product as well as a substrate for the microbial processes, and 2) the shift of nitrous oxide from product to substrate can occur by minute changes in highly variable parameters such as easily available carbon or soil water contents.

Oxygen and soil water status

Denitrification in soils is negligible at soil moistures below 2/3 of the water holding capacity, but the process may occur in anaerobic microsites within an otherwise aerobic soil (*Parkin, 1987; Bouwman, 1990; Christensen, 1990*). In aquatic systems the sediments are usually anaerobic and thus holds good conditions for denitrification.

A tendency for increased nitrous oxide release has been observed with improved aeration status of soils. This might be caused by a slowdown of the denitrification process (allowing more nitrous oxide to escape unreduced) or by stimulation of nitrification of ammonia whereby nitrous oxide may be one of the products.

The release of nitrous oxide from soils is stimulated by alternating drying and wetting cycles. When the soil is wetted conditions for denitrification are favoured and nitrous oxide will be produced more rapidly than it is reduced. Irrigation and precipitation are often followed by rapid increases in nitrous oxide emissions (*Ryden, Lund, 1980; Mosier, Hutchinson, 1981; Mosier et al., 1982; Goodroad et al., 1984*). If the soil dries fast enough, nitrous oxide reduction to molecular nitrogen is prevented.

Continuous flooding as found in aquatic environments makes molecular nitrogen the main product of denitrification with little nitrous oxide production (*Denmead et al., 1979; Smith et al., 1982; Goodroad, Keeney, 1986a; Sahrawat, Keeney, 1986; Van Breemen, Feijtel, 1990*).

Soil texture can also influence the extent of anaerobic sites in a soil. Compared to e.g. a sandy soil a heavy clay soil will contribute more to the development of anaerobic conditions thereby giving higher rates of denitrification (*Burford et al., 1981; Nommik, Larson, 1989*).

Temperature

There is a strong diurnal variation of nitrous oxide emission rates (e.g. *Christensen, 1983; Minami, 1987*) and a great seasonal variability (e.g. *Bremner et al., 1980*) partly as a result of temperature changes.

The optimum temperature for the denitrification process is 25°C and above, while the process is slow at 2°C. In nitrification the temperature optimum lies between 30 and 35°C, while below 5°C the activity is very low (*Bouwman, 1990*).

Both the rate of emission and the form of the products of denitrification and nitrification depend on temperature. Important losses of nitrous oxide from soils has been reported at quite low temperatures, like 8°C and large emissions can be observed during spring thaw (*Goodroad, Keeney, 1985; Cates, Keeney, 1987b; Schmidt et al., 1988; Christensen, Tiedje, 1990*).

Nitrate

A high concentration of nitrate often results in a high level of denitrification and also in a high ratio of nitrous oxide to molecular nitrogen as products of the denitrification process (e.g. *Christensen, 1985b*).

Soil organic matter

A good supply of readily decomposable organic matter feeds denitrification with energy and nitrification with ammonia (*Burford et al., 1981; Sahrawat, Keeney, 1986*). Nitrous oxide fluxes from organic soils can be many times higher than in mineral soils (*Duxbury et al., 1982*).

pH

Many of the denitrifying bacteria are sensitive to high hydrogen ion concentrations. However, under certain conditions denitrification may still be rapid at pH 4.7 (*Bouwman, 1990*).

Typically, the rate of nitrification falls off markedly below pH 6.0 and generally becomes negligible below pH 5.0, but some soils nitrify at pH 4.0 due to the presence of acid-adapted strains or chemical differences between the habitats (*Richards, 1987*).

In denitrification the composition of the gaseous products is governed by pH. From acid habitats nitrous oxide frequently makes up more than half of the gases evolved (*Bouwman, 1990*). Virtually no N₂ information is found at or below pH 5 (*Christensen, 1985a; Christensen et al., 1990*). At low pH nitrate also inhibits nitrous oxide reduction to molecular nitrogen but at higher pH this inhibition is only temporary.

The interactions between pH and nitrous oxide emission are very complex and emissions are not always correlated to hydrogen ion concentrations (see e.g. *Weier, Gilliam (1986) and Goodroad et al. (1984)*), though sometimes rates of emission will increase with increases in pH (*Goodroad, Keeney, 1984b*).

Plant cover

Plant growth may stimulate denitrification by providing easily decomposable carbon substrates and by consuming oxygen (*Vinther, 1984; Beck, Christensen, 1987; Klemetsson et al., 1987; Umarov, 1990; Christensen et al., 1991*). On the other hand, plants may reduce denitrification and also the nitrous oxide fraction of the gaseous products by depleting the inorganic nitrogen--pool (*Klemetsson et al., 1987; Bouwman, 1990*).

4. Nitrous oxide uptake by natural areas

Under certain circumstances aquatic systems (Cohen, 1978; Elkins *et al.*, 1978; Chan, Knowles, 1979) and soil systems (Blackmer, Bremner, 1976; Brice *et al.*, 1977; Bremner *et al.*, 1980; Ryden, 1981, 1983) can act as sinks for atmospheric nitrous oxide. This is, however, not considered as globally significant (Knowles, 1985) and nitrous oxide consumption is rarely of such a strength as to cause depletion of nitrous oxide from the surrounding air.

Virtually all denitrifying bacteria have the capacity to reduce nitrous oxide to molecular nitrogen (Blackmer, Bremner, 1976; Knowles, 1985). Ryden (1981) found that the prevailing soil conditions during periods of sink activity in a grassland soil were moderate to high (>20%) soil water content. Even small levels of nitrate seem to suppress nitrous oxide consumption in both aquatic (Chan, Knowles, 1979) and terrestrial systems (Blackmer, Bremner, 1976; Firestone *et al.*, 1979; Ryden, 1981, 1983).

Negligible sink strength

It is very likely that only a negligible area of natural ecosystems in Denmark are actually acting as sinks for atmospheric nitrous oxide.

It is important to bear in mind that even though a system does not act as a sink for nitrous oxide a significant proportion of the nitrous oxide evolved might be consumed by the system itself before entering the atmosphere. It has been observed that denitrifiers can act as a sink for nitrous oxide produced by other sources in the system (Robertson, Tiedje, 1987).

More work needs to be done to establish the importance of nitrous oxide consumption by microorganisms as nitrous oxide produced in subsoils and water columns diffuse to the soil or water surface, and also on the influence of land-use changes and nitrate concentration on nitrous oxide consumption.

5. Nitrous oxide emission from natural areas

Estimates of regional and global emissions of nitrous oxide from soils are hampered by large, both spatial and temporal, variations in measured flows. The size of reported nitrous oxide emissions from comparable soils often range over one or two orders of magnitude.

Besides this, very little work on nitrous oxide fluxes has been carried out on Danish soils. Most fluxes used in the following chapters are from sites with edaphic and climatic conditions which compares fairly well to Danish conditions. Also, levels of nitrous oxide emissions seem to be determined by a complex array of management and environmental factors that can not easily be transferred from one site to another.

Summarizing, emission from soils is a poorly understood source of nitrous oxide. Unlike emission from e.g. combustion of fossil fuels the processes governing the fluxes from soils are like a "black box" which we are only starting to understand. It is noted that all measurements of nitrous oxide emission rates from terrestrial systems have been carried out at the soil surface and hence do not consider any possible production or consumption of nitrous oxide in e.g. forest canopies.

5.1. Cultivated soils

Much of work has been carried out on nitrous oxide emission from cultivated soils in temperate or near-temperate regions (see appendix 1, which summarizes the reports on fluxes from cultivated soils in these regions).

However, as emission rates depend on a complex array of factors like soil structure, pH, climate, crops, C-content of the soil, water status, and amount and kind of N-fertilizer cultivated soils show a wide range of emission rates from -0.6 to 165 kg N-N₂O ha⁻¹ yr⁻¹.

First approach: Amount of applied fertilizer

Besides moisture status of the soil, the single most important factor influencing nitrous oxide emission from cultivated soils seems to be the level of N-fertilizer application. *Bouwman* (1990) found the following mathematical relation between N-fertilizer application and emission rate for croplands in temperate regions, excluding application rates of over 250 kg N ha⁻¹ yr⁻¹ as well as poorly drained soils:

$$(10) \quad \text{N}_2\text{O emission (kg N-N}_2\text{O ha}^{-1} \text{ yr}^{-1}) = 1.8785 + 0.00417 * \text{N-fertilizer level (kg N ha}^{-1} \text{ yr}^{-1})$$

Though the data are more limited a more steep relationship seems to exist between emission rates from grasslands and N-fertilizer application (see *Bouwman* 1990). The relationship is assumed to be double as steep as for ordinary cropland. In Denmark cultivated land (excluding grasslands) covers 2 227 000 ha (*Danmarks Statistik*, 1990). An average of 194 kg N ha⁻¹ yr⁻¹ is applied as fertilizer (135 kg N ha⁻¹ yr⁻¹ as commercial fertilizer (*Danmarks Statistik*, 1990) and 59 kg N ha⁻¹ yr⁻¹ as manure (*Danmarks Statistik*, 1986)). This results in an emission rate of 2.7 kg N-N₂O ha⁻¹ yr⁻¹ and a total emission of nitrous oxide of 6.0 kt yr⁻¹.

Permanent grasslands out of rotation and grasslands in rotation cover 219 000 ha and 328 000 ha, respectively (*Danmarks Statistik*, 1990). Permanent grasslands are assumed to receive 59 kg ha⁻¹ yr⁻¹ of manure on average from grazing livestock. Grasslands in rotation receive a total of

approximately 300 kg N ha⁻¹ yr⁻¹ (Jensen, Reenberg, 1986; Danmarks Statistik, 1990). From permanent grasslands and grasslands in rotation estimated emission rates are thus 2.4 kg N-N₂O ha⁻¹ yr⁻¹ and 4.3 kg N-N₂O ha⁻¹ yr⁻¹ resulting in total emissions of 0.5 kt yr⁻¹ and 1.4 kt yr⁻¹, respectively.

*Second approach:
Loss of fertilizer*

Eichner (1990) has reviewed the literature on N-fertilizer-induced nitrous oxide emissions. She gives an average annual emission from soils planted with grains of 0.0099 kg N-N₂O ha⁻¹ yr⁻¹ per kg N-fertilizer applied. The same value for grasslands is 0.017 kg N-N₂O ha⁻¹ yr⁻¹ kg N⁻¹. Using these data total emission from cropland is 4.3 kt yr⁻¹, from permanent grassland 0.2 kt yr⁻¹, and from grassland in rotation 1.6 kt yr⁻¹, totalling 6.1 kt yr⁻¹.

*Third approach:
Effect of fertilizer type*

A third approach is estimating the total emission from the percentage of applied N-fertilizers lost as nitrous oxide. This value depends on fertilizer type with losses of 2.7% for anhydrous ammonia, 0.44% for ammonium nitrate, 0.25% for ammonium chloride and sulphate, 0.11% for urea, and 0.07% for nitrate (Eichner, 1990). The corresponding value for manure depends on the type of manure (Bremner, Blackmer, 1981), but an estimated general factor of 0.5% seems appropriate.

Galbally (1985) reviewed fertilizer-loss as nitrous oxide and found that each of the emission factors varies by a factor of five around the median value; then estimating nitrous oxide fluxes from fertilizer application is subject to a great deal of uncertainties.

It must be noted that the emission factor for anhydrous ammonia (2.7%) is based on very few reports (Bremner *et al.*, 1981; Breitenbeck, Bremner, 1986a, 1986b) of which only one (Breitenbeck, Bremner, 1986a) compared the loss of anhydrous ammonia with the loss from other types of fertilizer applied to the same soils. It must be noted that Hutchinson and Mosier (1979) found a smaller loss (1.3%) of applied anhydrous ammonia, and that their report is not included in the review by Eichner (1990).

Breitenbeck and Bremner (1986a, 1986b) explain the high nitrous oxide loss from anhydrous ammonia by the method of applying this fertilizer by injection into the soil which produces highly alkaline zones of high ammonium-N concentration that do not occur when urea or aqueous ammonia fertilizers are broadcast and incorporated into the soil. The annual use of commercial fertilizers in Denmark by fertilizer type is: 80.2 kt anhydrous ammonia, 286.9 kt ammonia nitrate, 0.5 kt ammonium chloride and sulphate, 5.2 kt urea, and 4.2 kt calcium, potassium and sodium nitrate (Danmarks Statistik, 1990; Plantedirektoratet, 1990) and about 83.8 kt of manure (Danmarks Statistik, 1986).

Thus, total fertilizer-induced emission from cultivated soils is 3.9 kt yr⁻¹ plus non-fertilizer-induced emission which is around 4.3 kt yr⁻¹ (an emission

factor of $1.6 \text{ kg N-N}_2\text{O ha}^{-1} \text{ yr}^{-1}$ is the average from unfertilized cropland and grassland soils according to appendix 1) totalling 8.2 kt yr^{-1} .

Other contributions

The estimates above have not considered the contribution of nitrogen from precipitation, irrigation and sludge which add an average of $12\text{-}22 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ to the farmland (*Danmarks Statistik et al.*, 1990). This might raise the above total emission rates (first to third approach) by 5-10% resulting in a total emission of nitrous oxide from cultivated soils in Denmark of $6.4\text{-}9.0 \text{ kt yr}^{-1}$.

The median of this range suggests a total emission rate of 7.7 kt yr^{-1} for cultivated land. This value is shown in Table 1. This estimate is highly uncertain, however, and a more appropriate range is $3\text{-}15 \text{ kt yr}^{-1}$.

A lesser known factor in nitrous oxide fluxes from cultivated soils is irrigation. It has been found (for Californian climate, though) that emission from irrigated and fertilized soils can be several times higher than from non-irrigated soils (*Ryden, Lund*, 1980). These very high fluxes are probably connected to the combination of irrigation, very high levels of N-fertilizer application, and a warm climate.

In 1982 400 000 ha, or 14%, of the cultivated land in Denmark had facilities for irrigation and another 25 000 ha, or 1%, is added annually (*Jensen, Reenberg*, 1986). But until more work has been done on the influence of irrigation no estimate on the emission from irrigated soils in Denmark can be given.

Trends

Since 1980 the total area of cultivated land has decreased by about $14\,500 \text{ ha yr}^{-1}$ (*Danmarks Statistik*, 1990) of which $10\,000 \text{ ha yr}^{-1}$ has been given over to infrastructure and municipal development (*Skov- og Naturstyrelsen*, 1988). A decline of $14\,500 \text{ ha yr}^{-1}$ corresponds to an annual decline in nitrous oxide emission from cultivated land of about $0.03\text{-}0.05 \text{ kt yr}^{-1}$. It might be expected that in the coming years even more farmland will be converted to other uses as farmers are inclined to give up less productive areas.

In this century the use of N-fertilizers has increased dramatically, but during the last five years the consumption per ha has remained unchanged (*Danmarks Statistik*, 1990).

5.2. Forest soils

Only little has been reported on nitrous oxide emission from temperate forest soils (see appendix 2).

As nitrous oxide emission from cultivated soils seems to be tightly linked to N-fertilization the same is assumed to apply to forest soils (see *Keller et al.*, 1988). None of the works cited in appendix 2 state what the investigated

forest sites had received any N- fertilizer, but presumably they had not. In Denmark the use of N- fertilizers in forestry is very limited, averaging less than 1 kg N ha⁻¹ (Plum, 1991).

Unlike most cultivated soils which are usually limed, forest soils often have a low pH which is likely to influence nitrous oxide emission. In acid forest soils bacterial nitrification and denitrification are assumed to be restricted by the acidity. In these soils assimilatory nitrate-reducing fungi, which are considered to be of minor importance in agricultural soils, may be the most important nitrous oxide producers (Klemedtsson, Svensson, 1988).

Nitrous oxide production in soil cores from forest sites indicate that nitrous oxide production in forest soils under some circumstances are enhanced by low soil pH (Martikainen, 1985). But too little is known about the effect of pH on the emission of nitrous oxide from forest soils to speculate about the effects of liming forests.

The limited data in appendix 2 can not justify separate values for nitrous oxide emissions for different forest types: the emission rate for the Wisconsin coniferous forest seems higher than for the deciduous sites but this coniferous forest was situated on a fairly fertile soil quite unlike the Danish coniferous forests which are often situated on poor soils.

The average emission rates from the forests listed in appendix 2 are about 1.0 kg N-N₂O ha⁻¹ yr⁻¹. Due to their large ranges the Swedish sites have been excluded from this calculation, the same apply to the Danish waterlogged alder stand which due to its very special environmental conditions can not be considered representative for an average Danish forest.

Trends

At the latest forest survey in 1976, Denmark had 406 000 ha of forest (Danmarks Statistik, 1983). Since 1976 about 1500 ha have been planted with forest each year (Skov- og Naturstyrelsen, 1990) giving an estimated total forest area of about 427 000 ha in 1990. This correspond to a total emission of nitrous oxide from forest soils of 0.4 kt yr⁻¹.

The Danish Ministry for the Environment has put as a goal that the total forested area in Denmark will double within the next 80-100 years (Skov- og Naturstyrelsen, 1988). It is of course uncertain how much forest will actually be planted during the next decades. Estimates ranging from 50 000 to 500 000 ha have been given but 5000 ha yr⁻¹ seems appropriate (Skov- og Naturstyrelsen, 1990). The present 1500 ha of newly planted forest per year correspond to an annual increase in source strength of forest soils of 0.002 kt yr⁻¹.

5.3. Other terrestrial systems

About 70 000 ha of Denmark are covered by heathland and moors while another 7600 ha are covered by natural commons situated on well-drained soils (*Danmarks Statistik et al.*, 1990).

No value for the emission factor from sites comparable to these areas could be found in the literature. But since they are all situated on poor and often well drained soils with a low turn-over of nitrogen it is expected that the emission rates are small. North American prairies might be comparable to these areas with respect to nitrous oxide emission. *Bouwman* (1990) states that temperate forests produce more nitrous oxide than prairies.

Assuming that the nitrous oxide emission rate from heathlands, moors, and commons is half the value for temperate forests then these lands emit about $0.55 \text{ kg N-N}_2\text{O ha}^{-1} \text{ yr}^{-1}$ (which is in accordance with *Galbally* (1985)) giving a total emission of 0.04 kt yr^{-1} .

5.4. Drainage water

An overlooked source of atmospheric nitrous oxide is the degassing from drainage water from cultivated land and disturbed land like newly clear-cut forests.

Very few workers have adressed the potential of nitrous oxide emissions from drainage water, but the scattered reports indicate that drainage water might be an important source of atmospheric nitrous oxide (*Dowdell et al.*, 1979; *Conrad et al.*, 1983; *Bowden, Bormann*, 1986; *Minami, Ohsawa*, 1990).

It has been found that nitrous oxide flux from drainage water from a japanese vegetable field (*Minami, Ohsawa*, 1990) and from english fields during winter and spring (*Dowdell et al.*, 1979) are comparable to or even higher than from the field itself. *Minami, Fukushi* (1984) measured nitrous oxide emission of $17.5\text{-}104 \text{ kg N-N}_2\text{O ha}^{-1} \text{ yr}^{-1}$ in drainage water from cultivated soils.

Minami, Ohsawa (1990) found that the amount of dissolved nitrous oxide in drainage water correlated well with nitrate concentration, which indicates that the source strength of drainage water from cultivated land depends on the amount of applied N-fertilizer.

Bowden, Bormann (1986) found that the export of nitrous oxide by degassing of soil water from a clear-cut forest was similar to the soil-air diffusion from the logged area, but that the emission from drainage water from an undisturbed forest site was insignificant.

The assumption that the amount of nitrous oxide emitted from drainage water seeping from cultivated land or clear-cut forest, is equal to the direct fluxes from soil surfaces of cultivated land and clear-cut forest, makes drainage water a major source of nitrous oxide.

Water from cultivated land

As the fluxes measured by *Dowdell et al.* (1979) cover the season with excess precipitation over evapotranspiration and as *Minami, Ohsawa* (1990) reported fluxes from a Japanese field receiving $500 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ as fertilizer, a much lower total emission from drainage water from cultivated soils in Denmark is expected. It seems more likely that drainage water from cultivated land emit 5-50% of the nitrous oxide emission from the soil surface of cultivated soils, giving a total emission of 0.35 to 3.5 kt yr^{-1} .

Drainage water from clear-cut forests

Unfortunately, data on the area of forest that is annually clear-cut are not easily available (*Hedegaard*, 1991). But as e.g. the Danish beechwood forests are rarely clear-cut the annual area of clear-cut forest is probably only 1% of the total forest area amounting to around 4000 ha. Drainage water from clear-cut forest sites thus seems to be a negligible source of nitrous oxide in Denmark.

5.5. Groundwater

Ronen et al. (1988) found that groundwater contaminated by anthropogenic activities can contain high levels of nitrous oxide. *Jacobsen and Vinther* (1990) also found very high concentrations of nitrous oxide in a cultivated sandy soil at just above the saturated zone. However, it is not known how much of this nitrous oxide will eventually be released to the atmosphere through 1) springs and streams that are groundwater outlets, 2) through marine environments where groundwater discharges, and 3) as a result of the use of groundwater on the land surface for municipal and agricultural purposes. Alternatively, the nitrous oxide will be biologically converted to molecular nitrogen.

Based on this scanty evidence it is not possible to estimate the release of nitrous oxide from groundwater to the atmosphere in Denmark.

5.6. Freshwater systems

Due to the waterlogged conditions in sediments of freshwater systems the potential for denitrification is generally high and apparently denitrification is paramount to nitrification as a source of nitrous oxide in these systems. However, the nitrous oxide emission is usually small compared to terrestrial

systems since denitrification is limited by low nitrate concentrations in the anaerobic zone and furthermore mainly produces molecular nitrogen at these conditions.

The literature only gives very little information on nitrous oxide fluxes from freshwater systems (appendix 3) and emission rates range between 0.1 and 2.7 kg N-N₂O ha⁻¹ yr⁻¹.

Nitrogen loading

The values given in appendix 3 are too few to allow any reliable estimate of the nitrous oxide flux from Danish freshwater systems. But a different approach seems possible. As the nitrous oxide emission from these systems depend on the nitrogen loading the data compiled by *Seitzinger* (1988) can be used to estimate the nitrous oxide emission.

The Danish streams, lakes, and freshwater marshes annually receive 140-145 kt N (1984-figure) from cultivated land, farms and other anthropogenic sources (*Danmarks Statistik*, 1990). The rivers and the Danish lakes referred to by *Seitzinger* (1988) removes 7-35% of the nitrogen input by denitrification. The ratio of N₂O:N₂ emission from freshwater systems are determined by e.g. oxygen and hydrogen sulfide (H₂S) concentration, pH, and nitrogen input but is usually less than 5% (*Seitzinger*, 1988). An average N₂O:N₂ ratio of 1% is considered reasonable for Danish conditions. This gives a total nitrous oxide emission from Danish freshwater systems of 0.1-0.5 kt yr⁻¹.

Trends

About 90% of the nitrogen input to freshwater systems in Denmark comes from farms and cropland (*Danmarks Statistik et al.*, 1990). According to a major national environmental plan the nitrogen input to freshwater systems from agriculture will be halved by 1990 (*Danmarks Statistik et al.*, 1990), but apparently this goal has not yet been achieved. If it is achieved by 1995 the total emission of nitrous oxide from freshwater systems will be then be 0.05-0.25 kt yr⁻¹, corresponding to an annual decline of 0.01-0.05 kt yr⁻¹.

However, the emission can not be expected to drop at the same rate as the nitrogen input. It is more likely to drop at a slower rate because of the time-lag induced by the turnover time of nitrogen in the sediments of freshwater systems. On this background the annual decline is estimated to 0.005-0.05 kt yr⁻¹.

Denmark has 47 000 ha of lakes, 50 000 ha of freshwater marshes, bogs etc., and 40 000 km of streams (*Danmarks Statistik et al.*, 1990). If the average width of the streams is estimated to 1.5 meter the total area of freshwater systems is 103 000 ha. If the range in emission rates from appendix 3 are applied to this area, it results in a total emission of nitrous oxide from freshwater systems of 0.01-0.3 kt yr⁻¹. This value is lower than the value estimated from N-input. The reason for this could be that the sites in appendix 3 are not as heavily eutrophicated as Danish freshwater systems.

5.7. Coastal and marine areas

Open sea

The marine area within Danish territorial limits is 10 400 000 ha (*Skov- og Naturstyrelsen*, 1988) which is 0.029% of the global area of oceans.

No nitrous oxide emission rates for open seas comparable to Danish conditions could be found. Using the total global emission from oceans of 1-4.4 Tg N-N₂O yr⁻¹ (*Bolle et al.*, 1986; *Bouwman*, 1990; *Law, Owens*, 1990) the emission from Danish seas is 0.3-1.3 kt N-N₂O yr⁻¹. As the sea surrounding Denmark is generally very productive and nutrient rich compared to a global average the higher value is probably the most realistic.

Since 1980 the concentration of dissolved nitrogen in Danish seas within the northern-most part of Jutland (an area covering 60 000 000 ha (*Skov- og Naturstyrelsen*, 1988)) has increased with an average rate of about 4% yr⁻¹ (*Danmarks Statistik et al.*, 1990). As the nitrous oxide emission probably increases with nitrogen loading an annual increase in emission of nitrous oxide from Danish seas of 4% yr⁻¹ or 0.05 kt yr⁻¹ is expected. This increase will probably phase out some time in the future when the nitrogen input from anthropogenic land-based sources decreases according to the national plan for lowering the eutrophication of fresh- and saltwater systems.

Coastal areas

The many fjords, estuaries and other shallow brackish or saltwater areas in Denmark hold a great potential for nitrous oxide emission. *Jørgensen and Sørensen* (1985) reported emissions of 1.4 and 15 kg N-N₂O ha⁻¹ yr⁻¹ for a shallow Danish estuary.

Unfortunately, a nationwide geodetic survey of the shallow waters surrounding Denmark has never been carried out so areal data covering these areas are not available (*Plato*, 1991). If an emission rate of 5 kg N-N₂O ha⁻¹ yr⁻¹ and an estimated area of shallow coastal areas of 200 000 ha are used, it results in a total nitrous oxide emission from coastal areas of 1 kt yr⁻¹.

Salt and brackish marshes and meadows

Results from a Danish marine meadow of low salinity and a subtropical salt and brackish marsh in USA give emission rates of 1.3 and 0.3-0.5 kg N-N₂O ha⁻¹ yr⁻¹, respectively (appendix 4). An emission rate of 1 kg N-N₂O ha⁻¹ yr⁻¹ is indicative of the size of the average emission rate from such areas.

42 000 ha of Denmark are covered by salt and brackish marshes and meadows (*Danmarks Statistik et al.*, 1990) which are then estimated to emit a total of 0.04 kt yr⁻¹ of nitrous oxide.

6. Discussion

Total emission

The estimated emissions of nitrous oxide in Denmark from the different natural and agriculture-related sources are summarized in table 1. The total emission amounts to about 13 kt N-N₂O per year. This emission plus the emission from other anthropogenic sources (which is not the scope of this study) has to be compared to the atmospheric destruction of nitrous oxide. For a total global destruction of 10.5 Tg yr⁻¹ (Bouwman, 1990) the destruction "above" Danish land plus sea territory would be approximately 3 kt yr⁻¹. Clearly, on a relative global scale Denmark must rank high among contributors of atmospheric nitrous oxide.

Other investigations

The EEC has within the CORINAIR project also tried to estimate nitrous oxide emissions in Denmark. Their total emission from natural and agriculture-related sources is 20 kt yr⁻¹. Their estimate is rather rough and e.g. only operates with a single emission factor for all cultivated lands, and this emission factor (4.5 kg N-N₂O ha⁻¹ yr⁻¹) is probably too high. The emission factor 5.4 kg N-N₂O ha⁻¹ yr⁻¹ for coniferous forests also seem very high. The CORINAIR emission factors are European averages which does not, however, imply that they can be used for Danish conditions. On the other hand the CORINAIR estimate does not include emission from marine systems, drainage water, groundwater and terrestrial areas like heathland.

Table 1. Estimated annual emission of nitrous oxide from natural and agriculture-related sources in Denmark.

	Estimated annual emission (kt N-N ₂ O)	Estimated range (kt N-N ₂ O)	Estimated annual change (kt N-N ₂ O yr ⁻¹)
Cultivated land	7.7	3-15	-0.05- -0.03
Forest	0.4	0.04-4	+0.002
Other land areas	0.04	0.01-0.1	ns
Drainage water	1.9	0.1-5	ns
Groundwater	?	?	?
Freshwater systems	0.3	0.01-2	-0.05- -0.005
Open sea	1.3	0.1-10	0- +0.05
Shallow coastal areas	1	0.1-10	?
Salt & brackish marshes	0.04	tr-0.4	ns
Sum	12.7		

ns= the change is probably insignificant relative to the total emission from Denmark.

tr= trace amounts.

?= the figure can not be estimated at the present.

The role of fertilizers

The estimated ranges in Table 1 indicate that a high degree of uncertainty is connected to each of the emission rates. However, the estimated emissions indicate that cultivated land is the main source of nitrous oxide in Denmark.

But, more important, fertilizer use and fertilizer induced eutrophication seem to be a key factor contributing significantly to the fluxes from cultivated land, drainage water, and freshwater as well as marine systems. The fertilizer-related emission is probably responsible for 25-50% of the total emission from natural and agriculture-related sources.

National budget

Even if the fertilizer-related emission is taken out of the total emission the emission from natural sources is well above the atmospheric sink strength over Danish territory. This seems likely as Danish soils and coastal areas are quite productive by global standards.

Summarizing the estimated annual change in emission from the different sources (Table 1) indicates that a minor annual decrease (approximately 0.04 kt yr^{-1}) in total emission will take place in the years to come. This decrease equals about 0.3% of the total emission from the sources included in this study. But due to the long lifetime of atmospheric nitrous oxide and to the intense use of fertilizers the total Danish sources are elevated so much above unperturbed steady-state natural source-strength that Denmark for years will continue as a contributor to the increasing atmospheric concentration of nitrous oxide.

Possible reductions

It is not realistic to reduce the emission of nitrous oxide to natural levels as N-fertilization is an integrated part of Danish agriculture. But significant reductions seem possible. The emission from freshwater systems, shallow coastal waters and partly from the open sea will probably be reduced by measures already taken against the eutrophication of these systems.

The emission from cultivated land could be reduced by a reduction in the amount of applied N-fertilizer and perhaps also by changes in the kind of fertilizers used. If e.g. the 80.2 kt of anhydrous ammonia used annually was replaced by an equal amount of ammonium nitrate a reduction of emission by 1.8 kt nitrous oxide per year might be achieved. However, more research is needed to establish the potential of anhydrous ammonia as a source of nitrous oxide.

The use of anhydrous ammonia has decreased significantly during the 1980's as the use in 1982 and 1985 were 153.7 kt and 96.7 kt respectively (*Danmarks Statistik*, 1983, 1986) compared to the 80.2 kt used in 1989. This trend in the use of anhydrous ammonia might continue in the next years, but measures should be taken to reduce the use even further, if the potential of anhydrous ammonia as a source of nitrous oxide is established.

Gaps in knowledge

Table 1 indicates that several gaps exist in our knowledge of nitrous oxide emission from natural and agriculture-related sources. Further research has to address these gaps if we are to understand the emission of nitrous oxide from Denmark.

Especially, the potential of drainage water and groundwater as sources of nitrous oxide and the emission from freshwater systems and shallow coastal areas should be monitored, all of them with a special focus on the influence of N-fertilizers. Other areas to address are the possible impact of deposition of atmospheric nitrogen on nitrous oxide loss from forested areas and the loss of N-fertilizers as nitrous oxide from cultivated land.

7. Conclusion

The estimated emissions of nitrous oxide in Denmark indicate that cultivated land is the major source, and that the widespread use of N-fertilizer in agriculture has profound effects on the total emission. The present total Danish source strength is elevated much above natural unperturbed steady-state source-strength.

It seems unrealistic to reduce the present total emissions to natural levels. But the emission caused by N-fertilizer induced eutrophication will probably be reduced by national measures already being taken to reduce the eutrophication of aquatic systems. If future research establishes the importance of anhydrous ammonia as a source of nitrous oxide measures should be taken to significantly reduce the use of anhydrous ammonia by changing to the use of e.g. ammonium nitrate.

It is important that future research will address the gaps in our understanding of nitrous oxide emission in Denmark in order to make recommendations for a further reduction in emission rates.

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Appendices

Appendix 1a. Grasslands

Appendix 1b. Cropland

Appendix 2. Forests

Appendix 3. Freshwater systems

Appendix 4. Saltwater systems

Appendix 1a. Grasslands

N-fertilizer (kg N ha ⁻¹)	Soil type	Location	Emission rate (kg N-N ₂ O ha ⁻¹ yr ⁻¹)	Fertilizer type	Reference	Remarks
0	Fine loamy (typic hapludalf)	USA (Wisconsin)	0.3	-	Cates, Keeney (1987)	pH 6.7
0	Clay loam	USA (Iowa)	1.3	-	Breitenbeck et al. (1980)	pH 8.2
0	Sandy loam over clay loam	U.K.	0.8-2.7	-	Egginton, Smith (1986b)	pH 6.5, imperfectly drained
0	Clay/silt loam	U.K.	0.8-1.0	-	Webster, Dowdell (1982)	
0	Sandy loam over clay loam	U.K.	1.3-5.8	-	Egginton, Smith (1986a)	pH 6.5, imperfectly drained
0	Sandy loam over clay loam	U.K.	0.2-0.7	-	Egginton, Smith (1986b)	pH 6.5, imperfectly drained
0	Organic (peat) soil	USA (Florida)	16-97	-	Duxbury et al. (1982)	
0	Fine sandy loam over clay	Australia	0.2	-	Denmead et al. (1979)	pH 6.2, dry soil
0	Fine sandy loam over clay	Australia	2.2-9.1	-	Denmead et al. (1979)	pH 6.2, moist soil
210	Heavy clay	U.K.	0.6	NH ₄ NO ₃	Colbourn et al. (1984)	
250	Sandy loam over clay loam	U.K.	0.5-2.0	NO ₃	Egginton, Smith (1986a)	pH 6.5, imperfectly drained
250	Sandy loam over clay loam	U.K.	2.6-11.2	NO ₃	Egginton, Smith (1986a)	pH 6.5, imperfectly drained
250	Loam ochraqualf	U.K.	3.5	NH ₄ NO ₃	Ryden (1983)	
298	Sandy loam over clay loam	U.K.	0.5-2.0	Organic	Egginton, Smith (1986a)	pH 6.5, imperfectly drained
300	Loam (typic xerothent)	USA (California)	1.8-4.3	NO ₃	Rolston et al. (1978)	pH 6.5, imperfectly drained
325	Sandy loam	Denmark (Askov)	50	Cow slurry	Christensen (1981)	Soil constantly wet pH 6.0
400	Silt loam	U.K.	4.0-6.0	NO ₃	Webster, Dowdell (1982)	Well drained
400	Clay loam	U.K.	6.0-8.0	NO ₃	Webster, Dowdell (1982)	Drainage restricted
450	Loam, fine sandy loam	USA (Colorado)	0.8	Urea	Mosier et al. (1981)	
500	Loam ochraqualf	U.K.	8.0	NH ₄ NO ₃	Ryden (1983)	
700	Sandy loam over clay loam	U.K.	13.4	NO ₃	Egginton, Smith (1986b)	Imperfectly drained
700	Sandy loam over clay loam	U.K.	3.3	Organic	Egginton, Smith (1986b)	Imperfectly drained
1230	Sandy loam over clay loam	U.K.	1.2-5.3	Organic	Egginton, Smith (1986a)	pH 6.5, imperfectly drained

Appendix 1b. Cropland

N-fertilizer (kg N ha ⁻¹)	Soil type	Crop	Location	Emission rate (kg N ₂ O ha ⁻¹ yr ⁻¹)	Fertilizer type	Reference	Remarks
0	Loam, ochraqualf	Weeds	U.K.	-0.6	-	Ryden (1981)	pH 6.5
0	Ustic torriorthents	Barley	USA (Colorado)	1.2	-	Mosier et al. (1982)	pH 8.1
0	Silt loam	Alfalfa	USA (New York)	2.3-4.2	-	Duxbury et al. (1982)	
0	Silt loam	Weeds	USA (New York)	0.9-1.7	-	Duxbury et al. (1982)	
0	Oragnic (peat) soil	Sugar cane	USA (Florida)	7-48	-	Duxbury et al. (1982)	
0	Organic (peat) soil	Fallow	USA (Florida)	59-165	-	Duxbury et al. (1982)	
0	Clay loam	Soybean	USA (Iowa)	0.3-2.0	-	Bremner et al. (1980)	pH 6.5-8.1
0	Haplagnolls/calciagnolls	?	USA (Iowa)	0.4-0.7	-	Breitenbeck, Bremner (1986a)	pH 6.9-7.9
40	Sandy loam	Barley	Denmark (Askov)	2.6	NH ₄ NO ₃	Christensen (1981)	pH 6.0
56	Sandy loam	Barley	USA (Colorado)	2.2	NH ₄ NO ₃	Mosier et al. (1982)	pH 8.1
71	Ustic torriorthents	Barley	USA (Colorado)	2.5	Organic	Mosier et al. (1982)	pH 8.1
80	Sandy loam	Barley	Denmark (Roskilde)	3-20	NH ₄ NO ₃	Christensen (1985b)	pH 5.3
80	Silt loam/typic argiudolls	Cabbage	USA (Wisconsin)	0.2-2.2	NH ₄ NO ₃	Goodroad et al. (1984)	
82	Sandy loam	Barley	Denmark (Askov)	4.5	Cow slurry	Christensen (1981)	pH 6.0
105	Heavy clay	?	U.K.	0.5-1.0	NH ₄ NO ₃	Burford et al. (1981)	
105	Heavy clay	?	U.K.	0.9-5.6	NH ₄ NO ₃	Burford et al. (1981)	
105	Heavy clay	?	U.K.	1.5-2.1	NH ₄ NO ₃	Burford et al. (1981)	
105	Heavy clay	?	U.K.	5.4-8.6	NH ₄ NO ₃	Burford et al. (1981)	
107	Sandy loam	Barley	Denmark (Roskilde)	4-11	2:1 Org./NH ₄ NO ₃	Christensen (1985b)	pH 5.3
112	Ustic torriorthents	Barley	USA (Colorado)	2.5	NH ₄ NO ₃	Mosier et al. (1982)	pH 8.1
120	Silty sand (inceptisol)	Wheat	Germany	4	NH ₄ NO ₃	Benckiser et al. (1987)	
125	Clay loam	Uncropped	USA (Iowa)	1.4	NO ₃	Breitenbeck et al. (1980)	pH 8.2
125	Clay loam	Bare soil	USA (Iowa)	2.5	NH ₃ /NH ₄	Breitenbeck et al. (1980)	pH 8.2
125	Clay loam	Bare soil	USA (Iowa)	1.9	Urea	Breitenbeck et al. (1980)	pH 8.2
130	Silt loam	Corn	USA (New York)	1.6-3.8	Organic	Duxbury et al. (1982)	
130	Silt loam	Corn	USA (New York)	2.2-2.9	NH ₄ NO ₃	Duxbury et al. (1982)	

N-fertilizer (kg N ha ⁻¹)	Soil type	Crop	Location	Emission rate (kg N-N ₂ O ha ⁻¹ yr ⁻¹)	Fertilizer type	Reference	Remarks
160	Sandy loam	Barley	Denmark (Roskilde)	6-41	NH ₄ NO ₃	Christensen (1985b)	pH 5.3
160	Sandy loam	Barley	Denmark (Roskilde)	5.2	NH ₄ NO ₃	Christensen (1981)	pH 6.0
170	Organic (peat) soil	Sweetcorn	USA (New York)	76-152	NH ₄ NO ₃	Duxbury et al. (1982)	
170	Organic (peat) soil	Onions	USA (New York)	72-85	NH ₄ NO ₃	Duxbury et al. (1982)	
180	Haplaquolls/calciaguolls	?	USA (Iowa)	1.9-4.4	Anhydrous NH ₃	Breitenbeck, Bremner (1986a)	pH 6.9-7.9
180	Haplaquolls/calciaguolls	?	USA (Iowa)	0.5-9.0	Aqueous NH ₃	Breitenbeck, Bremner (1986a)	pH 6.9-7.9
180	Haplaquolls/calciaguolls	?	USA (Iowa)	0.4-0.8	Urea	Breitenbeck, Bremner (1986a)	pH 6.9-7.9
180	Calciaguolls	?	USA (Iowa)	0.4	CaNO ₃	Breitenbeck, Bremner (1986a)	pH 7.9
181	Fine loam	Corn	USA (Wisconsin)	3.6	Organic	Cates, Keeney (1987b)	pH 6.7
200	Aridic argillstolls	Corn	USA (Colorado)	2.5	NH ₃ /NH ₄	Mosier, Hutchinson (1981)	
200	Silt loam (typic argiudolls)	Corn	USA (Wisconsin)	0.3-6.3	NH ₄ NO ₃	Goodroad et al. (1984)	pH 6.8
210	Heavy clay	Winterwheat	U.K.	1.7-2.0	NH ₄ NO ₃	Colbourn et al. (1984)	
224	Sandy loam	Barley	USA (Colorado)	3.3	NH ₄ NO ₃	Mosier et al. (1982)	pH 8.1
237	Fine loam	Corn	USA (Wisconsin)	5.2	Organic	Cates, Keeney (1987b)	pH 6.7
245	Silt loam (typic argiudolls)	Cabbage	USA (Wisconsin)	0.3-3.2	2:1 Org./NH ₄ NO ₃	Goodroad et al. (1984)	pH 6.8
250	Clay loam	Uncropped	USA (Iowa)	1.4	NO ₃	Breitenbeck et al. (1980)	pH 8.2
250	Clay loam	Uncropped	USA (Iowa)	2.4	NH ₃ /NH ₄	Breitenbeck et al. (1980)	pH 8.2
250	Clay loam	Uncropped	USA (Iowa)	2.4	Urea	Breitenbeck et al. (1980)	pH 8.2
300	Loam (typic xerorthent)	Uncropped	USA (California)	0.6-2.1	NO ₃	Rolston et al. (1978)	Constantly wet
300	Loam (typic xerorthent)	Uncropped	USA (California)	5.4-9.9	NO ₃	Rolston et al. (1978)	Constantly wet
356	Sandy loam	Barley	USA (Colorado)	9.9	Organic	Mosier et al. (1982)	pH 8.1
410	Silt loam (typic argiudolls)	Cabbage	USA (Wisconsin)	1.2-6.1	4:1 Org./NH ₄ NO ₃	Goodroad et al. (1984)	pH 6.8
424	Sandy loam	Barley	Denmark (Roskilde)	15-29	3:1 NH ₄ NO ₃ /Org.	Christensen (1985b)	pH 5.3
430	Fine loamy pachic haploxeroll	Artichokes	USA (California)	20-27	NO ₃	Ryden, Lund (1980)	Irrigated
440	Silt loam (typic argiudolls)	Barley	USA (Wisconsin)	0.2-1.6	Sludge	Goodroad et al. (1984)	pH 6.8
620	Fine loamy pachic haploxeroll	Lettuce	USA (California)	20-42	NO ₃	Ryden, Lund (1980)	Irrigated
680	Fine loamy pachic haploxeroll	Cauliflower	USA (California)	27-29	NO ₃	Ryden, Lund (1980)	Irrigated

Forest type	Soil type	Location	Emission rate (kg N-N ₂ O ha ⁻¹ yr ⁻¹)	Reference	Remarks
Deciduous	Sandy brown soil	Germany	0.4-0.9	Schmidt et al. (1988)	pH 3.7, 1-3 cm humus layer
Deciduous	Grey-brown podzol, loess	Germany	0.3-0.8	Schmidt et al. (1988)	pH 3.6, 1-5 cm humus layer
Deciduous	Pseudogley soil, loess	Germany	0.5-6.6	Schmidt et al. (1988)	pH 3.4, 1-3 cm humus layer
Deciduous	Grey-brown podzol, loess	Germany	0.5-0.7	Schmidt et al. (1988)	pH 3.7, 1-2 cm humus layer
Deciduous	Brown soil, loess loam	Germany	0.1-0.3	Schmidt et al. (1988)	pH 4.7
Deciduous	Pelosoil, loess loam	Germany	0.4-0.9	Schmidt et al. (1988)	pH 6.7
Deciduous		USA (New Hampshire)	0.15	Keller et al. (1983)	65 years old trees
Hardwood	Silt loam	USA (New York)	0.9	Duxbury et al. (1982)	
Hardwood	Sandy loam	USA (Massachusetts)	0.02	Bowden et al. (1990)	pH 3.3, 80 years old, well drained
Oak	Loess over glacial till	USA (Wisconsin)	0.5-1.3	Goodroad, Keeney (1984a)	pH 5.0
Beech		Denmark (Strødam)	0.3	Christensen (unpubl.)	
Beech		Sweden	0.01-2.0	Klemedtsson, Svensson (1988)	100 years old, lower rates dominated
Ash		Denmark	0.3	Struwe, Kjølner (1989)	pH 6.5-7.5, waterlogged
Alder		Denmark	1.8	Struwe, Kjølner (1989)	pH 6.5-7.0, waterlogged
Alder		Sweden	0.2-4.0	Klemedtsson, Svensson (1988)	
Pine	Loess over glacial till	USA (Wisconsin)	2.4-3.0	Goodroad, Keeney (1984a)	
Pine	Sandy loam	USA (Massachusetts)	0.01	Bowden et al. (1990)	pH 4.5, 40 years old
Spruce		Denmark (Gribskov)	0.8	Christensen (unpubl.)	pH 3.2, 62 years old, well drained
Spruce		Sweden	0.01-2.0	Klemedtsson, Svensson (1988)	5-6 years old, clearcut site, lower rates dominated
Spruce		Sweden	0.01-1.0	Klemedtsson, Svensson (1989)	40-60 years old, lower rates dominated

Appendix 3. Freshwater systems

Ecosystem	Location	Emission rate (kg N-N ₂ O -1 ha ⁻¹ yr ⁻¹)	Reference	Remarks
Meadow	Denmark (Viby Sj.)	0.6	Christensen (unpubl.)	
Wet meadow	USA (Wisconsin)	2.7	Goodroad, Keeney (1984a)	Disturbed sedge overgrown with reed canary grass
Marsh	USA (Louisiana)	0.55	Smith et al. (1983)	Only lightly eutrophied
Marsh	USA (Louisiana)	0.35	Smith et al. (1983)	Open water site, only lightly eutrophied
Marsh	USA (Wisconsin)	0.1	Goodroad, Keeney (1984a)	pH 7.0, cattails (<i>Typha angustifolia</i>) dominating
Drained marsh	USA (Wisconsin)	5.7-13.1	Goodroad, Keeney (1984a)	pH 6.1, old sedge meadow overgrown with bushes/small trees

APPENDIX 4. NITROUS OXIDE EMISSION RATES - TEMPERATE SALTWATER SYSTEMS.

Ecosystem	Location	Emission rate (kg N-N ₂ O -1 ha ⁻¹ yr ⁻¹)	Reference	Remarks
Brackish meadow	Denmark (Sydanager)	1.3	Christensen (unpubl.)	Low salinity
Brackish estuary	Denmark (Norsminde)	1.4-15	Jørgensen, Sørensen (1985)	Two sites: Salinity 0.05-0.8‰ & 0.8-2.3‰.
Brackish marsh	USA (Louisiana)	0.5	Smith et al. (1983)	Little eutrophication
Brackish marsh	USA (Louisiana)	0.2	Smith et al. (1983)	Open water site, little eutrophication
Salt marsh	USA (Louisiana)	0.3	Smith et al. (1983)	Little eutrophication
Salt marsh	USA (Louisiana)	0.1	Smith et al. (1983)	Open water site, little eutrophication
Marine sediment	USA (Rhode Island)	0.02	Seitzinger et al. (1980)	Relatively unpolluted

Appendix 4. Saltwater systems

Ecosystem	Location	Emission rate (kg N-N ₂ O -1 ha ⁻¹ yr ⁻¹)	Reference	Remarks
Meadow	Denmark (Viby Sj.)	0.6	Christensen (unpubl.)	
Wet meadow	USA (Wisconsin)	2.7	Goodroad, Keeney (1984a)	Disturbed sedge overgrown with reed canary grass
Marsh	USA (Louisiana)	0.55	Smith et al. (1983)	Only lightly eutrophied
Marsh	USA (Louisiana)	0.35	Smith et al. (1983)	Open water site, only lightly eutrophied
Marsh	USA (Wisconsin)	0.1	Goodroad, Keeney (1984a)	pH 7.0, cattails (<i>Typha angustifolia</i>) dominating
Drained marsh	USA (Wisconsin)	5.7-13.1	Goodroad, Keeney (1984a)	pH 6.1, old sedge meadow overgrown with bushes/small trees

APPENDIX 4. NITROUS OXIDE EMISSION RATES - TEMPERATE SALTWATER SYSTEMS.

Ecosystem	Location	Emission rate (kg N-N ₂ O -1 ha ⁻¹ yr ⁻¹)	Reference	Remarks
Brackish meadow	Denmark (Sydamager)	1.3	Christensen (unpubl.)	Low salinity
Brackish estuary	Denmark (Norsminde)	1.4-15	Jørgensen, Sørensen (1985)	Two sites: Salinity 0.05-0.8% & 0.8-2.3%.
Brackish marsh	USA (Louisiana)	0.5	Smith et al. (1983)	Little eutrophication
Brackish marsh	USA (Louisiana)	0.2	Smith et al. (1983)	Open water site, little eutrophication
Salt marsh	USA (Louisiana)	0.3	Smith et al. (1983)	Little eutrophication
Salt marsh	USA (Louisiana)	0.1	Smith et al. (1983)	Open water site, little eutrophication
Marine sediment	USA (Rhode Island)	0.02	Seitzinger et al. (1980)	Relatively unpolluted

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The National Environmental Research Institute - NERI - is a research institute of the Ministry of the Environment. Neri's tasks are primarily to do research, collect data and give advice on problems related to the environment and nature.

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