COMPILATION OF A GLOBAL INVENTORY OF EMISSIONS OF NITROUS OXIDE

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COMPILATION OF A GLOBAL INVENTORY OF EMISSIONS OF NITROUS OXIDE

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Distikstofoxide (*Eng.* nitrous oxide), N_2O, lachgas, een kleurloos gas dat als narcosemiddel wordt gebruikt. Lachgas wordt o.a. bereid door verhitting van natriumnitraat met ammoniumchloride. Het komt in geringe concentraties in de atmosfeer voor. In 1799 werd ontdekt dat lachgas dempend werkt op het bewustzijn, en tot bewusteloosheid en ongevoeligheid voor pijnprikkels kan voeren. De naam lachgas slaat op de beginfase van de narcosetoestand die op een alcoholroes lijkt. Lachgas is een bruikbaar gas bij de totaalanesthesie, voor operaties en andere ingrepen (De Grote Oosthoek Encyclopedie en Woordenboek, Oosthoek's Uitgeverij bv, Utrecht).

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JUPITER ET LE MÉTAYER

Jupiter eut jadis une ferme à donner.
Mercure en fit l'annonce, et gens se présentèrent,
Firent des offres, écoutèrent :
Ce ne fut pas sans bien tourner ;
L'un alléguoit que l'heritage
Étoit frayant et rude, et l'autre un autre si.
Pendant qu'ils marchandoient ainsi,
Un d'eux, le plus hardi, mai non le plus sage,
Promit d'en rendre tant, pourvú que Jupiter
Le laissât disposer de l'air,
Lui donnât saison à sa guise,
Qu'il eût du chaud, du froid, du beau temps, de la bise,
Enfin du sec et du mouillé,
Aussitôt qu'il auroit bâillé.
Jupiter y consent. Contrat passé; notre homme
Tranche du roi des airs, pleut, vente, et fait en somme
Un climat pour lui seul : ses plus proches voisins
Ne s'en sentoient non plus que les Américains.
Ce fut leur avantage : ils eurent bonne année,
Pleine moisson, pleine vinée.
Monsieur le Receveur fut très-mal partagé.
L'an suivant, voilà tout changé :
Il ajuste d'une autre sorte
La temperature des cieux.
Son champ ne s'en trouve pas mieux;
Celui de ses voisins fructifie et rapporte.
Que fait-il? Il recourt au monarque des Dieux,
Il confesse son imprudence.
Jupiter en usa comme un maître fort doux.

Concluons que la Providence
Sait ce qu'il nous faut mieux que nous (VI, iv).

La Fontaine (1668)

Voor mijn vader

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FOREWORD

In recent years we have become increasingly aware of how strongly the physical and chemical properties of the Earth's atmosphere are influenced by fluxes of trace gases from natural and anthropogenic sources. One of these gases is nitrous oxide (N_2O), or laughing-gas. It plays a role in the atmospheric radiative balance and in the stratospheric ozone chemistry. Many major and minor natural and anthropogenic sources of nitrous oxide have been identified, yet there is considerable uncertainty in the source strengths. Part of the uncertainty arises from the paucity of measurements of N_2O fluxes. Another problem is to extrapolate local measurements to larger scales. Perhaps all biological processes involving oxidation or reduction of nitrogen through the +1 or +2 oxidation state yield trace amounts of nitrous oxide. Several industrial processes have also been identified as sources of nitrous oxide. An increase of nitrogen inputs to the environment through increased biological and industrial nitrogen fixation, for example in the form of synthetic nitrogen fertilizers, will eventually lead to increased nitrous oxide injection into the atmosphere.

This study concentrated on the nitrous oxide emissions from soils. However, for validation of source estimates an atmospheric transport model was used, which in turn required inclusion of estimates for all known sources in the global inventory. The results of this study form only another step towards a better understanding of the global distribution of sources of nitrous oxide. The research was carried out in the period 1990-1994. The chapters are not presented in chronological order. Chapter 2, describing a simple model for N_2O production in soils, is the result of a short period in 1990 as guest-researcher at NASA-Goddard Institute for Space Studies. Chapter 3 describes results of the data collection and analysis done in the period 1990-1994. Chapter 4 presents the emission inventories that were compiled in 1992-1994 in the framework of the EDGAR and IMAGE projects at RIVM. Chapter 5 was prepared in collaboration with Douwe van Dam of the Soil Science and Geology Department of the Wageningen Agricultural University in 1994. Chapter 6 is the result of a collaborative study with John Taylor of the Australian National University in Canberra, Australia, in the period 1992-1994.

Most of the chapters presented in this thesis have been published before, or will be published in the near future. Where relevant, the literature reference of each chapter, the co-authors and their affiliation are printed on the page opposite the chapter's title page.

The preliminary inventory described in this dissertation is a contribution the Global Emission Inventories Activity (GEIA), a project of the Global Atmospheric Chemistry Programme (IGAC).

Wageningen, December 1994.

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The National Institute of Public Health and Environmental Protection (RIVM) allowed me to work part of my time on the study described in this dissertation. In particular I am indebted to Ton Bresser and Rob Swart for stimulating me in this effort.

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In 1992 I met John Taylor in Tsukuba, Japan, where we first discussed the idea to start the modelling exercise described in Chapter 6. We met a few more times, but most of our communication was via electronic mail. It was exciting to actually talk via the computer, sometimes very early in the morning.

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Wageningen, December 1994.

SUMMARY

A global inventory with 1°x1° resolution was compiled of emissions of nitrous oxide (N_2O) to the atmosphere, including emissions from soils under natural vegetation, fertilized agricultural land, grasslands and animal excreta, biomass burning, forest clearing, oceans, fossil fuel and biofuel combustion, and industrial sources.

A simple global model of the production potential of N_2O in soils under natural vegetation was developed to analyze the relative importance of five major controls on N_2O production: (i) input of organic matter, (ii) soil fertility, (iii) soil moisture status, (iv) temperature; and (v) soil oxygen status. Indices for the controls were derived from global gridded (1° × 1° resolution) data bases of soil type and texture, normalized difference vegetation index (NDVI) and climate. The model explains close to 60% of the variability found in measurements reported at about 30 sites in six different ecosystems throughout the world. The model results confirm conclusions from earlier studies that the major natural source regions of N_2O are in the tropics.

Literature data on N_2O flux measurements from agricultural fields show that the fertilizer-induced N_2O emission is higher for measurements covering longer periods than for measurements which represent short periods. A method to estimate the total annual direct N_2O emission from fertilized fields was based on measurements covering periods of one year: N_2O-N emission = 1 kg N $ha^{-1}yr^{-1}$ plus 1.25 ± 1% of the amount of fertilizer N applied (kg N $ha^{-1}yr^{-1}$). This relationship was used to compile a global 1° x 1° resolution inventory of emissions of N_2O from fertilized arable land.

The inventory of N_2O emission from animal excreta was based on estimates of nitrogen (N) excretion by various categories of domestic animals. The estimated global amount of N in animal excreta (~100 x 10^{12} g N yr^{-1}) suggests that the associated N_2O emission may be of the same order of magnitude as that caused by the use of synthetic N fertilizer (~80 x 10^{12} g N yr^{-1}).

To illustrate the difficulty to describe the cycling of N in ecosystems, N budgets were compiled for a deforestation sequence in the Atlantic Zone of Costa Rica. After forest clearing an important part of the soil organic N is mineralized. Part of the nitrate formed by nitrification of the mineralized N is lost via leaching, while most of the N loss occurs through denitrification. After a period of 3 to 5 years most of the easily decomposable material is lost, and denitrification and N_2O fluxes decrease with time to levels lower than in the undisturbed forest. The global estimate of enhanced soil N_2O emission from denitrification following tropical forest clearing, accounting for this decline of N_2O fluxes along with ageing of the clearing, indicates that deforestation is an important global source.

Global 1° x 1° resolution inventories were also compiled for N_2O emissions from fossil fuel and fuelwood combustion, and industrial N_2O sources. For N_2O emissions from oceans and biomass burning, inventories from the literature were used. The complete inventory of annual N_2O emissions including all sources, was compared with source estimates inferred from inverse modeling. The N_2O inventories are in general agreement with inverse modeling results. However, there are major uncertainties, particularly in the tropics. The comparison between the emission inventory and the source estimates from inverse modelling, resulted in improved understanding of some sources:

- The oceanic N_2O emission may be higher than previous estimates; the 0°N-30°N latitudinal zone and the Antarctic ocean show much higher N_2O fluxes than the mean global oceanic flux.
- Most of the N_2O from arable lands and grasslands including effects of synthetic fertilizers and animal excreta comes from the northern hemisphere. Inputs of N to soils from N deposition and from N fixation by leguminous crops are of the same order of magnitude as synthetic N fertilizer use. Their associated N_2O release may also be similar in magnitude.
- Fossil-fuel combustion and industrial N_2O sources are dominant in the 30°N-90°N zone, while N_2O from fuelwood combustion is mainly produced in the 0°N-30°N zone.
- The major part of the N_2O emitted from coastal marine and fresh water systems probably stems

from the northern hemisphere.

Global monthly estimates of N_2O emissions were used to prescribe a three-dimensional atmospheric transport model. The simulated northern hemispheric N_2O surface concentration was ~1 ppb higher than in the southern hemisphere. This is in general agreement with atmospheric observations. The modeled N_2O concentrations over strong source regions in continental interiors were up to 5 ppb higher than those over oceans. Predicted concentrations for the northern hemisphere were somewhat higher in summer than in winter, in agreement with the seasonality of N_2O emissions. However, the atmospheric N_2O concentration measurements show no seasonal variation in the northern hemisphere. The small seasonality in modeled atmospheric N_2O concentrations for the southern hemisphere is more consistent with measurements. Inconsistencies between predicted and observed atmospheric N_2O concentrations may be caused by overestimation of the seasonality in the northern hemisphere. The global N_2O inventory used does not account for soil N_2O consumption in temperate N-limited ecosystems and observed episodic emissions in temperate ecosystems during winter, early spring and autumn. These potential errors and possible underestimation of N_2O emissions from combustion in winter may exaggerate the simulated seasonal trends.

Another possible reason for the inconsistencies found between predicted and measured N_2O concentrations is that seasonal trends in atmospheric N_2O concentrations remain unobserved, because of the remote location of most monitoring stations. In addition, the precision of N_2O measurements is not adequate for resolving seasonal trends.

The major part of the atmospheric N_2O increase stems from sources related to human food production. A growing world population will inevitably lead to more food demand. Moreover, an increasing portion of the synthetic fertilizers is used to increase animal production. At present, close to 40% of the global cereal production and 25% of the production of root and tuber crops is fed to animals. An important global reduction of N_2O emission associated with food production could be achieved by a shift-away from animal production, by more efficient agricultural use of N, and by advanced fertilization techniques.

Solutions to reduce N_2O emission from fossil fuel combustion include technical options (e.g. development of new catalysts) and energy saving. Several industrial and chemical processes generate N_2O, but so far only the N_2O production from nitric and adipic acid production have been quantified. The global N_2O emission from adipic acid production is currently decreasing as the major producers have agreed to reduce N_2O emissions.

Chapter 1

GENERAL INTRODUCTION

1.1. BACKGROUND

The climate of the Earth is controlled by the radiative balance of the atmosphere. On a long term basis the energy of incoming radiation is balanced by that of outgoing radiation. About half of the incoming solar radiation is absorbed at the Earth's surface. The Earth gives off radiant energy known as thermal radiation or planetary infrared radiation. A few trace gases in the Earth's atmosphere – "greenhouse gases" – have an important physical property that has the potential to make our planet habitable: they absorb radiant energy at infrared wavelengths much more efficiently than they absorb radiant energy at solar wavelengths, and thus trap radiant heat emitted from the Earth's surface before it can escape into space. The most important greenhouse gases are water vapor, carbon dioxide (CO_2) and trace amounts of methane (CH_4), ozone (O_3), nitrous oxide (N_2O) and other trace gases. This trapping of energy is very efficient: only 5% of the outward directed long wave radiation can escape from Earth's surface into space. The remainder is absorbed by the atmospheric greenhouse gases and clouds, and 90% of the absorbed radiation is radiated back to the Earth's surface. This atmosphere-surface cycling is the "greenhouse" effect, causing the earth's surface to be about 33°C warmer than would otherwise be the case (Graedel and Crutzen, 1993). Water vapor and CO_2 account for respectively 60 - 70% and 25% of the 'natural' greenhouse effect.

Perturbations to the energy balance alter climate by creating radiative forcing of climate. The energy balance can be perturbed by variation in solar radiation, variations in the earth's orbit, and changes in the greenhouse effect. Currently there is concern over a possible enhancement of the greenhouse effect caused by increasing concentrations of atmospheric trace gases. To compare the effects of the different gases on the greenhouse effect, an index has been developed that is commonly referred to as the Global Warming Potential (GWP). The GWP combines the capacity of a gas to absorb infrared radiation, its residence time in the atmosphere and a time frame over which climatic changes are evaluated. Estimates of GWPs for some atmospheric trace gases are presented in Table 1.

Table 1. Global warming potential (GWP) and other properties of CO_2, CH_4 and N_2O

Gas	Concentration†	Annual increase†	Lifetime (yr)	Relative absorption capacity‡	GWP§	Sign of indirect effects
CO_2	355 ppmv	1.8 ppmv	120	1	1	none
CH_4	1.72 ppmv	10-13 ppbv	12-17	58	24.5¶	positive
N_2O	310 ppbv	0.8 ppbv	120	206	320	uncertain

Compiled from Shine et al. (1990), Isaksen et al. (1992), and IPCC (1994).

† ppmv = parts per million by volume; ppbv = parts per billion by volume.

‡ Per unit mass change from present concentrations, relative to CO_2.

§ Direct Global Warming Potential following addition of 1 kg of each gas, relative to CO_2 for a 100-year time horizon.

¶ Including the direct effect of CH_4 and indirect effects due to the production of tropospheric ozone and stratospheric water vapor.

1.2. NITROUS OXIDE IN THE ATMOSPHERE

Although N_2O occurs in the atmosphere in minute quantities compared to CO_2 and water vapor, its contribution to the greenhouse effect is considerable. This is caused by its long residence time and high relative absorption capacity per unit mass (Table 1). The direct contribution of N_2O to radiative forcing of climate may have increased from 4% over the period 1950-1990 to 6% during 1980-1990 (Kroeze, 1993) and it may progressively increase to 10% in the coming decades (Dickinson and Cicerone, 1986).

In the troposphere considerable amounts of N_2O may be formed by photochemical NH_3 oxidation (Dentener, 1993). Most of this N_2O formation is related to high NH_3 and NO_x emissions from tropical biomass burning. Nitrous oxide may also be formed in exhaust gases from combustion processes (Khalil and Rasmussen, 1992), and other by processes such as lightning and corona processes around high voltage eletrical transmission lines. To date no reliable global estimates for these processes have been published (see section 1.3).

In the troposphere N_2O is stable and thus inert. It is carried into the stratosphere where it is removed by photolysis and reaction with excited oxygen atoms, $O(^1D)$, as shown by reaction equations (1) - (3) from Warneck (1988). The $O(^1D)$ is mainly produced by photolysis of ozone (O_3).

$$N_2O + h\nu \rightarrow N_2 + O(^1D) \quad (1)$$
$$O(^1D) + N_2O \rightarrow N_2 + O_2 \quad (2)$$
$$O(^1D) + N_2O \rightarrow NO + NO \quad (3)$$

About 90% of the destruction of N_2O is by photolysis according to reaction (1) (Minschwaner et al., 1993). The rest of the N_2O is removed by reactions (2) (4% of the N_2O) and (3) (6%) (Crutzen and Schmaichl, 1983). Oxidation of N_2O according to reaction (3) is the major source of NO in the stratosphere between 20 and 40 km altitude, and NO plays an important role in the destruction of stratospheric ozone (Crutzen, 1970). Hence, increases in N_2O input in the stratosphere may result in ozone loss. Banin et al. (1984) speculated that an increase by 10% of atmospheric N_2O may result in a loss of the average ozone column by 1.5%. However, stratospheric NO may also mitigate chlorine-catalyzed ozone destruction by sequestering harmful ClO into the stable reservoir species $ClONO_2$ (Isaksen and Stordal, 1986). The indirect climatic effects of N_2O are yet uncertain (Table 1). There may be mutual influences of stratospheric ozone depletion and global warming. However, it is likely that the effect on climate of stratospheric ozone depletion caused by N_2O is relatively unimportant (Isaksen et al., 1992).

Analysis of antarctic ice core samples suggests that the atmospheric N_2O concentration has risen from about 270 ppbv 400 years ago to about 293 ppbv in the beginning of the 20th century (Zardini et al., 1988) and to 310 ppbv now (Khalil and Rasmussen, 1992). Records from longer periods indicate that the atmospheric N_2O concentration was at least 30% lower during the last Glacial Maximum than during the Holocene epoch, and that present-day N_2O concentrations are unprecedented in the past 45000 years (Leuenberger and Siegenthaler, 1992). The atmospheric N_2O concentration started to increase rapidly during this century, but an accelerated increase may have started only after 1940 (Stauffer and Neftel, 1988). The N_2O concentration may even be increasing at a faster rate in recent years than during the middle 1970s (Khalil and Rasmussen, 1992). The observed increase of atmospheric N_2O during the 1980s was 0.25-0.31% yr^{-1} (Prinn et al., 1990). However, the trends over the last decade are extremely variable. Over 3-year periods increase rates have ranged from 0.5 ± 0.2 ppbv yr^{-1} to 1.2 ± 0.1 ppbv yr^{-1}, averaging 0.8 ± 0.2 ppbv yr^{-1} over 1977-1988 (Khalil and Rasmussen, 1992). In 1992 the increase was about half that in the 1980s (Swanson et al., 1993). This was perhaps a result of the observed global cooling caused by aerosols emitted by Mt. Pinatubo in 1991 (Dutton and Christy, 1992). The cooling may have caused lower N_2O production in soils[1] and oceans (J. Elkins, personal communication). The growth of atmospheric N_2O started increasing again from late 1993 (J. Elkins, personal communication).

The atmospheric burden of N_2O is about 1500 Tg N_2O-N (Minschwaner et al., 1993). The annual increase of atmospheric N_2O during the 1980s of 0.25-0.31% was caused by an imbalance of sources over sinks of 3 - 4.5 Tg N_2O-N yr^{-1}, accounting for 30% or more of the total source of 13 - 16 Tg N_2O-N yr^{-1} (Minschwaner et al., 1993; Khalil and Rasmussen, 1992).

The stratospheric destruction by reactions (1), (2) and (3) removes 10.5 ± 3 Tg N_2O-N yr^{-1} (McElroy and Wofsy, 1986) to 12.2 ± 3.7 Tg N_2O-N yr^{-1} (Minschwaner et al., 1993). The atmospheric lifetime of N_2O based on these destruction rates and the atmospheric burden amounts to respectively 100 - 200 and 120 ± 40 years. These estimates do not account for other possible removal processes, such as uptake of N_2O by soils and aquatic ecosystems.

1.3. SEARCH FOR N_2O SOURCES AND SINKS

Probably most of the N_2O in the earth's atmosphere is from microbiological processes in soils and aquatic systems. In soils the major sources of N_2O are generally accepted to be denitrification and nitrification (Williams et al., 1992). In subsurface environments denitrification is the major source of N_2O (Rice and Rogers, 1993). Kim and Craig (1990) suggested that nitrifying bacteria are the dominant N_2O source in the oceans, based on isotopic ratios of N_2O. However, Yoshida et al. (1990) suggested that nitrification is the primary source in near surface ocean waters, but denitrification was perhaps the primary source of N_2O at depth in the oxygen minimum zone. In rivers, streams, lakes, coastal marine systems and continental shelves most N_2O stems from the sediments, primarily produced by denitrification with minor amounts coming from nitrification. Nitrification and denitrification and N_2O production also occur in the water column of these aquatic systems (Seitzinger, 1990).

Banin et al. (1984) noted that under reducing conditions with no other available source of N, soils may act as sinks for N_2O. Uptake of N_2O by the ocean surface has also been observed (see Nevison, 1994). Due to lack of knowledge of the conditions at which soils and aquatic systems act as sinks for N_2O, and the parameters affecting the influx when they do so, is too limited to evaluate their importance at the global scale.

It is not certain what causes the increase of atmospheric N_2O. The N_2O emission from fossil fuel combustion and biomass burning has long been considered the major cause. Recently Muzio and Kramlich (1988) discovered that when mixtures of flue gases such as in power plant effluent, are stored, reactions occur in the presence of SO_2, NO_x and H_2O that can produce substantial amounts of N_2O. All past work on N_2O from coal combustion had relied on stored samples and became suspect. Based on new analyses Khalil and Rasmussen (1992) concluded that direct N_2O emission from stationary fossil fuel combustion contributes less than 1% of the global source. However, as yet unknown amounts of N_2O may be formed in smoke plumes and during catalytic reduction of NO_x. The recent history of global "budgets" of emissions of N_2O illustrates the change in views before and after the discovery of the sampling artifact (Table 2).

Recently, Khalil and Rasmussen (1992) listed a great number of source candidates, including agricultural soils, natural soils, aquatic sources, biomass burning, land use changes, fossil fuel combustion, automobiles, industrial and other sources (Table 2). The major sources in most N_2O budgets are formed by soils under natural vegetation, followed by oceans and agricultural soils.

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[1] E. Kreileman, unpublished calculations with the model discussed in chapters 2 and 4, showing a 0.4 Tg N_2O-N decrease in global soil emissions associated with the cooling observed by Dutton and Christy (1992) and later observations from unpublished data provided by J. Elkins. Tg = teragram; 1 Tg = 10^{12}g.

Table 2. Comparison of global N_2O estimates presented between 1984 and 1992 in Tg N_2O-N yr^{-1}. NK = indicated by authors as unquantified potential source

Source	Reference †				
	1	2	3	4	5
Soils under natural vegetation	2.6-25	3.5-11.5	3-9	2.7-7.7	7.6
Cultivated soils	1.6-5.3	3.5-5.9	0.5-2.5	0-3	0.3-2
Grasslands		8.1			
Animal waste					0.2-0.6
Biomass burning	1-2	0.5-0.9	1-2	0.2-1	0.1-2
Land use changes			0.2-0.6		0.4
Oceans	1-10	1-3	1-3	1.4-2.6	1.9
Fossil fuel combustion	1-2	3-5	1-3	0.1-0.3	0-0.1
Industry				0.5-0.9	0.4
Mobile sources				0.2-0.6	0.1-1.3
Aquifers					0.5-1.3
Sewage					0.2-1.9
Waste water treatment	1-2				
Global warming					0-0.6
Atmosph. formation‡	0-21				NK
Trash incineration					NK
Total	9-67	9-22	7-21	10-18	13-16

† 1 = Banin et al. (1984), estimate for 1970s; 2 = McElroy and Wofsy (1986); 3 = Seiler and Conrad (1986); 4 = Watson et al. (1992); 5 = Khalil and Rasmussen (1992)

‡ including excited species chemistry (20 Tg N_2O-N yr^{-1}), N_2O formation during lightning (<0.01 Tg yr^{-1}), and corona processes in the atmosphere surrounding high voltage electrical transmission lines (0-0.5 Tg yr^{-1})

Most anthropogenic sources are minor, and precise estimates of emissions will probably not be known for a long time (Khalil and Rasmussen, 1992). Industrial sources include the production of adipic acid, primarily used for nylon 6,6 polyamide, and the production of nitric acid, mainly used for fertilizers and other intermediate products including adipic acid. Additional, yet unidentified industrial processes, may also generate N_2O.

Formation of N_2O in the atmosphere was believed to be a significant source by some authors (Banin et al., 1984), though later ignored by others (Seiler and Conrad, 1987; Watson et al., 1992). In the most recent budget atmospheric chemistry is listed again without quantification.

Recently global warming has also been mentioned as a potential N_2O source (Table 2). Other known minor sources not listed in Table 2 include fresh water and coastal marine waters, effects of N deposition on soil N_2O emission, the production and use of explosives, medical and industrial use of N_2O, and use of N_2O as aerosol propellant (Granli and Bøckman, 1994).

Individual sources may be of great local or regional importance. For example, worldwide production of adipic acid occurs in a limited number of plants, while deforestation and biomass burning are of particular regional importance in the tropics.

Several studies have recently reported that isotopic ratios can be used to determine the mechanism of N_2O production. Wahlen and Yoshinari (1985) showed that $^{18}O/^{16}O$ isotopic ratios of N_2O can be sensitively measured, and found that $\delta^{18}O$ values for N_2O derived from nitrification are lower than those for N_2O from denitrification. Assuming that these isotopic differences are uniform among different systems, several authors claim that the process of formation of N_2O can be determined (Yoshida and Matsuo, 1983; Yoshinari and Wahlen, 1985; Yoshida, 1988). Nitrous oxide in soil and groundwater may be significantly depleted in ^{15}N and ^{18}O relative to tropospheric N_2O (Ueda, 1991; Kim and Craig, 1993). In surface ocean waters down to 600 m depth N_2O is depleted in both heavy isotopes, but at greater depth N_2O is enriched in ^{15}N and ^{18}O. Coal plant and engine exhaust have been shown to be enriched in ^{18}O relative to N_2O in the troposphere and in soil and groundwater (Kim and Craig, 1990; 1993; Wahlen and Yoshinari, 1985; Yoshinari and Wahlen, 1985). The N_2O from the stratospheric backflux to the troposphere may be heavier than tropospheric N_2O. However, Kim and Craig (1993) concluded that at present the isotopic ratios of N_2O in the atmosphere have not been sufficiently investigated to be used to identify and quantify N_2O sources.

1.4. PURPOSE AND OUTLINE OF THIS STUDY

The aim of this study is to develop and validate a global inventory of emissions of N_2O – with emphasis on the soil-borne sources. The inventory must describe both the spatial and temporal patterns of emissions to allow for validation with three-dimensional atmospheric transport models. Seasonality in the atmospheric N_2O concentrations has been observed (Khalil and Rasmussen, 1983). Spatial and temporal distributions of emissions may be used in forward runs with three-dimensional atmospheric transport models to predict atmospheric N_2O concentrations. Comparison of predicted and observed seasonal patterns in the atmospheric N_2O concentration may lead to better understanding of the source distributions. Inverse modeling techniques may be used to infer regional source distributions.

It is difficult to verify source estimates in N_2O budgets. The stratospheric N_2O destruction and the atmospheric increase are relatively well known, but these constraints can be satisfied on the basis of different assumptions for the N_2O sources. There is considerable uncertainty in all source estimates (Table 2). The uncertainty arises from the paucity of measurements of N_2O fluxes and the difficulty in making extrapolations. For the anthropogenic sources political, economic and cultural factors cause major uncertainties in making extrapolations. For biogenic fluxes from soil and aquatic systems, extrapolation of flux measurement data to larger scales is difficult, because of the extreme spatial and temporal heterogeneity of regulating factors of N_2O production, consumption and emission in these systems. For example, most estimates of emissions from soils, the major global N_2O source, were made one the basis of a few measurements that were multiplied by the land area of the globe or the extent of a particular ecosystem. Trace gas fluxes from water surfaces cannot be directly measured. Generally, fluxes from oceans are modeled from N_2O supersaturation and the water-air gas transfer coefficient. Measurements of N_2O supersaturation in the surface water of oceans represent only a minor part of the area covered by the world's oceans.

Matson et al. (1989) recommended to stratify ecosystems and to make geographically explicit descriptions of regulating factors of N_2O emission in ecosystems to improve the understanding of the variability within ecosystems. Since both denitrification and nitrification play a role in terrestrial and aquatic systems, it is obvious that N_2O fluxes are strongly linked to the nitrogen cycle. Therefore, one way of studying the global sources of N_2O is by describing the global nitrogen cycle. For example, in terrestrial systems there may be fluxes of nitrogen from various sources, including mineral fertilizers, animal excreta, biological N fixation, N mineralization and deposition. Quantification of most of these fluxes requires description of completely different biological, chemical, physical and atmospheric processes. System losses of nitrogen include denitrification, volatilization, run-off and leaching. Even for intensively studied sites it is difficult to estimate the individual loss-components, and the spatial and temporal variability is extremely high. Knowledge of the local hydrology is required to estimate the fluxes of nitrogen from the subsoil to aquifers, via aquifers to surface waters and to deeper ground water. Denitrification in these subsurface environments occurs, but the fate of the N_2O formed is not well known (Rice and Rogers, 1993). Geographically explicit calculations being a requirement, a comprehensive and descriptive model of the global N cycle is an impossible goal as yet.

Therefore, in this study individual sources of N_2O were described in a simple way. First a geographically explicit global model will be presented of N_2O production in soils under natural vegetation, the major single global source (chapter 2). This 1°x1° resolution model is based on the current understanding of the factors regulating N_2O production. It is simple, because it needs to be applicable at the global scale. The direct emission of N_2O to the atmosphere from mineral fertilizer use and nitrogen inputs from animal excreta will be discussed on the basis of the measurements published in the past decades in chapter 3.

During the work it became clear that validation of emission inventories necessitates the inclusion of estimates for all sources or at least the major ones. Chapter 4 describes estimates for the major known sources of N_2O, and a comparison of emission estimates with inverse modelling results from literature. Chapter 5 discusses the difficulty of describing the cycling of N through soils. The example taken is for the Atlantic Zone in Costa Rica, where enhanced N_2O fluxes were observed in recently cleared fields. The results of the comparison made in chapter 4 form a priori estimates that are validated with a three-dimensional atmospheric transport model in chapter 6.

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Global analysis of the potential for N_2O production in natural soils

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Chapter 2

GLOBAL ANALYSIS OF THE POTENTIAL FOR N_2O PRODUCTION IN NATURAL SOILS

Abstract. A simple global model of the production potential of nitrous oxide (N_2O) in natural soils is developed to analyze the relative importance, both geographically and seasonally, of the different controls on N_2O production at the global scale. Five major controls on N_2O production are included: (1) input of organic matter, (2) soil fertility, (3) soil moisture status, (4) temperature, and (5) soil oxygen status. Indices for the controls are derived from global gridded (1°×1° resolution) data bases of soil type, soil texture, NDVI and climate. The model explains close to 60% of the variability found in measurements reported at about 30 sites in six different ecosystems throughout the world. Although this result is reasonable for global analyses, the correlation is considered insufficient to make global estimates of nitrous oxide emission with confidence. The model confirms conclusions from earlier studies that the major source regions of nitrous oxide are in the tropics.

2.1. INTRODUCTION

Nitrous oxide (N_2O) is present in the atmosphere in trace quantities. Its concentration in 1990 was about 310 parts per billion by volume (ppbv), about 1000 times less than that of CO_2, and it is increasing at the rate of about 0.8 ppbv y^{-1} (Watson et al., 1992). The seemingly small growth rate, about 0.25% y^{-1}, is the result of a large imbalance (about 30%) between the sources and sinks (Prinn et al., 1990; Khalil and Rasmussen, 1992). Despite its low abundance in the atmosphere, N_2O plays an important role. Its long lifetime of ~132 years (Isaksen et al., 1992) means the system has a long memory of its emission history. The radiative forcing of N_2O is, molecule for molecule, about 200 times that of CO_2 (Isaksen et al., 1992). In the stratosphere the principal mechanism for N_2O destruction is by photolysis; in addition, N_2O is destroyed by reaction with excited oxygen atoms (Crutzen, 1976). The latter reaction is the largest source of stratospheric NO (nitric oxide), initiating a complex set of gas phase reactions that lead to catalytic ozone destruction.

Neither the sources nor the causes for the increase in N_2O are well known. It is generally accepted that the most important source is natural soils, seconded by emissions from the oceans (Seiler and Conrad, 1987) although there is significant uncertainty regarding the distribution and magnitude of the sources themselves. For some time it was thought that, like CO_2, the primary cause for the increasing concentration was combustion of fossil fuels, in particular, coalburning power plants producing electricity (Hao et al., 1987). However, identification of an artifact in the flask sampling procedure ruled out combustion (including biomass burning) as the major cause of the trend (Muzio and Kramlich, 1988). Minor sources identified so far include agricultural fields amended with nitrogenous fertilizers, animal manure, aquifers, sewage, industry, automobiles, biomass burning, land clearing, and trash incineration (Watson et al., 1992; Khalil and Rasmussen, 1992). It is not clear if these sources can account for the secular trend.

In soils and aquatic systems, microbial processes are responsible for both the production and consumption of atmospheric nitrous oxide and nitric oxide. In vitro experiments demonstrate significant N_2O production by both denitrifiers (Firestone et al., 1980) and nitrifiers (Yoshida and Alexander, 1970; Blackmer et al., 1980; Lipschultz et al., 1981; Poth and Focht, 1985). Recently, it was observed that N_2O may also be formed by other processes (Robertson and Tiedje, 1987; Tiedje, 1988), but the significance of these processes on a global scale, and their mechanisms and regulators, are poorly known.

Denitrification comprises the group of processes whereby nitrogenous oxides, principally nitrate (NO_3^-) and nitrite (NO_2^-), are reduced to dinitrogen gases (N_2), nitrous oxide and nitric oxide (Firestone and Davidson, 1989). It occurs under oxygen-limited conditions; under strictly anaerobic conditions, nitrous oxide and nitric oxide may also serve as electron acceptors. Several processes, including abiotic ones, all match this definition (Firestone and Davidson, 1989). Under natural conditions, the nitrate available for denitrification depends on the rate of nitrification (Myrold and Tiedje, 1985; Tiedje, 1988). With relatively high nitrate supply, denitrification is related primarily to the amount of available organic compounds (Ottow et al., 1985) and N_2O production shows a strong positive correlation with both CO_2 evolution and available carbon (Eaton and Patriquin, 1989).

In nitrification, ammonia (NH_4^+) is oxidized to nitrite (NO_2^-) or nitrate (NO_3^-). In natural soil ecosystems, the ammonia comes mainly from decomposition and mineralization of organic matter. Nitrogen inputs to natural ecosystems come from N deposition and biological dinitrogen fixation (Boring et al., 1988). Major regulators of nitrification are temperature, oxygen, nutrient availability, carbon and nitrifiable N sources. Under oxygen-limited conditions, nitrifiers can use NO_2^- as a terminal electron acceptor to avoid accumulation of the toxic NO_2^-, whereby N_2O is produced (Poth and Focht, 1985). While denitrification is inhibited by oxygen, nitrification is an aerobic process. Therefore denitrification in constantly anaerobic systems is low since under such conditions, nitrification is blocked by lack of oxygen (Sahrawat and Keeney, 1986; Bowden, 1986).

There is considerable controversy about the relative importance of nitrification and denitrification for N_2O production. Many researchers have suggested that nitrification is the major source of N_2O in "aerobic" agricultural soils (Ryden et al., 1978; Breitenbeck et al., 1980; Bremner and Blackmer, 1981; Klemedtsson et al., 1988) and in natural semiarid "well aerated" soils (Anderson et al., 1988; Parton et al., 1988). In peat sediments (Gordon et al., 1986), certain tropical forests (Livingston et al., 1988; Keller et al., 1988; Robertson and Tiedje, 1988) and some temperate forests (Eaton and Patriquin, 1989), denitrification may be the major process involved. Apparently, nitrification is a relatively constant process across ecosystems while denitrification rates are temporally and spatially variable (Firestone and Davidson, 1989).

Estimates of the annual N_2O release from natural soils illustrate the uncertainty about this source. The ranges are 7-16 Tg (Tg = 10^{12}g) (Bowden, 1986), 3-25 Tg (Banin, 1986), 3-9 Tg (Seiler and Conrad, 1987), and 2.8-7.7 Tg (Watson et al., 1992). Most of these studies rely on one or a few flux measurements, multiplied by areas of broad vegetation groups, to derive global emissions. These "representative measurement" approaches cannot explain the extreme variability of fluxes observed both in space and time. Fluxes of N_2O from temperate grasslands have been modeled extensively (e.g., Parton et al., 1988); a model based on rainfall events and detailed description of microbiological and physical soil processes was developed by Li et al. (1992). While conceptual models of nitrification and denitrification exist (e.g., Robertson, 1989; Firestone and Davidson, 1989; Davidson, 1991), extrapolation from site-specific measurements or models to regional or global emissions is a considerable challenge. Schimel et al. (1988) and Matson et al. (1989) stressed the need for stratification of ecosystems to describe the variability of environmental conditions responsible for N_2O fluxes: Matson and Vitousek (1990) stratified tropical forests primarily on the basis of soil fertility. Such approaches lead to better-justified regional estimates of N_2O emissions.

The global scale of the modeling presented here makes description of soil processes a difficult task. It is necessary to identify the major controls, and to describe and quantify their relative importance in a simple way. Our working hypothesis is that, since N_2O can be produced by both nitrifiers and denitrifiers, the potential for N_2O production in upland soils under natural conditions is related to the amount of nitrogen cycling through the soil-plant-microbial biomass system (Matson and Vitousek, 1987; Firestone and Davidson, 1989), hence to the N mineralization, nitrification, and denitrification rates (Matson and Vitousek, 1987; Robertson and Tiedje, 1984; Martikainen, 1985). In this "process pipe" or "hole in the pipe" concept (Firestone and Davidson, 1989; Davidson, 1991) the size of the holes or leaks in the pipe through which N_2O and NO escape is determined by factors that control the partitioning of the reacting N species to N_2O or more reduced/oxidized products. The rate at which N moves through the process pipe determines the importance of the leaks. This means that low denitrification rates lead to low trace gas release, regardless of the relative proportions of end products (Firestone and Davidson, 1989; Davidson, 1991).

We present a simple global model to analyze the geographic and seasonal variations of the various controls of N_2O production and their relative importance in the total annual emission. The focus is natural ecosystems, the largest single source term in the present-day N_2O budget. Given the paucity of available flux measurements, an immediate goal is to test the simple model of N_2O fluxes and to identify gaps where measurements and analyses are needed to reduce uncertainties about this source term.

The major controls on N_2O emissions in the model are discussed in section 2. The synthesis of these regulators into a simple schematic model for determining the N_2O emission potential is described in section 3. The one-dimensional model is applied at every 1°×1° gridbox globally. Modeled distribution of N_2O production potential and its sensitivity to the controlling factors are presented in sections 4.1 to 4.3. Regression relationships between modeled and observed N_2O emission values are presented in section 4.4, yielding an equation to translate the modeled non-dimensional N_2O potentials into dimensional fluxes (section 4.5). Global data sets of surface climate, soil properties, and satellite observations of the normalized difference vegetation index (NDVI), used to derive the geographic variation of the controls used in the model, are discussed in the appendix.

2.2. MAJOR CONTROLLING FACTORS OF N_2O PRODUCTION IN SOILS

The global model of N_2O production potential is based on the hypothesis that N_2O fluxes are directly related to the rate of nitrogen cycling through the soil-plant-microbial system. Hence we need to describe general patterns of decomposition and N mineralization, and, given the amount of N available in the soil, the major determinants of nitrification and denitrification.

Climate, vegetation, and soil are fundamentally interrelated to determine the ecosystem that develops in a particular area. The parent rock and soil type determine the type of vegetation via nutrient availability. Sites with low nutrient availability select for species with low nutrient demands producing low-quality litter which decomposes slowly and further reduces nutrient availability (Anderson, 1992). Soil properties are also molded by the vegetation, particularly the amount and nature of organic matter formed during decomposition of plant detritus. Soils can have low fertility for various reasons. Low pH causes low solubility of a number of major and minor trace elements while the solubility of other elements (e.g., aluminum) increases. Highly weathered soils of the tropics (e.g., Ferralsols and Acrisols) usually contain less phosphorous than younger, less weathered soils of the temperate regions; they may even show P-fixation (Jordan, 1985; Duxbury et al., 1989; Driessen and Dudal, 1991).

Decomposition rates (Jordan, 1985) and mineralization processes are closely related to soil fertility because of feedbacks between litter quality, decomposition rates and nutrient availability (Adams et al., 1970; Zimka and Stachurski, 1976; Miller, 1981; Vitousek, 1982). In general, concentrations of all major nutrients, including nitrogen, in aboveground and belowground forest biomass are markedly higher in fertile soils than in poor ones (Vitousek and Sanford, 1986).

Decomposition and mineralization rates are determined also by litter properties, most notably lignin concentration and C/N ratios. With low lignin concentrations (~10-15%) decomposition rates can be predicted from nitrogen concentrations or C/N ratios, while with high lignin concentrations the lignin/N ratio or initial lignin determines decomposition (Anderson, 1992). The C/N ratio of organic material supplied to the soil influences the amount of N immobilized in microbial biomass. This influences N availability which in turn may affect nitrification and denitrification activities

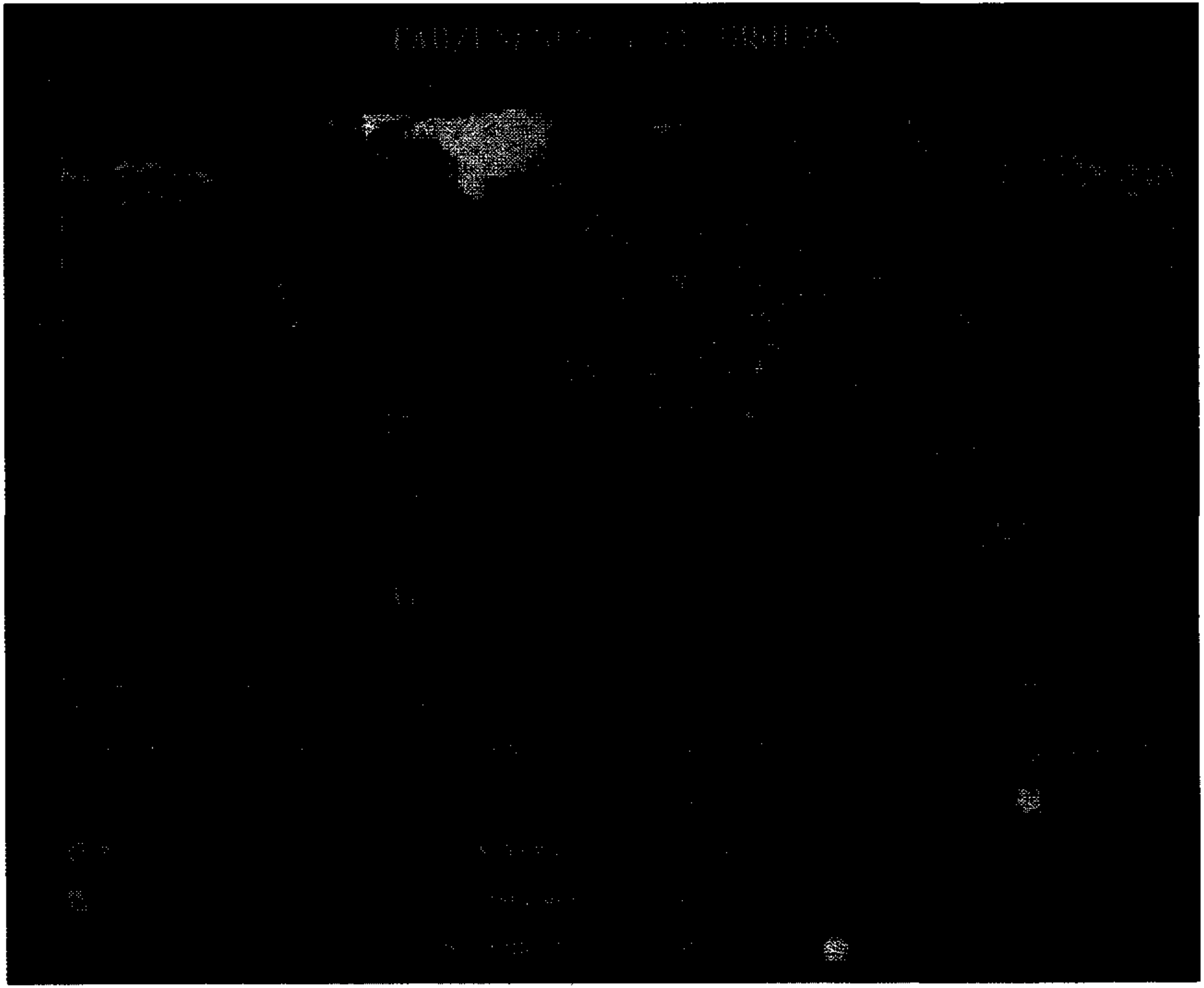

Plate 1. Distribution of major soil groups from the data set of Zobler (1986) digitized from FAO (1974-1981) soil maps. See Table 1 for descriptions.

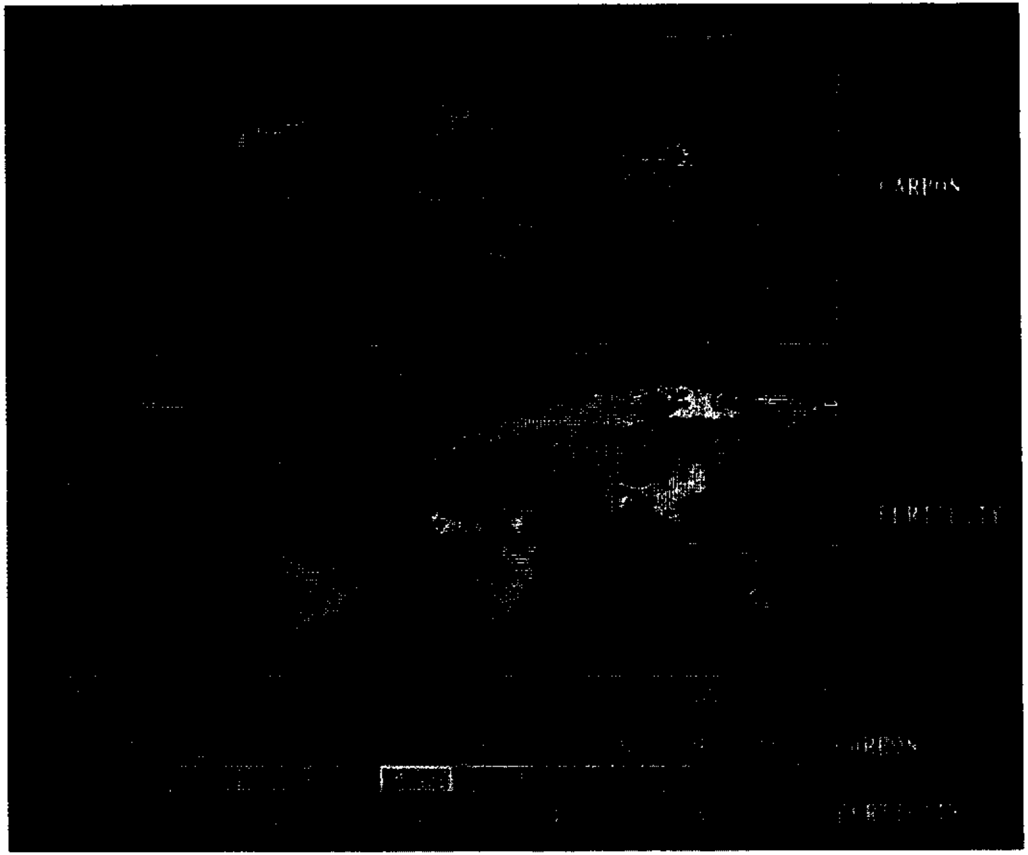

Plate 2. Distribution of factors used in the model: CARBON (upper panel), representing the input of organic matter, is NDVI scaled from 0-10, and FERT (lower panel), soil fertility, scaled from 1-5 (see Table 1).

and products of denitrification (see Ottow, 1985). The low C/N ratio in tropical forests leads to rapid and more complete decomposition of organic matter inputs (Duxbury et al., 1989) and high N mineralization rates in these soils (Vitousek and Sanford, 1986).

Soil moisture and temperature control soil processes at all levels and are important factors governing the speed of organic matter decomposition. Because of year-round high temperatures, soil processes in the humid tropics can remain active throughout the year, and annual nutrient cycling can be faster than in regions where cold or drought interrupts these processes. Jenkinson and Ayanaba (1977) estimated that decomposition rates in the tropics are four times those in temperate zones, while decomposition in mediterranean regions has been found to be twice that in temperate climates (Oades, 1988). Measurements of soil organic matter stocks and leaf litterfall suggest that decomposition rates mimic global patterns of temperature and moisture. Post et al. (1985) noted that the C/N ratio is generally lower in warm climates. This is consistent with the observation that tropical forests generally cycle 2-4 times more nitrogen between soil and vegetation than do most temperate ecosystems (Vitousek, 1984; Jordan, 1985). However, this may be caused by ecosystem characteristics other than temperature. Apparently, nitrogen is not limiting growth in these tropical systems but other nutrients, such as phosphorous in strongly leached soils, may be limiting (Vitousek, 1984). Because of nitrogen limitation, temperate forests generally have higher C/N ratios in litter and slower N mineralization rates than do tropical forests (Robertson and Tiedje, 1984; Jordan, 1985; Vitousek and Sanford, 1986; Vitousek and Matson, 1988). Fluxes in temperate forests are much lower than those in most tropical environments, which may be explained by lower N availability and less favorable moisture and temperature conditions.

Soil-water content is the balance of inputs from rainfall, leaching and capillary rise, and consumption by plants. It has been shown to influence N_2O emissions from well-drained soils (Mosier and Parton, 1985; Klemedtsson et al., 1988; Groffman and Tiedje, 1988). Aerobic microbial activity increases with soil-water content until water displaces air and restricts oxygen diffusion so that maximum rates of microbial respiration, nitrification and mineralization occur at the highest water content at which soil aeration remains nonlimiting. This critical water content is strongly dependent on soil texture. In general, microbial activity peaks at 60-80% of field capacity (Linn and Doran, 1984), the amount of water held at soil-water potentials of about 10 kPa (10 kPa = 0.1 bars). Nitrification and associated N_2O production also show maximum activity at 60-80% of field capacity (Greaves and Carter, 1920; Davidson, 1991).

Optimum conditions for denitrification may occur at water contents from 80% to over 100% of field capacity (Linn and Doran, 1984; Klemedtsson et al., 1988). There is, however, a clear hysteretic effect whereby respiration and denitrification rates slowly decrease monotonically during the drying phase (Groffman and Tiedje, 1988). Soils close to saturation show low N_2O production (Davidson, 1991) because N_2O diffusion may be limited (Sahrawat and Keeney, 1986; Keller et al., 1986) resulting in a greater fraction of N_2O reduced to N_2. Uptake of N_2O from the atmosphere by wet soils may occur under very wet conditions (Ryden, 1981, 1983; Letey et al., 1981; Smith et al., 1983; Keller et al., 1986).

Wetting of dry soils causes pulses in N mineralization, nitrification and N_2O fluxes (Denmead et al., 1979; Letey et al., 1981; Mulvaney and Kurtz, 1984; Sextone et al., 1985; Mosier et al., 1981, 1986; Parton et al., 1988; Vitousek et al., 1990). Letey et al. (1981) showed that release of N_2O from the soil to the atmosphere is enhanced by alternate drying and wetting of soils although peaks in N_2O production may decline with subsequent wetting events.

Overall denitrification activity is strongly stimulated at low oxygen pressures (Colbourn and Harper, 1987; Firestone and Davidson, 1989) but associated N_2O fluxes are low under anaerobic conditions (e.g., Terry et al., 1981; Davidson, 1991). Oxygen status of soils is controlled by the interplay between water inputs, oxygen supply, and oxygen consumption. Soil-water content is crucial as oxygen diffuses 10^4 slower in water than in air (Nye and Tinker, 1977).

Soil oxygen is consumed by root respiration and microbial activity. Oxygen consumption by microorganisms is driven by the supply of carbon and, under dry conditions, by water as a stimulator of metabolic activity. Denitrification is often found in "hot spots" created by decomposing organic matter which generates anaerobic microsites (Dowdell and Smith, 1974; Duxbury et al., 1982; Parkin, 1987; Schmidt et al., 1988). This phenomenon may explain some of the high spatial variability of soil denitrification commonly observed (Ryden et al., 1978; Rolston et al., 1978; Breitenbeck et al., 1980; Bremner et al., 1980; Mosier et al., 1981; Folorunso and Rolston, 1984; Colbourn et al., 1984; Goodroad and Keeney, 1985; Colbourn and Harper, 1987).

A strong relationship exists between soil texture and denitrification activity (Groffman and Tiedje, 1989). Fine-textured soils have more capillary pores within aggregates than do sandy soils, thereby holding soil water more tightly. As a result, anaerobic conditions may be more easily reached and maintained for longer periods within aggregates in fine-textured soils than in coarse-textured soils. Soil-drainage characteristics also influence denitrification activity (Colbourn and Harper, 1987; Groffman and Tiedje, 1989) by affecting soil aeration.

Studies on the effect of temperature on N_2O fluxes show Q_{10} values of 5 at 10°C to 1.5 at 30°C (Mosier and Parton, 1985), 2.8 between 15° and 25°C (Denmead et al., 1979), 2.28 between 10° and 20°C, 1.99 from 20° to 30°C and 1.35 at 30° to 40°C (Blackmer et al., 1982). Temperature may not control the release of N_2O in ecosystems where soil nitrogen temporarily accumulates due to wet-dry cycles or freeze-thaw cycles (Anderson and Poth, 1989). Denitrification during early spring and autumn in temperate climates may account for a significant portion of the annual N_2O released (Keeney et al., 1979; Goodroad and Keeney, 1984a; Schmidt et al., 1988; Christensen and Tiedje, 1990).

Soil pH also has a marked effect on the products of denitrification. Denitrification rates are low under acid conditions and more rapid under slightly alkaline conditions, but the N_2O fraction may be larger at low soil pH (Focht, 1974; Goodroad and Keeney, 1984b; Martikainen, 1985; Eaton and Patriquin, 1989; Brumme and Beese, 1992), particularly with an adequate nitrate supply. This is commonly attributed to the sensitivity of N_2O reductase to proton activity (Alexander, 1977). Simply lowering soil pH does not always increase the N_2O ratio suggesting that the soil and its resident organisms, rather than pH per se, determine the N_2O fraction (Ottow et al., 1985). This may be due to the observed apparent adaptation of strains of denitrifiers to low pH (Parkin et al., 1985).

2.3. MODEL DESCRIPTION

The following parameters were selected as control variables in the N_2O model: (1) input of organic matter (CARBON); (2) soil fertility (FERT); (3) temperature effect on organic matter decomposition and nitrogen mineralization (SOD); (4) soil-water availability affecting decomposition, mineralization and nitrification (H2O); and (5) soil-oxygen limitation affecting denitrification (O2).

We assume that organic matter input is proportional to litter amount which in turn is proportional to net primary productivity of the vegetation. The mobilization rate of nitrogen in organic matter is proportional to the rate of decomposition and mineralization as determined by soil temperature, soil moisture and soil fertility. We recognize that composition of the organic material is also an important determinant of decomposition rate. However, there is no global dataset or proxy dataset for lignin content or C/N ratios in litter, and so we have not included these controls explicitly. As C/N ratio may be related to temperature (Post et al., 1985) and soil fertility (Vitousek and Sanford, 1986), their effect on decomposition and mineralization rates may be indirectly captured by temperature (SOD) and fertility (FERT) factors.

Plate 3. Distribution of soil characteristics derived from the soil data base. Texture class (upper panel) determines the soil storage capacity (SSC; middle panel). Refer to Table 2 for descriptions of numbered texture classes and soil storage capacity. Drainage indices (lower panel) for soil units are listed in Table 1.

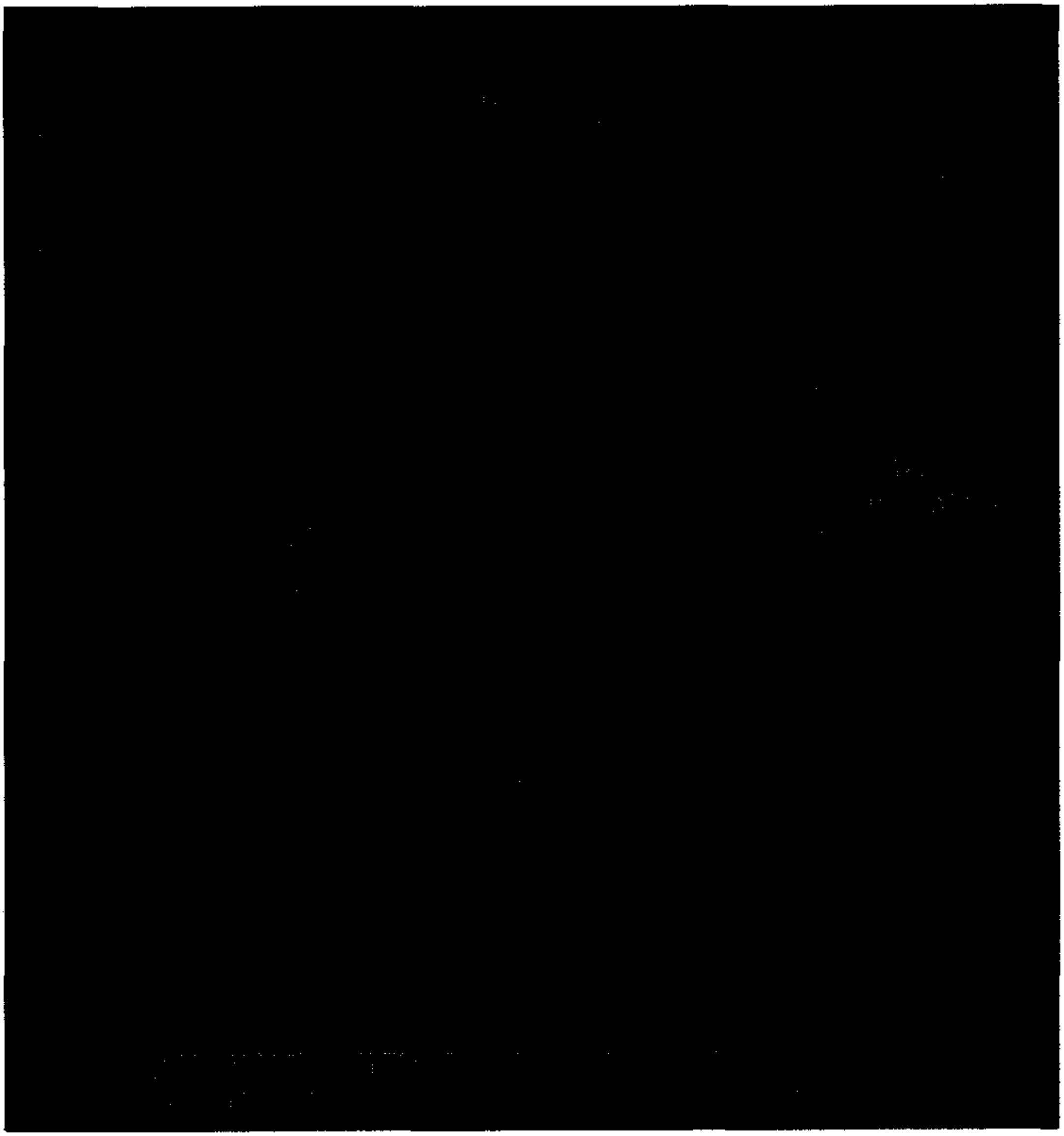

Plate 4. Distribution of annual sums of monthly indices for soil organic matter decomposition for the reference case (SOD1; upper panel), soil-water availability (H2O; middle panel) and soil-oxygen limitation (O2; lower panel).

Soil pH has also been identified as a regulator for N_2O production. Because global data on soil pH is lacking, this soil property is not included as a separate factor in this study. Instead, we assume that soil pH and soil fertility are closely related and hence complementary.

A central part of the model is a description of soil-water regulation of nitrification / denitrification. We use a bucket model of water balance, which includes variations in soil drainage and topsoil texture and determines monthly water status of the topsoil. If the soil-water status is high, there is high water availability and low oxygen availability - conditions favorable for denitrification. The model is described below and illustrated schematically in Figure 1. Three of the five regulators vary monthly: SOD, H2O and O2. The remaining two, FERT and CARBON, are constant through the year.

Because of the lack of quantitative relationships between N_2O fluxes and the various controlling parameters for soil types on a global scale, our strategy was to first translate ideas about relative importance into ranked nondimensional indices. The indices range from 0 to 10 or from 1 to 5, with high numbers signifying importance for N_2O production. Such translations may be straightforward for numeric data such as temperature. For control factors such as soil fertility, a subjective ranking of soil units was carried out. The suite of control indices were then combined to form indices for N_2O fluxes.

The focus of the model requires data which are global in domain and which span at least a year. The finest spatial resolution of the primary data sets used is 1° latitude by 1° longitude, about 110 km × 110 km at the equator. For these reasons, the model cannot resolve episodic effluxes of N_2O after rainstorms, localized "hot spots" and high spring and autumn emissions in temperate ecosystems which are often reported. The importance of such high-frequency, local events in the global budget has not been established.

The primary gridded data bases on climate, soil type, soil texture, and vegetation are discussed in the appendix. The global distribution of soil groups is shown in Plate 1.

- *Input of organic matter (CARBON).* Under natural conditions, litterfall and root decay are the major sources of carbon and nitrogen to the soil. Most ecosystems have abundant surface litter throughout the year. Therefore the seasonal variation of C and N mobilization in the litter is governed more by decomposition rates rather than by seasonal variations in litterfall. We assume that the geographic pattern of annual litterfall is the same as that of annual net primary productivity (NPP). NPP is the result of soil and environmental conditions, and for many temperate ecosystems also the result of nitrogen availability. Hence NPP may only be correlated indirectly to nitrogen.

The satellite-derived normalized difference vegetation index (NDVI; see appendix) has been shown to be a good correlate for NPP (Goward et al., 1986; Box et al., 1989). The annual integral of the NDVI, rather than monthly NDVI values, is used because litterfall is asynchronous with productivity and with nitrogen mineralization and nitrogen may be immobilized in microbial biomass before it is liberated.

Monthly NDVI composites for 1984 are gridded at 1° resolution for the globe, and summed to produce the annual integral. Monthly NDVI values range from -0.1 to 0.5, and annual totals range from -0.1 to 4.0. For consistency with other factors used in the study, the NDVI totals are rescaled to range from 0 to 10, for the index CARBON (Plate 2). Use of the NDVI captures the variability of NPP at the same resolution as that of the soil data.

- *Soil fertility (FERT).* In this study, soil fertility is the inherent capability of soil material to supply nutrients to plant roots (see Sanchez, 1976; Brady, 1976). This definition excludes soil N availability which is a combined result of vegetation, climate, and soil. With this definition, mineralization of nitrogen is indirectly determined by the combination of CARBON, FERT, SOD and H2O.

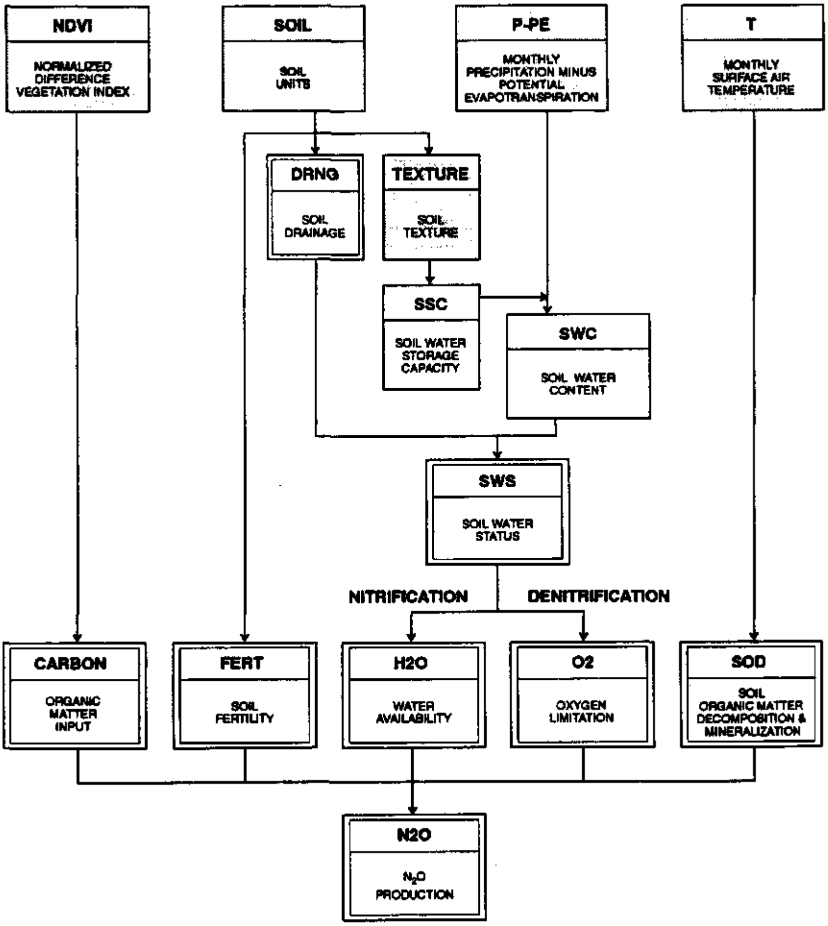

Figure 1. Schematic diagram of the model of N_2O production. Double bordered boxes denote nondimensional indices.

Soil characteristics important for inherent fertility are soil pH, cation exchange capacity, base saturation, and amounts of weatherable minerals; in some cases P fixation also plays a role. The scale for soil fertility is adapted from Food and Agriculture Organization (FAO) (1981) for the major FAO/Unesco soil groups (Table 1); indices for some individual soil units within groups vary due to their diagnostic horizons or properties. For example, Nitosols (Table 1, soil group 12) are highly fertile soils common to the tropics. Within the group of Nitosols, the dystric Nitosols are assigned a lower index because their base saturation is less than 49% of the cation exchange capacity (FAO/Unesco, 1974-1981). In general, leached soils in wet climates have lower fertility than soils with a less pronounced downward flow of water over the year. Ferralsols (soil group 13) are strongly leached soils with variable charge characteristics, dominated by kaolinite and hydrated oxides of alumina and iron; their fertility is low due to low cation exchange capacity, the presence of alumina at the exchange complex, low content of weatherable minerals and

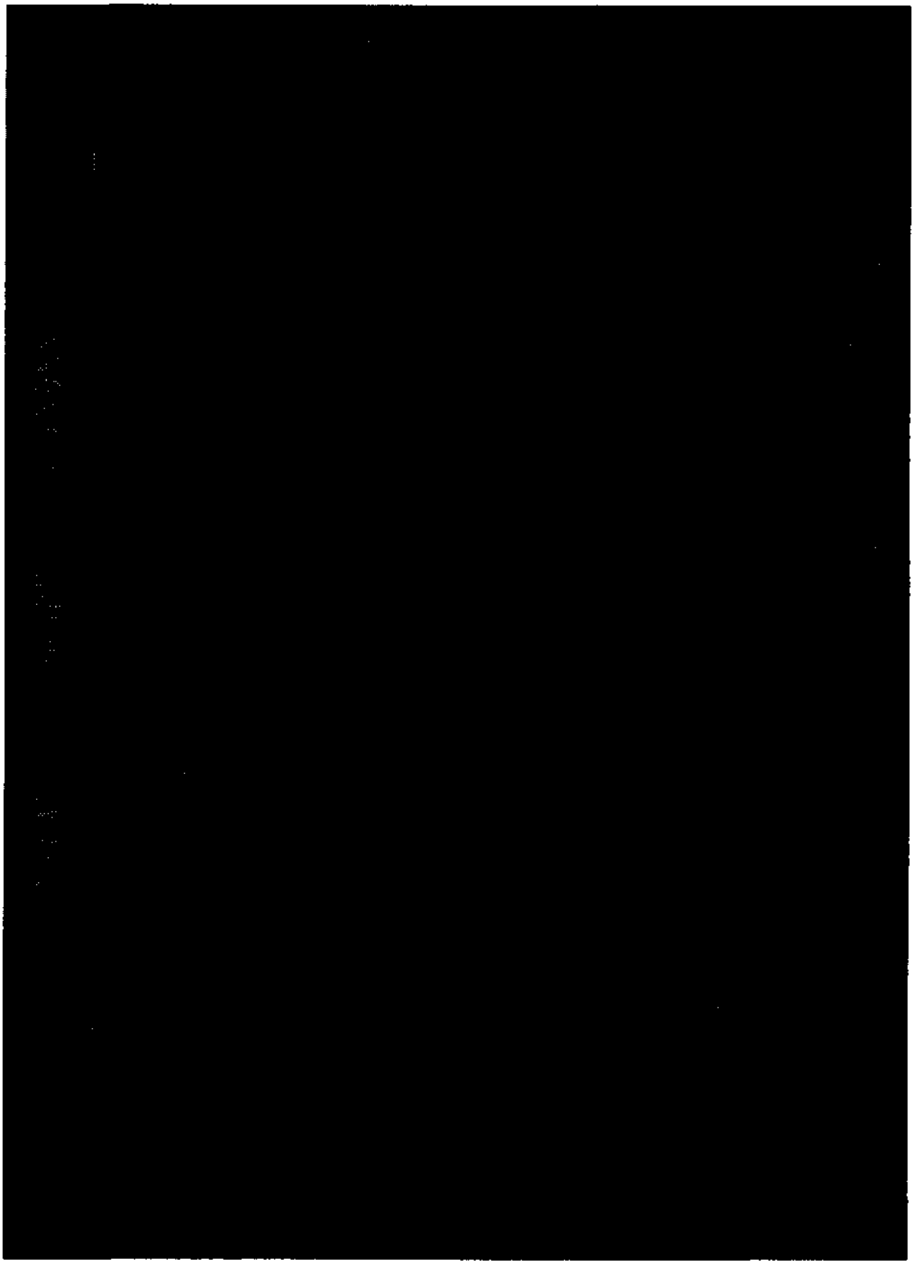

Plate 5. Distribution of annual sum of monthly N_2O production potential N2O

phosphorous fixation. Podzols (soil group 2) are also infertile, generally because of low cation exchange capacity, low content of weatherable minerals and alumina saturation.

The global distribution of soil fertility index (FERT) is shown in Plate 2. Globally, soils are characterized predominantly by moderately low fertility: soils covering slightly more than half the ice-free land area have fertility indices of two. About one quarter of the land is of intermediate fertility, while the other fertility classes account for <10% each.

- *Temperature effect on rate of soil organic matter decomposition and N mineralization (SOD).* Rates of decomposition and nitrogen mineralization are regulated by a number of soil factors including temperature, moisture, fertility, and texture. All these factors, except soil temperature, are represented in the factors H2O and FERT. Lacking a global data set on soil temperatures, we use monthly climatologies of surface air temperature (Shea, 1986); this might introduce timing errors of up to 1-2 months particularly in middle to high latitudes. In this study, we investigated three temperature dependencies for SOD. The three SOD functions are shown in Figure 2.

The first (SOD1) is an exponential function obtained for semiarid grasslands (Mosier and Parton, 1985; Parton et al., 1988) describing the temperature effect on N_2O fluxes: SOD1 = 10 at T = 50°C, and SOD1 = 0 for T < 0°C, with a rapid increase between 10° and 30°C. The second equation (SOD2) is a quadratic function adapted from Parton et al. (1987) describing decomposition of soil organic matter in grasslands. The optimum (SOD2 = 10) is at T = 33°C. To avoid negative values, SOD2 is set to zero at T < 0°C and at T > 49°C. The third relationship (SOD3) is a set of linear functions for four broad ecosystem groups: broadleaf vegetation, needleleaf vegetation, grassland, and tropical vegetation. The functions are derived from observations of CO_2 evolution from soils and mean monthly temperature (Fung et al., 1987): SOD3 = 10 at T = 50°C. The different slopes of the four functions reflect the differences in litter composition for the broad biome groups. For comparable temperature ranges, grassland, with a high fraction of easily decomposable detrital material, has a faster relative decomposition rate than that of needleleaf woody vegetation.

Differences between the SOD1 and SOD2 functions are minor: the most marked differences are expected in temperate climates with mild winters. At temperatures between 0° and 10°C, SOD2 yields the lowest values; in that temperature range, SOD3 gives much higher values than the other functions. The three functions give similar results for the tropics. The SOD1 equation yields the best correlation with the measurement data (see Section 4.4). For that reason, SOD1 was chosen as the reference case. The global distribution of SOD1 is shown in Plate 4 (annual total of the monthly values).

- *Soil water and soil oxygen.* A key to determining moisture conditions for decomposition and for distinguishing the pathways of nitrification versus denitrification is the degree of saturation and aerobicity of the soil which are determined by the soil water content (SWC) and its relation to the maximum amount of water held in the soil (soil water storage capacity, SSC), and by soil drainage properties. Topsoils are the primary sites for N_2O production since maximum microbial activity and the most intensive rooting occur in this soil layer. Nitrification may dominate in the upper few millimeters (Seiler and Conrad, 1981) while denitrification may dominate below, particularly during periods of high water content (Goodroad and Keeney, 1985).

Therefore, we consider the topsoil, i.e. upper 30 cm of soil, as the zone of N_2O production. Some physical properties in zones below 30 cm, that influence water and air movement in the topsoil, are included in the soil drainage characteristics.

Table 1. Indices of soil fertility (FERT) and drainage (DRNG) assigned to different soil units †

	Soil Unit	FERT	DRNG	Area (10^6 ha)	
I	SOILS WITH PERMAFROST				674
1	**Soils with permafrost within 200 cm**				674
	gelic Cambisols	3	5	232	
	gelic Gleysols	3	5	148	
	gelic Histosols	2	5	66	
	gelic Planosols	2	5	-	
	gelic Regosols	2	5	228	
II	SOILS CONDITIONED BY SUBHUMID CLIMATES				885
2	**Soils with illuviation of organic matter and/or sesquioxides**				493
	ferric Podzols	2	1	-	
	gleyic Podzols	2	3	44	
	humic Podzols	2	1	31	
	leptic Podzols	2	1	19	
	orthic Podzols	2	1	398	
	placic Podzols	2	2	1	
3	**Soils with strong textural differentiation with slowly permeable subsoil**				141
	dystric Planosols	1	3	1	
	eutric Planosols	2	3	63	
	humic Planosols	2	3	1	
	mollic Planosols	2	3	16	
	solodic Planosols	1	3	60	
4	**Soils with argic horizon ‡ underlying a bleached subsurface horizon**				251
	dystric Podzoluvisols	2	2	66	
	eutric Podzoluvisols	3	2	159	
	gleyic Podzoluvisols	3	3	26	
III	SOILS CONDITIONED BY DRY SUBHUMID (SUB-) TROPICAL OR SUBHUMID TEMPERATE CLIMATES				950
5	**Soils with argic horizon***				950
	albic Luvisols	3	2	114	
	calcic Luvisols	4	2	63	
	chromic Luvisols	4	2	249	
	ferric Luvisols	3	2	264	
	gleyic Luvisols	4	3	73	
	orthic Luvisols	4	2	162	
	plinthic Luvisols	3	4	23	
	vertic Luvisols	4	3	2	
IV	SOILS CONDITIONED BY STEPPE CLIMATES				897
6	**Soils with argic horizon* and organic matter accumulation**				27
	gleyic Greyzems	3	3	11	
	orthic Greyzems	3	2	17	
7	**Soils with organic matter accumulation, annual P > E §**				153
	calcaric Phaeozems	5	1	4	
	gleyic Phaeozems	5	3	11	
	haplic Phaeozems	5	1	58	
	luvic Phaeozems	5	2	80	
8	**Soils with organic matter accumulation, annual P = E §**				225
	calcic Chernozems	5	1	32	
	glossic Chernozems	5	1	6	
	haplic Chernozems	5	1	124	
	luvic Chernozems	5	2	63	
9	**Soils with organic matter accumulation, annual P < E §**				492
	calcic Kastanozems	5	1	14	
	haplic Kastanozems	5	1	225	
	luvic Kastanozems	5	2	253	
V	SOILS CONDITIONED BY LIMITED AGE				758
10	**Weakly developed soils**				758
	calcic Cambisols	4	1	86	
	chromic Cambisols	3	1	81	
	dystric Cambisols	3	1	245	
	eutric Cambisols	4	1	230	
	ferralic Cambisols	3	1	23	
	gleyic Cambisols	3	3	15	
	humic Cambisols	4	1	60	
	vertic Cambisols	4	2	18	
VI	SOILS CONDITIONED BY WET (SUB-) TROPICAL CLIMATES				2228
11	**Leached soils with argic horizon***				903
	ferric Acrisols	3	2	164	
	gleyic Acrisols	2	3	40	
	humic Acrisols	3	2	41	
	orthic Acrisols	3	2	576	
	plinthic Acrisols	2	4	82	

Table 1. (continued)

	Soil Unit	FERT	DRNG	Area (10^6 ha)	
12	**Strongly weathered soils with deeply developed argic horizon**[‡]				209
	dystric Nitosols	3	1	114	
	eutric Nitosols	5	1	80	
	humic Nitosols	5	1	15	
13	**Strongly leached soils dominated by hydrated oxides**				1116
	acric Ferralsols	1	1	68	
	humic Ferralsols	2	1	27	
	orthic Ferralsols	2	1	506	
	plinthic Ferralsols	1	4	38	
	rhodic Ferralsols	2	1	73	
	xanthic Ferralsols	2	1	404	
VII	SOILS CONDITIONED BY (SEMI-)ARID CLIMATES				2108
14	**Desert soils**				1876
	calcic Xerosols	2	1	253	
	calcic Yermosols	2	1	314	
	gypsic Yermosols	2	1	69	
	haplic Yermosols	2	1	743	
	gypsic Xerosols	2	1	6	
	haplic Xerosols	2	1	133	
	luvic Xerosols	2	2	107	
	luvic Yermosols	2	2	235	
	takyric Yermosols	2	1	16	
15	**Saline and alkaline soils**				232
	gleyic Solonchaks	1	3	27	
	mollic Solonchaks	2	1	5	
	orthic Solonchaks	1	1	92	
	takyric Solonchaks	1	1	3	
	gleyic Solonetz	1	4	1	
	mollic Solonetz	2	3	29	
	orthic Solonetz	1	3	75	
VIII	SOILS CONDITIONED BY THEIR PARENT MATERIAL				1146
16	**Heavy textured cracking soils dominated by clays with swell-shrink properties**				314
	chromic Vertisols	3	3	205	
	pellic Vertisols	3	3	109	
17	**Soils formed in volcanic ash**				111
	humic Andosols	4	1	20	
	mollic Andosols	4	1	12	
	ochric Andosols	4	1	27	
	vitric Andosols	4	1	52	
18	**Weakly developed soils with sand texture**				721
	albic Arenosols	1	1	19	
	cambic Arenosols	2	1	316	
	ferralic Arenosols	1	1	302	
	luvic Arenosols	2	2	84	
IX	SOILS CONDITIONED BY THEIR PHYSIOGRAPHIC POSITION				3377
19	**Soils influenced by a floodplain regime**				245
	calcaric Fluvisols	4	1	61	
	dystric Fluvisols	3	1	35	
	eutric Fluvisols	4	1	138	
	thionic Fluvisols	1	5	11	
20	**Soils influenced by groundwater**				412
	calcaric Gleysols	3	5	16	
	dystric Gleysols	2	5	161	
	eutric Gleysols	3	5	134	
	humic Gleysols	3	5	27	
	mollic Gleysols	3	5	72	
	plinthic Gleysols	2	5	2	
21	**Leptosols (shallow soils, mostly <10 cm thick)**				2273
	Lithosols	2	1	2224	
	Rankers	1	1	3	
	Rendzinas	3	1	46	
22	**Weakly developed soils formed in unconsolidated non-alluvial material**				447
	calcaric Regosols	2	1	189	
	dystric Regosols	1	1	87	
	eutric Regosols	3	1	171	
X	ORGANIC SOILS				172
23	**Histosols (peat soils)**				172
	dystric Histosols	1	5	125	
	eutric Histosols	2	5	47	

† Low index values indicate low fertility and free drainage. Soil units are classified by soil groups (arabic numbers) and soil clusters (roman numbers); the areas of each unit is given in column 4, while those of each group and cluster are in column 5. Total ice-free land area is 13,195 × 10^6 ha. Land ice is 1666 × 10^6 ha.

‡ argic horizon = subsurface horizon with distinctly higher clay content than overlying horizon.

§ P = precipitation; E = evapotranspiration

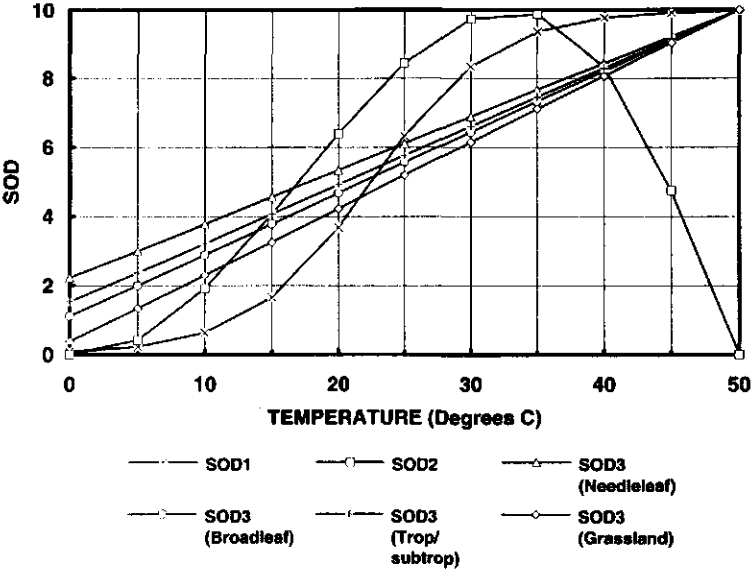

Figure 2. Index of the rates of soil organic matter decomposition and N mineralization (SOD) as a function of temperature. SOD1 (reference) is taken from Mosier and Parton (1985) and Parton et al. (1988); SOD2 from Parton et al. (1987); SOD3 from Fung et al. (1987).

- *Soil water storage capacity (SSC).* Soil water storage capacity is the maximum amount of water held in the upper 30 cm of the soil at field capacity (soil water potential of 10 kPa = 0.1 bar), i.e. when internal drainage and redistribution have ceased. In reality, redistribution of water after wetting is continuous and the field capacity is reached after prolonged periods if at all. Nevertheless, for the scale and purpose of this study, the above definition of SSC is appropriate.

SSC values are listed in Table 2. For most soils, water storage capacity is derived from soil texture based on average soil physical characteristics of major texture classes (Euroconsult, 1989; Landon, 1984). The global distributions of soil texture and SSC are shown in Plate 3. For several soils with special physical properties influencing field capacity, alternative water storage capacities are assigned regardless of soil texture. These are discussed in some detail, since the soils involved cover about 30% of the ice-free land surface of the Earth.

Vertisols (Table 1) are cracking clay soils dominated by montmorillonitic clays with swell-shrink properties. Their water storage capacity may be lower than expected on the basis of their clay content because it is usually difficult to wet these soils. After the initial rain, infiltration rates may decrease as the clay swells and closes the cracks (see Blokhuis, 1991). On the basis of these characteristics, Vertisols are assigned a low storage capacity similar to coarse- and medium-textured soils. In addition, soil units with vertic properties are assigned SSC values similar to those of medium-textured soils.

Because particle size distributions of Andosols and Ferralsols are not easily determined (Dijkerman, 1991), texture information from the FAO/Unesco maps may be less reliable than for other soil groups. In Ferralsols, positively charged hydrated oxides and negatively charged kaolinite form stable aggregates with many biopores resulting in water retention characteristics similar to those of sands or medium-textured soils, although some Ferralsols are fine-textured. Ferralsols are assigned a water-storage capacity of 80 mm, equal to that of medium-textured soils.

Table 2. Soil water storage capacity (SSC) of the upper 30 cm

Texture Class	Soil Texture	SSC † (mm)	γ ‡ (no dim)	δ ‡ (no dim)
1	coarse	40	15	0
2	coarse/medium	60	9	0
6	organic	60	18	0
3	coarse/fine	80	9	0
3	coarse/medium/fine	80	9	0
3	medium	80	9	0
4	medium/fine	100	9	0.1
5	fine	120	6	0.1
	Soil Type §	SSC † (mm)	γ ‡ (no dim)	δ ‡ (no dim)
	Rendzinas	15	15	0
	Lithosols	15	15	0
	Rankers	15	15	0
	Vertisols	60	6	0.1
	vertic Cambisols	80	9	0.1
	Ferralsols	80	9	0
	vertic Luvisols	80	9	0.1
	Andosols	120	9	0

† SSC is shown for major texture classes (Zobler, 1986) and for some soil types (FAO/Unesco, 1974-1981) with limited soil depth or clay minerals having specific soil-moisture retention characteristics. Based on Landon (1984) and Euroconsult (1989).

‡ γ and δ are used in Equation (2) in the computation of transpiration and soil evaporation from the soil- water content; δ expresses the intercept of the soil- water extraction curve β.

§ A full list of soil units and their fertility and drainage indices is presented in Table 1.

Andosols are soils formed in volcanic ash, commonly with high aggregate stability. Water availability in Andosols is generally not lower, and is often higher, than in other mineral soil materials (Van Reeuwijk, 1991), which may not be reflected by their texture. Therefore Andosols are assigned a topsoil storage capacity of 120 mm, equal to SSC values for fine textures.

Leptosols, shallow soils with a depth of ~10 cm or less, are assigned a low storage capacity of 15 mm since their shallowness is assumed to exert primary influence on water storage.

- *Soil water budget model.* The monthly change in soil moisture is the difference between the supply, and demand of moisture at the surface. Supply is governed mainly by precipitation, while demand is governed by evaporation from soils and transpiration by plants.

Several soil moisture models are used in general circulation models, ranging from the simple bucket model of Manabe (1969), where SSC is uniformly 15 cm, to the simple biosphere model (SiB) of Sellers et al. (1986) and the complex biosphere-atmosphere scheme (BATS) of Dickinson et al. (1986) which take into account differential effects of biomes on soil-water balance. The recent models distinguish soil evaporation from plant evaporation and transpiration, as well as model explicitly vertical profiles of soil moisture. We note that in a bucket model where supply and demand are independent of soil moisture itself, the solution to the soil moisture equation is not uniquely determined; it is dependent on the initial soil water content assumed unless SWC =

0 or SWC = SSC (runoff) sometime during the year.

We employed a simple soil-moisture model whose solution does not depend on an arbitrarily chosen initial condition. We adapted the Mintz and Serafini (1981) model for calculating monthly soil-water content. In this model, net supply is the difference between monthly precipitation (P_m) and evaporation from wet canopies (EI_m), while demand is the sum of transpiration through plants and evaporation from soils (ETS_m).

$$SWC_m = SWC_{m-1} + (P_m - EI_m) - ETS_m \qquad (1)$$

Potential evapotranspiration (PE_m), maximum moisture demand by the atmosphere, is calculated from the surface air temperature according to Thornthwaite (1948). Mean monthly surface air temperatures (T_m) and precipitation (P_m) are obtained from Shea (1986). Three moisture regimes are considered, depending on the relation between precipitation and potential evapotranspiration:

$$\begin{array}{ll} P_m = 0 & EI_m = 0 \\ & ETS_m = PE_m \times \beta_m \times \alpha \\ P_m < PE_m & EI_m = P_m \\ & ETS_m = (PE_m - P_m) \times \beta_m \times \alpha \\ P_m \geq PE_m & EI_m = PE_m \\ & ETS_m = 0 \end{array} \qquad (2)$$

where: $\alpha = 0.4$ and

$$\beta_m = 1 - e^{-\gamma[\{SWC_{m-1} + (P_m - EI_m)/2\}/SSC - \delta]}$$

The coefficient α expresses the ratio of the amount of water extracted from the topsoil to that extracted from the full rooting zone. The function β describes maximum water extraction as a function of soil water content and soil characteristics. It is calculated for 50% of the monthly addition, $(P_m - EI_m)/2$, to arrive at the mean monthly water extraction. Its parameter γ depends on topsoil texture and mineralogy, while δ represents the water unavailable to plants, i.e. the intercept of the water extraction curve β. The values of δ and γ (Table 2) are based on Hillel (1980). For clays, $\gamma = 6$, resulting in a strong decrease in water extraction below SWC/SSC about 50%. Due to the selected value of δ for clays, water extraction at SWC/SSC < 10% is reduced. In sands and medium-textured soils, β sharply decreases at values of SWC/SSC of about 40% and 20%, respectively. Because SWC is a function of β, the monthly equilibrium SWC is achieved independently of the initial water content at the start of the simulation.

- *Effects of Soil Drainage (DRNG).* In the calculation of soil-water content above, drainage properties are not considered. Drainage is a soil property that determines the removal of excess water from the soil, and is an indicator of soil aeration. Soil drainage indices are used to estimate soil-water status and are prescribed for soil units on the basis of several soil properties including the presence of impermeable and less permeable horizons (Table 1). The global distribution of the drainage index (DRNG) is shown in Plate 3. We recognize that the soil data base may not represent the local spatial variations in drainage (see appendix), particularly for small areas of hydromorphic soils which may be important for N_2O fluxes.

Very poorly drained soils (DRNG = 5) include those soil groups strongly influenced by groundwater such as the Gleysols and Histosols, as well as soils with permafrost within 200 cm of the soil surface (gelic soil units). Soils with somewhat impeded drainage and moderately restricted aeration (DRNG = 4) include gleyic Solonetz and soils with plinthite; plinthic Acrisols and plinthic Ferralsols are wet tropical soils, usually with fluctuating water tables, and with firm plinthic subsurface layers within 50-125 cm of the surface that are iron-rich and humus-poor.

Gleyic Solonetz are compact soils with hydromorphic properties and clay in the subsoil. The intermediate class (DRNG=3) indicates impeded drainage and restricted aeration; it encompasses gleyic soil units (in which only the subsoil is influenced by groundwater or with a seasonally perched water table within the profile), Planosols (a slowly permeable subsoil), Vertisols (soils with swell/shrinkage properties and montmorillonitic clays dominating the clay fraction) and the nongleyic Solonetz (compact alkaline soils with a natric horizon). About 20% of the land surface, with a drainage index of 2, comprises soils with an increase in clay content in the subsoil (argic horizon), (FAO/Unesco, 1988). Water stagnation and anaerobic conditions may occur in these soils during periods of high precipitation. About two thirds of the land lacks the above properties and is considered freely drained (DRNG=1). The effects of drainage on soil oxygen limitation and water availability are difficult to quantify. The drainage indices are used to estimate soil water status.

- *Soil-Water Status (SWS).* Soil-water status of the topsoil is scaled on the basis of soil-water content and drainage as shown in Table 3. Although it is difficult to combine drainage and soil-water status at their intermediate values in the index, we note several points: (1) distinguishing saturation levels <20% is not important; (2) we increase the SWS scale linearly up to the saturation; (3) we fill in the rest of the table by assuming that N_2O production likely asymptotes at high saturation and poor drainage. While it is clear that the highest SWS rank of 10 should be assigned to a poorly drained soil when the monthly soil-water content approaches storage capacity, the SWS scale is somewhat arbitrary. It represents a first attempt at quantifying our understanding of the effects of soil drainage characteristics on soil oxygen and soil moisture.
- *Effects of water status and oxygen status on decomposition, nitrification and denitrification.* Two factors are derived to describe these effects: water availability (H2O) and oxygen limitation (O2). H2O describes the influence of soil water on decomposition, mineralization and nitrification processes. In general, water contents of 60-80% of field capacity are favorable for these processes while nearly saturated and anaerobic soils have low H2O values (Linn and Doran, 1984; Davidson, 1991). H2O (Table 4) is based on the soil-water status of the previous and current months. Wetting of dry soils is assumed to be more favorable, and drying soils less favorable, than are conditions of constant water content (Groffman and Tiedje, 1988).

Although we do not attempt to simulate the reported pulses in N_2O production after the wetting of dry soils, we hypothesize that pulses of N_2O at the onset of a wet season give a higher average monthly N_2O flux than in wet months preceded by moist conditions.

The regulator O2 expresses effects of soil-oxygen status on denitrification. It is obtained from the soil-water status of the previous and current months (Table 5) and includes the effect of wetting and drying of soils. Oxygen limitation at soil-water contents of 60-80% is treated as most favorable for N_2O production by nitrification while denitrification is more prominent at water contents of >80%. In general, high levels of wetness result in greater oxygen limitation; wetting of dry soils is assigned higher O2 indices than constantly wet or moist soils. The information in H2O and O2 is very similar at SWC values between 7 and 9. Under saturated conditions, low H2O counteracts high O2 values reflecting the simultaneous occurrence of conditions favorable for denitrification (O2), and unfavorable for NH_4^+ oxidation by nitrification. In one sensitivity experiment, only the current month determines H2O and O2; in that case the H2O and O2 scales are the diagonals in Tables 4 and 5, respectively. Annual sums of monthly H2O and O2 are shown in Plate 4.

- *N_2O production (N2O).* The controls on N_2O fluxes identified above are combined to yield monthly nondimensional N_2O indices at 1° resolution for the globe. We recognize that the five factors chosen are not independent. In particular, CARBON, scaled from the annual integral of the NDVI, captures geographic variations in temperature, soil moisture, and soil fertility.

Table 3. Scaled soil-water status (SWS) as a function of soil drainage (DRNG; see Table 1) and modeled soil-water content/soil-water storage capacity (SWC/SSC; see Table 2)

	SWC/SSC, %	Drainage Scale (DRNG)				
		1	2	3	4	5
1	0 - 20	1	1	1	1	2
2	20 - 30	2	2	2	2	3
3	30 - 40	3	3	3	4	5
4	40 - 50	4	4	4	5	7
5	50 - 60	5	5	5	6	8
6	60 - 70	6	6	6	7	9
7	70 - 80	6	7	7	8	9
8	80 - 90	7	7	8	9	10
9	90 -100	7	8	9	10	10
10	surplus (>100%)	8	9	10	10	10

Table 4. Scaled soil water availability (H2O) for the reference case and experiments 1-5 and 7.

	SWS in Current Month									
SWS in Preceding Month	1	2	3	4	5	6	7	8	9	10
1	1	2	4	6	9	10	10	10	6	1
2	1	1	3	5	8	9	10	10	6	1
3	1	1	2	4	7	8	9	10	5	1
4	1	1	2	4	7	8	8	8	4	1
5	1	1	2	4	6	7	7	7	3	1
6	1	1	2	4	6	7	7	6	2	1
7	1	1	2	4	6	7	7	6	2	1
8	1	1	2	4	5	6	6	5	2	1
9	1	1	2	3	4	6	6	4	2	1
10	1	1	2	3	4	5	6	4	2	1

H2O is a function of soil-water status (SWS, see Table 3) of the current month and of the preceding month. The H2O scale corresponding to SWS in the current month only, used in experiment 6, is given by the underlined numbers on the diagonal of the table (see Table 5).

There are many ways to combine the factors. Lacking information about relative importance, we assume that all five controlling factors are of equal importance, i.e. the maximum fertility factor has the same effect as the maximum oxygen limitation factor as far as N_2O production is concerned. Hence, although FERT is scaled from 1 to 5 because of our inability to discriminate further, FERT is multiplied by two (FERT*) to normalize to the other factors.

Table 5. Scaled oxygen limitation (O2) for the reference case and experiments 1-5 and 7.

	SWS in Current Month									
SWS in Preceding Month	1	2	3	4	5	6	7	8	9	10
1	1	1	1	4	6	8	10	10	10	10
2	1	1	1	4	6	8	10	10	10	10
3	1	1	1	3	5	7	9	10	10	10
4	1	1	1	3	4	6	8	9	10	10
5	1	1	1	2	3	5	7	8	9	10
6	1	1	1	1	2	4	6	7	8	9
7	1	1	1	1	2	3	5	6	7	8
8	1	1	1	1	2	3	4	5	6	7
9	1	1	1	1	2	3	4	5	6	7
10	1	1	1	1	2	3	4	5	6	7

O2 is a function of the soil-water status (SWS, see Table 3) of the current month and of the preceding month. The O2 scale used in experiment 6, which is a function of SWS in the current month only, is given by the underlined numbers on the diagonal of the table.

We model the nondimensional monthly N_2O production as the geometric mean of the five controlling factors. In this way, a low value for one of the factors automatically lowers the N_2O production index. For example, values of 1 and 9 for two factors are given less weight than 5 and 5, which yields the same arithmetic mean.

It is difficult to rank the above regulators a priori since the influences of these regulators varies among habitats (Tiedje, 1988). Therefore the monthly nondimensional N_2O production index, N2O is calculated for the reference case as:

$$\mathrm{N2O} = (\mathrm{O2} \times \mathrm{H2O} \times \mathrm{SOD} \times \mathrm{FERT^*} \times \mathrm{CARBON})^{1/5} \quad (3)$$

where N2O, O2, H2O, and SOD are indices calculated monthly, and FERT* and CARBON are site characteristics constant for every month. Under conditions where soil processes are inactive (months in which mean surface temperatures $< 0°C$), N2O is set to zero. N2O is also set to zero for months in which the monthly precipitation ≤ 5.0 mm and the soil-water content is $\leq 1\%$. When there is rainfall ($P > 0$) and the water budget equation predicts a dry soil at the end of that month ($SWC = 0$), the soil has not been dry throughout the month. In that case the indices for O2 and H2O are low, giving low N2O.

2.4. RESULTS AND DISCUSSION

The model was run globally at 1° resolution for 12 months. Model results for the reference case are discussed in section 4.1; sensitivity experiments analyzing the relative importance of the five factors are discussed in 4.2, and results for selected sites are described in section 4.3. Results are compared with measurements in section 4.4 and used to estimate a global flux in section 4.5.

2.4.1. *Global Results for the Reference Case*

The mean annual sum of N2O and its standard deviation (expressed as percentage of the mean) is presented for the reference case and the experiments for the 23 soil groups (Table 6). The standard deviation reflects the variation in annual sums over the areal extent of each soil group. Note that the SOD1 function is used in all cases. The results for the soil groups are presented separately for the tropics (Table 6a; within 30° of the equator) and the extratropics (Table 6b; poleward of 30° latitude). The annual sum of monthly N_2O indices for the reference case is shown in Plate 5; the latitudinal and seasonal distribution of N2O is shown in Figure 3. Since the index can range from 0 to 10 in any month, the potential annual maximum is 120, although the actual maximum is 84. The results for the reference case suggest that the wet tropics and subtropics have the highest levels of N_2O production. Mediterranean regions have intermediate values and soils in cold temperate and boreal regions, as well as deserts, have the lowest potential for N_2O production.

The most important soil group, in terms of areal extent and nitrous oxide production, is the Ferralsols (Table 1, group 13; Table 6a, group 13A). Because of their occurrence in somewhat drier climates, Acrisols (group 11) show slightly lower N2O than do Ferralsols, while the fertile Nitosols (group 12) have higher values than Ferralsols. The extensive subtropical and tropical Luvisols (Table 6a, group 5A) also exhibit high N_2O potential. Other soils with high N2O levels, but occupying minor areas, are tropical Podzols (group 2A), Vertisols (group 16), Andosols (group 17), Fluvisols (group 19) and Gleysols (group 20). The subtropical Phaeozems and Kastanozems (groups 7A and 9A, respectively), occurring mainly in South America, exhibit high N_2O indices but their global effect is moderated by their small areas. Their temperate counterparts (Table 6b, groups 7B and 9B) show much lower N2O levels. Another soil group of significant area is the shallow Leptosols (group 21). The model predicts that temperate Leptosols, when covered by their natural forest vegetation, have very low N_2O production, while tropical Leptosols are intermediate.

About 15% of the Earth's ice-free land area is covered by desert soils (group 14) which have low potential for N_2O production mainly due to precipitation limitation. Tropical organic soils (group 23) show much higher N2O values than their temperate counterparts.

Generally where N2O is low, percent standard deviations are high, although the absolute standard deviation may be low. The low annual N2O values of temperate areas have higher percent standard deviations than do the tropics even when occupied by the same soil. Highest standard deviations are found in both tropical and temperate extents of groups 14 (desert soils), 15 (saline and alkaline soils), 18 (sand soils), 21 (shallow soils), and 22 (weakly developed soils), (Table 1, Plate 1). Percent standard deviations are small for the temperate and tropical distributions of Vertisols (group 16).

Temporal variability (not shown in Table 6) is expectedly high for temperate soil groups because of effects from seasonal temperature and moisture regimes. Very high intra-annual variabilities are also found for soils in semiarid and arid climates (soil groups 14-15) in both the tropics and temperate zones presumably due to precipitation fluctuations. N_2O indices vary seasonally for soils conditioned by parent material (groups 16-18) and for soils conditioned by their physiographic position (groups 19-22) but this variability is probably induced by the occurrence of these soils over a broad climatic range. Tropical and subtropical soils of wet climates (groups 11-13), show muted variations in N2O during the year.

Figure 3 shows the seasonality of N_2O potential modeled in this study. Expectedly, the seasonal variation is greatest for temperate climates of the northern hemisphere; the seasonal variation in temperate climates of the southern hemisphere is about half that in the north. Tropical soils are active nitrous oxide sources throughout most of the year. Moreover, monthly N2O values are higher for most of the tropics than for temperate climates; annual N2O sums can be a factor 3-4

Table 6a. Mean and standard deviation[†] of modeled N2O by soil group in the tropics and subtropics for the reference case and seven sensitivity experiments

Soil Group	Area 10^6ha	REF	E1	E2	E3	E4	E5	E6	E7
2A	15	50	48	49	52	52	61	50	60
		(19)	(21)	(21)	(25)	(22)	(20)	(20)	(33)
3A	95	38	35	33	39	41	45	38	69
		(28)	(34)	(29)	(30)	(28)	(29)	(28)	(34)
5A	546	44	41	43	38	49	44	43	54
		(26)	(33)	(25)	(32)	(25)	(26)	(27)	(43)
7A	15	53	56	51	45	56	48	53	73
		(21)	(30)	(19)	(28)	(17)	(28)	(22)	(33)
9A	75	39	38	36	31	48	31	38	60
		(35)	(43)	(36)	(44)	(30)	(38)	(36)	(42)
10A	290	53	53	53	49	58	50	53	61
		(25)	(29)	(25)	(30)	(22)	(28)	(26)	(37)
11A	736	52	52	50	50	54	56	52	74
		(15)	(18)	(16)	(18)	(17)	(17)	(15)	(25)
12A	204	58	56	57	52	64	52	57	66
		(18)	(23)	(16)	(22)	(16)	(17)	(19)	(29)
13A	1113	56	52	51	60	59	58	56	82
		(16)	(16)	(16)	(16)	(15)	(13)	(17)	(18)
14A	1008	12	10	12	10	14	14	12	11
		(87)	(105)	(86)	(93)	(90)	(80)	(86)	(134)
15A	84	24	19	25	25	25	29	24	26
		(41)	(59)	(35)	(46)	(39)	(53)	(41)	(87)
16A	281	39	36	42	35	41	36	37	35
		(19)	(21)	(16)	(22)	(18)	(28)	(19)	(52)
17A	49	53	54	53	46	59	48	53	59
		(29)	(31)	(30)	(36)	(27)	(33)	(29)	(29)
18A	696	25	22	24	27	28	31	25	34
		(50)	(60)	(45)	(60)	(46)	(47)	(50)	(77)
19A	132	48	47	49	43	52	45	47	51
		(39)	(44)	(38)	(44)	(37)	(43)	(40)	(49)
20A	162	45	42	44	43	45	55	45	64
		(21)	(22)	(20)	(24)	(21)	(22)	(21)	(38)
21A	752	23	21	21	21	24	26	22	27
		(78)	(89)	(76)	(86)	(78)	(74)	(78)	(106)
22A	330	18	15	19	17	21	22	18	16
		(65)	(75)	(58)	(71)	(62)	(59)	(64)	(107)
23A	39	43	38	39	45	38	66	43	64
		(17)	(16)	(17)	(17)	(21)	(16)	(17)	(33)

† Standard deviations are in parentheses, and are expressed as a percent of the mean N_2O index of each soil group for each experiment.

higher in the tropics than in temperate zones. These broad-scale patterns from a simple model agree with variations in decomposition rates and amounts of carbon and nitrogen cycling through soils in tropical and temperate climates (Jenkinson and Ayanaba, 1977; Vitousek, 1984).

Table 6b. Like Table 6a, but for temperate and subpolar latitudes

Soil Group	Area 10^6ha	REF	E1	E2	E3	E4	E5	E6	E7
1B	674	3	9	3	2	3	3	3	7
		(106)	(42)	(106)	(117)	(102)	(99)	(106)	(61)
2B	478	10	25	9	9	10	12	10	22
		(64)	(45)	(72)	(79)	(62)	(62)	(62)	(60)
3B	46	29	36	26	30	29	35	29	57
		(29)	(24)	(26)	(33)	(26)	(31)	(29)	(36)
4B	251	10	21	10	9	11	10	11	21
		(32)	(28)	(33)	(36)	(33)	(30)	(33)	(33)
5B	404	22	37	21	19	24	20	23	40
		(48)	(35)	(47)	(49)	(45)	(44)	(45)	(45)
6B	27	11	22	11	10	13	11	12	23
		(24)	(18)	(23)	(23)	(19)	(22)	(25)	(20)
7B	138	31	48	30	25	34	27	32	52
		(48)	(33)	(44)	(53)	(42)	(48)	(46)	(43)
8B	225	15	25	16	11	17	13	15	21
		(23)	(29)	(22)	(24)	(20)	(23)	(24)	(30)
9B	417	15	25	17	11	18	14	16	21
		(43)	(46)	(35)	(47)	(42)	(35)	(42)	(69)
10B	468	21	38	21	18	23	19	22	36
		(63)	(52)	(62)	(69)	(62)	(58)	(62)	(62)
11B	166	36	54	32	34	37	36	36	72
		(27)	(13)	(29)	(30)	(26)	(32)	(27)	(20)
12B	5	52	66	45	47	53	44	52	79
		(23)	(9)	(26)	(26)	(26)	(29)	(22)	(23)
13B	3	49	56	43	48	48	46	50	76
		(12)	(10)	(14)	(16)	(8)	(15)	(11)	(18)
14B	868	15	19	16	12	16	16	15	15
		(46)	(54)	(48)	(57)	(49)	(43)	(45)	(75)
15B	148	16	19	17	16	16	16	16	19
		(58)	(56)	(52)	(64)	(52)	(71)	(58)	(102)
16B	33	38	45	37	34	37	37	37	54
		(22)	(16)	(18)	(26)	(18)	(30)	(23)	(45)
17B	62	23	50	23	20	24	20	24	43
		(65)	(34)	(62)	(70)	(63)	(67)	(65)	(62)
18B	25	18	25	18	17	19	21	19	25
		(55)	(39)	(51)	(63)	(57)	(48)	(51)	(74)
19B	114	16	25	16	12	17	14	15	21
		(82)	(67)	(79)	(93)	(79)	(73)	(80)	(86)
20B	250	14	24	13	11	13	14	14	24
		(77)	(58)	(83)	(88)	(72)	(85)	(76)	(77)
21B	1521	9	18	8	7	9	11	9	16
		(86)	(67)	(92)	(101)	(81)	(78)	(86)	(83)
22B	117	18	27	18	15	20	18	18	24
		(50)	(40)	(50)	(58)	(48)	(45)	(48)	(58)
23B	133	7	15	7	7	7	9	7	17
		(58)	(42)	(61)	(60)	(51)	(56)	(58)	(48)

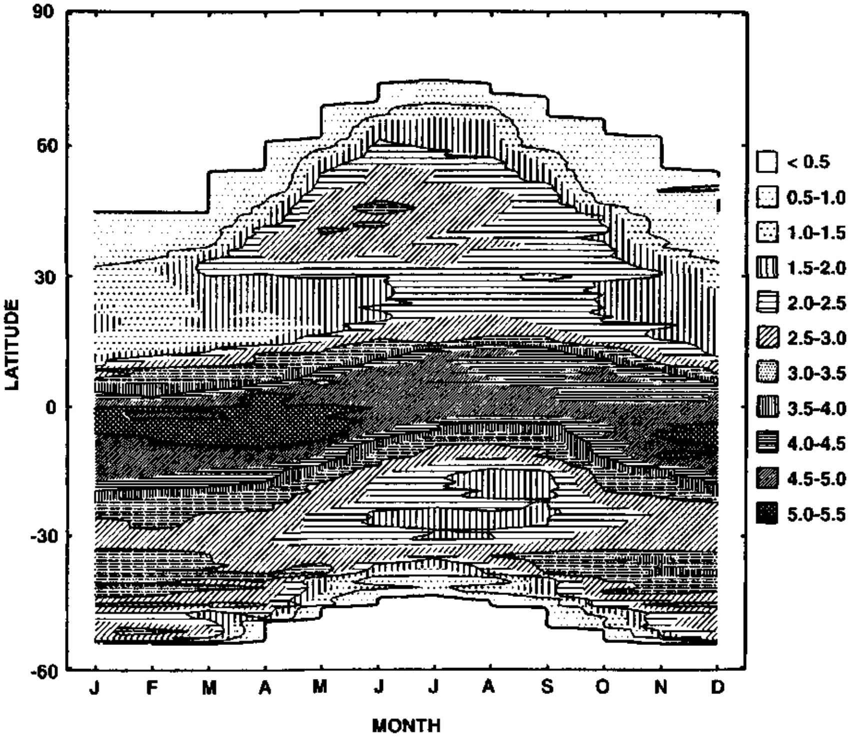

Figure 3. Seasonal and latitudinal distribution of the N_2O production potential (N2O). The zonal means are calculated over land areas.

2.4.2. *Sensitivity Experiments*

Characteristics of the sensitivity experiments are briefly described in Table 7. Annual N_2O production potential for the sensitivity experiments are shown in Table 6.

- *Experiment 1 (- SOD).* In temperate climates, this experiment yields much higher values for annual N2O than those of the reference case by removing temperature limitations in autumn, winter and spring (Table 6b). In the tropics, the experiment gives somewhat lower N2O values (e.g., Table 6a, soil groups 12A and 13A) than the reference case. The role of SOD is also expressed by differences in the variability between the reference case and experiment 1. The spatial variability as expressed by the percent standard deviation decreases considerably for most extratropical soils when SOD is excluded (except for the semiarid Kastanozems and desert soils - Table 6b, groups 9B and 14B, respectively). For most tropical soil groups, the percent standard deviation is higher in experiment 1 than in the reference case.
- *Experiment 2 (- CARBON).* For nearly all tropical and temperate soils, this experiment yields N2O values lower than those of the reference case. Results differ from the reference case most markedly in the wet tropics and subtropics, particularly for the Ferralsols (group 13). Over the distribution of these soils, exclusion of CARBON causes an average decline of 5 in the annual mean N_2O potential. In this experiment higher N2O values are calculated for temperate soil groups 8B and 9B (steppe soils), 14B and 15B (desert soils) and for some minor tropical soil groups (15A, 16A, and 22A). The effect of excluding CARBON is minor in semiarid, arid, and steppe areas because other factors limit N_2O production. In the desert soils (group 14) and temperate

steppe soils, N2O is low in both the reference case and experiment 2 due to moisture limitations.

The results of experiments 1 and 2 indicate that SOD and CARBON are driving N_2O production in wet and moist tropics. Input of organic matter plays a less important role in temperate climates, because the annual N2O is primarily determined by low temperatures in winter; and in dry climates, where soil moisture is the major regulator. These observations agree with the suggestion that carbon nowhere really prevents denitrification (Tiedje, 1988).

Excluding CARBON decreases the spatial variability for most temperate and tropical soil groups conditioned by their parent material (soil groups 15-18), but has no major effect on the variability in tropical soils in wet climates (Table 6a, groups 11A-13A). Exclusion of CARBON decreases the variability over tropical shallow soils (group 21A), whereas the variability increases for temperate zone distributions of these soils (group 21B). In the major climate-conditioned temperate soils (groups 3B-9B) exclusion of the organic input lowers spatial variability.

- *Experiment 3 (- FERT).* Exclusion of soil fertility strongly reduces the annual N2O for most highly fertile soils in temperate and tropical zones, for example, the Phaeozems, Chernozems, Kastanozems, and Nitosols (groups 7, 8, 9 and 12, respectively) and increases N2O for the infertile tropical soils (group 2A, 3A, 13A, 15A, 18A, and 23A in Table 6a). For other temperate and tropical soils with intermediate soil fertility, the effect of experiment 3 is minor. Excluding soil fertility increases the standard deviation for most temperate and tropical soil groups due to the simplicity of the fertility scale in which the same index is associated with most soil units within each group. However, within landscapes, soil fertility may be a major cause of variability in N_2O fluxes (Table 8; Livingston et al, 1988; Matson et al. 1990).

- *Experiment 4 (- O2).* The O2 factor provides information not included in the other four factors over the whole range of temperate and tropical ecosystems. Under wet conditions, high O2 raises the levels of expected denitrification. Similar conditions produce low H2O values simulating blockage of nitrification. Hence, because of the partial overlap of the scales of O2 and H2O, important effects on overall levels of N2O in this experiment are found only in well-drained soils. No effect, or a decrease relative to the reference case, is found for the poorly-drained Gleysols and Histosols (soil groups 20 and 23, respectively). Where aerobic conditions prevail, O2 depresses the N_2O index (Table 5). In most tropical and temperate soils, annual N2O calculated in exp. 4 is higher and variability is lower than in the reference case, consistent with Tiedje (1988), who concluded that in aerobic systems, oxygen is the principal factor limiting denitrification.

- *Experiment 5 (- soil variability - CARBON).* In experiment 5, CARBON is excluded and for all soils FERT = 3, DRNG = 1 (well drained) and texture is fine (high soil-water storage capacity). This eliminates variability produced by soil and vegetation differences and isolates the spatial variability caused by climate. The results can best be compared with experiment 2 (- CARBON). Smoothing soil differences increases spatial standard deviations for about half of the temperate soil types (Table 6b, groups 3B, 7B, 8B, 11-13B, 15-17B, 20B) and for most tropical soil groups (Table 6a). Tropical soils showing lower variability in this experiment are groups 12A-14A and 21A-23A. What this means can best be illustrated for the Ferralsols (group 13A), the major tropical soil group. These soils occur in relatively uniform climates so soil differences contribute to the variability. The high spatial variability within soil groups 15A-B to 22A-B, soils conditioned by factors other than climate, reflects the distribution of these soils over different climatic zones. As in the reference case and experiment 1, the percent standard deviation among temperate soil groups is higher than in most of the equivalent tropical soils.

- *Experiment 6 (not considering soil water and oxygen of the previous month).* Effects of wetting and drying are excluded in this experiment; H2O and O2 indices are given by the diagonals in Tables 4 and 5, respectively. The overall effect of considering water status only of the current month is minor and effects do not show patterns specific to soil groups or clusters.

Table 7. List of Experiments to Evaluate the Sensitivity of the N_2O Production Index to Different Controls.

Experiment	Description
Reference case:	All 5 factors are used; SOD1 is from Mosier and Parton (1985). $N2O = \sum_{1\,y} (O2)^{1/5} \times (H2O)^{1/5} \times (SOD)^{1/5} \times (FERT^*)^{1/5} \times (CARBON)^{1/5}$
Experiment 1 (- SOD)	As reference case, but SOD is excluded. $N2O = \sum_{1\,y} (O2)^{1/4} \times (H2O)^{1/4} \times (FERT^*)^{1/4} \times (CARBON)^{1/4}$
Experiment 2 (- CARBON)	As reference case, but CARBON is excluded. $N2O = \sum_{1\,y} (O2)^{1/4} \times (H2O)^{1/4} \times (SOD)^{1/4} \times (FERT^*)^{1/4}$
Experiment 3 (- FERT)	As reference case, but FERT* is excluded. $N2O = \sum_{1\,y} (O2)^{1/4} \times (H2O)^{1/4} \times (SOD)^{1/4} \times (CARBON)^{1/4}$
Experiment 4 (- O2)	As reference case, but O2 is excluded. $N2O = \sum_{1\,y} (H2O)^{1/4} \times (SOD)^{1/4} \times (FERT^*)^{1/4} \times (CARBON)^{1/4}$
Experiment 5 (- soil variability)	As reference case, but CARBON is excluded and the soil parameters are equal for all soils: FERT = 3, DRNG =1, and texture is fine. This experiment studies the effect of climate on the variability of N2O. $N2O = \sum_{1\,y} (O2)^{1/4} \times (H2O)^{1/4} \times (SOD)^{1/4} \times (FERT^*)^{1/4}$
Experiment 6 (H2O and O2 of current month only)	As reference case, but soil-water and oxygen status in the preceding month is not considered. H2O and O2 are the diagonals of Table 4 and 5, respectively.
Experiment 7 (only CARBON)	CARBON is used to predict N2O. $N2O = \sum_{1\,y} (CARBON)$

FERT* = 2 × FERT, the fertility index from Table 1. See text for explanation.

However, seasonal N2O patterns exhibit a response. Results for selected sites (section 4.3) illustrate cases in which monthly N_2O indices are equal to those of the reference case (e.g., the Brazilian site). However, in sites with marked seasonality of rainfall, experiment 6 exhibits lower N2O values at the onset of the wet season and higher values at the end of the growing period. The net result is no, or only minor, differences in the annual sum. For most soil groups conditioned by climate, spatial variability declines slightly in temperate regions (Table 6b, groups 2B, 5B, 7B, 9B, 10B, 12B-14B) and increases in the tropics (Table 6a).

- *Experiment 7 (only CARBON)*. Using CARBON as the sole predictor of N_2O production generally yields higher values than the reference case. Exceptions are tropical desert soils, Vertisols and Regosols (groups 14A, 16A, and 22A, respectively). N2O increases are largest in temperate soils and in soils of the wet tropics. Standard deviations in experiment 7 are generally elevated for the tropical soils. Among temperate soil groups, variability is both higher and lower than in the reference case suggesting no clear relation with climate. The CARBON factor represents organic matter inputs and not N inputs. In addition, CARBON includes much but not all information needed to characterize conditions responsible for N_2O production. In particular, soil aeration (drainage and soil wetness), soil-water availability, and possibly soil fertility, are probably not captured by the NDVI. Another problem of the underlying NDVI data is that NPP is overestimated in a number of ecosystems, particularly needleleaved forests and vegetation types with a strong seasonal pattern in growth (Box et al., 1989). The broad spatial patterns and overall levels of CARBON are, however, very similar to the distribution of N2O values from the reference case.

2.4.3. *Results for Selected Sites*

We selected four sites to discuss more fully. They are distributed throughout a range of climatic and ecological regimes and include locations representative of globally important soil groups, as well as regimes dominated by various factors in the model, for example, water balance, temperature, soil fertility.

One site represents a semiarid temperate climate, the second a humid temperate site, and two tropical sites include a seasonal climate and a per-humid climate. In the following discussions, the three functions describing soil organic matter decomposition (SOD) discussed above are referred to in abbreviated form as follows: SOD1, Mosier (from Mosier and Parton, 1985; Parton et al., 1988); SOD2, Parton (from Parton et al., 1987); SOD3, Fung (from Fung et al., 1987). The base or reference case is the Mosier function, SOD1 (Figure 2).

- *Colorado, Temperate Arid North America.* The first site a semiarid grassland region in Colorado in the western US (Figure 4a) occupied by shortgrass prairie with no woody cover. The rescaled NDVI sum representing carbon supply is only 3.4 which lies in the low end of the scale for this index, consistent with the low biomass semiarid ecosystem. The underlying soil, a luvic Kastanozem, is a highly fertile soil conditioned by steppe climates (Table 1) and occurs here under conditions of about 350-mm annual precipitation which is exceeded by potential evapotranspiration (about 630 mm per year). The continental location results in mean monthly temperatures that vary within a few degrees of 0°C for five months of the year and rise to the low twenties in July and August, increasing the potential production of N_2O. However, in this summer period, moisture deficits control the ecosystem due to consistently low precipitation. Soil-water deficits are extreme for the entire second half of the year, resulting in reduced O2 and H2O (denitrification and nitrification, respectively) factors for most of the year. Organic matter decomposition (SOD) is moderate at this site although the three SOD functions vary considerably. The lowest is from the Mosier curve (SOD1 = 16) followed by 25 and 33 from SOD3 and SOD2, respectively.

The annual N_2O production for the base case is low, depressed by several factors that act serially during the year. N_2O production shuts down from November through April due to low temperatures; warm summer temperatures, dovetailing with moderate rainfall, create dry well-aerated soils. As a result, the seasonal pattern of N2O is governed by elevated O2 (denitrification) and H2O (nitrification) indices in conjunction with low rates of decomposition in the spring, followed by trend reversals in these three factors in the summer.

Excluding the fertility factor (experiment 3) lowers N2O from 17 to 12, an expected response since the luvic Kastanozems here are associated with the maximum fertility index. Experiment 5 results in a similar decline in N2O for similar reasons: the annually constant fertility parameter is reset to a middle value and the effect of organic matter availability is removed. As an attempt to focus on the climate effect, experiment 5 reflects observations indicating that seasonal and relatively arid temperate climates carry inherently low potential for N_2O production. Experiments 2 and 4 exhibit moderate divergence from the base case with slight increases in the annual N2O value but little seasonal redistribution indicating that the factors evaluated in these experiments (carbon availability, oxygen limitation and soil-water history) exert approximately equal control over N2O modeled at this site.

The highest N2O value is for experiment 1 since seasonal temperature limitations on the decomposition of soil organic matter are relaxed in this experiment. In the reference case, monthly N2O values are zero from November through April, and range between 4 and 2 during the productive season of May to October; in experiment 1, N_2O production rises as high as 6 and remains elevated for a long season lasting from February to November. Overall, N_2O production in this semiarid temperate environment appears to be limited by low water availability and low organic matter input.

Overall N_2O production in this semiarid temperate environment appears limited by high aerobicity, low water availability and low carbon supply which are not sufficiently counteracted by the high summer temperatures.

- *Germany, Temperate Western Europe.* The site in western Germany is in a region of temperate deciduous forest admixed with evergreens (Figure 4b). CARBON is intermediate at 4.4; the underlying soil belongs to the dystric Cambisol group, moderately fertile soils with weak profile development, free drainage and good aeration (Table 1). The site is located in a cool wet climate characterized by a surplus of precipitation over potential evapotranspiration (820 mm and 594 mm per annum, respectively).

Organic matter decomposition at this site varies by a factor of four among the three functions; the lowest was for SOD1 (8), followed by SOD2 and SOD3 (30). SOD1 predicts low decomposition rates during a short season from May to September while SOD2 gives higher, slightly peaked, values in summer when temperatures are in the 10°-20°C range, where largest differences are expected. For the SOD3 relationship, temperature exerts weaker control on the level and duration of decomposition, resulting in relatively low and constant decomposition throughout the year. Annual N2O estimated via the three relationships exhibits patterns similar in seasonality to their respective SOD factors and to the overall trend in temperature and H2O; they range from the base case of 20 (SOD1) to 33 (SOD2) and 51 (SOD3). Although the predicted N2O level is similar among the three relationships for months when they all indicate N_2O activity, the productive seasons are longer by several months with the SOD2 and SOD3 relationships.

The sensitivity experiments generally produced minimal variations on the seasonal and total values of N2O indicating that most parameters play equal roles in annual N2O estimates at this site, as well as reflecting the temporally constant status of water and oxygen parameters. The only major effect, increasing N2O to about 2.5 times that of the base case, is in experiment 1 which virtually eliminates the seasonal low-temperature limitation on decomposition and N mineralization; N2O levels for individual months in experiment 1 are somewhat higher than those for the reference case and rise above zero for a long season extending from February through December in contrast to the five-month productive season for the base case. Since the system is well supplied with moisture throughout the year, soil-water history is not a crucial factor in the production of N_2O (experiment 6). The results of the reference case and sensitivity experiments indicate that N_2O production in fertile, humid forest zones in moderate climates is controlled predominantly by temperature. Moisture-related parameters exert little limitation except that the relative constancy of moist soil conditions throughout the year, while contributing to conditions amenable to N_2O production, does not produce the high N2O pulses that accompany the wetting of dry soils.

- *Venezuela, Tropical Seasonal South America.* The South American site is occupied by drought-deciduous forest underlain by orthic Acrisols with mildly impeded drainage characteristic of wet tropical and subtropical climates with seasonal rainfall (Figure 4c). Almost half of the ~900-mm annual precipitation falls from June to August; the drier winter months (January-March) have ≤ 20 mm rainfall monthly. Constant high temperatures produce small variations in potential evapotranspiration, which ranges from 100 mm in February to 143 mm in May. This relatively flat trend in PE, combined with highly seasonal precipitation, produces conditions of moisture deficit from September to May; the season of small moisture surplus is confined to June/July and precipitation following the winter aridity produces a seasonal spike in nitrification (H2O). As expected, the temperature-dependent decomposition functions are very similar and essentially constant through the year. Other site characteristics (fertility and drainage) are intermediate but the organic matter supply is elevated reflecting the higher deciduous biomass of the vegetation.

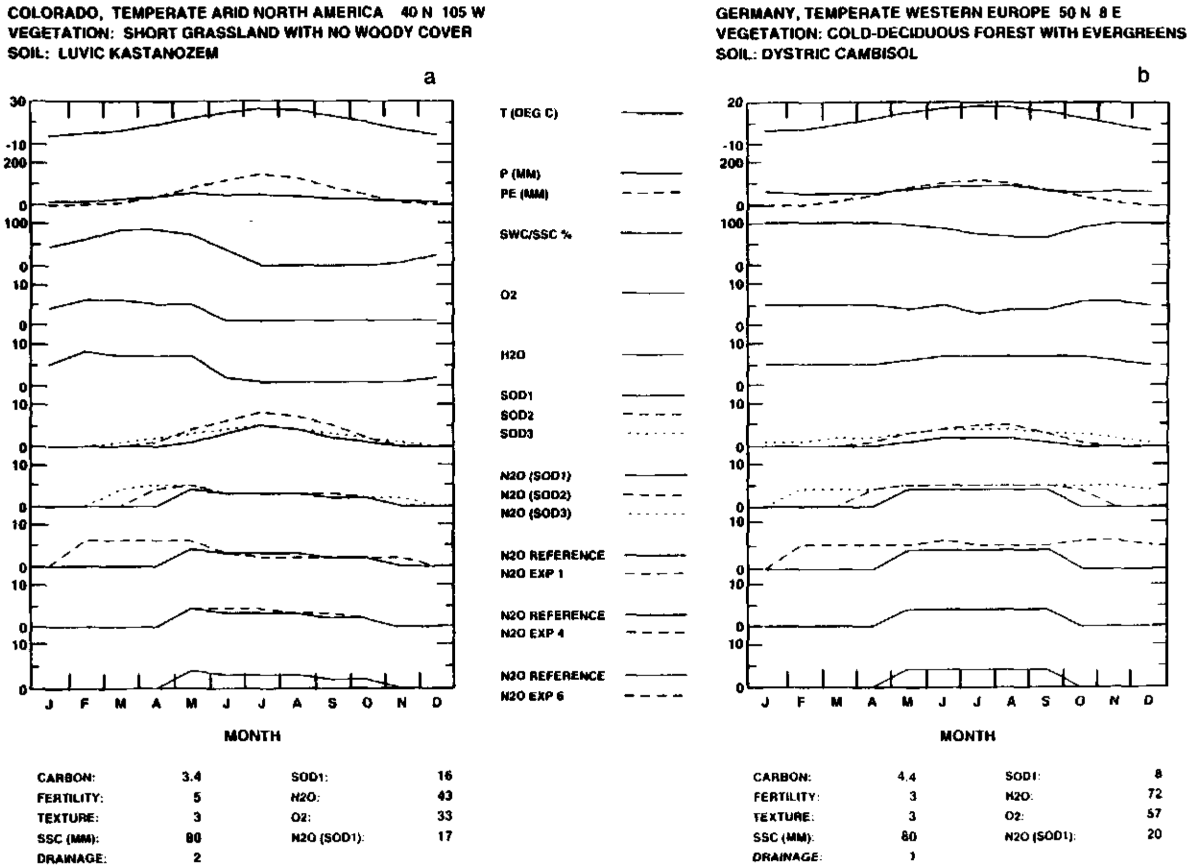

Figure 4. Environmental characteristics, control indices, and N_2O production indices at four sites: (a) Colorado, temperate arid North America, (b) Germany, temperate Western Europe, (c) Venezuela, tropical seasonal South America, and (d) Brazil, tropical humid South America. The three modeled N_2O indices are derived from the three decomposition functions shown in Figure 2; SOD1 is used in the reference case.

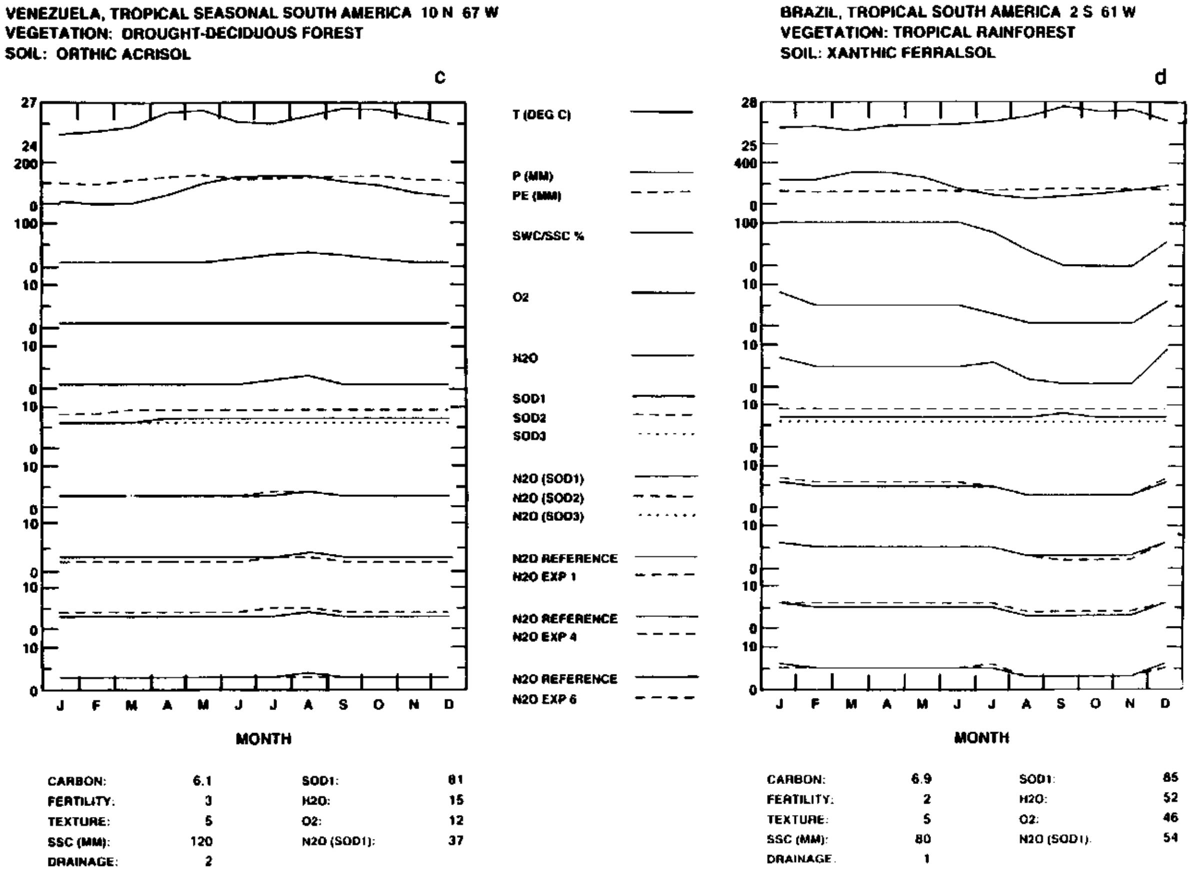

Figure 4. (Continued).

The reference N2O value is less than one half the global maximum; the alternative SOD formulations are essentially equal to the reference case, i.e., highly seasonal values mimic the H2O curve. This seasonal tropical forest site exhibits only moderate responses to the series of sensitivity experiments because of the generally neutral values of site characteristics. Although temperature is not a major controller of N_2O production in terms of seasonality, constant high temperatures throughout the year result in rapid decomposition so that the exclusion of SOD (experiment 1) produces a decline of ~30% in modeled N2O. For reasons mentioned above, removal of parameters seasonally constant for the site (CARBON, FERT and a combination of CARBON and soil factors) all depress the N2O values slightly. The exclusion of the O2 denitrification parameter (experiment 4), which mildly limits overall N_2O production here, produces the only increase in N_2O potential among the sensitivity experiments.

Under conditions of sufficient organic matter, efficiently delivered via fertile soils, the major features controlling the monthly and annual production of nitrous oxide in the tropical seasonal forest appear to be moisture conditions, reflected here in O2 (denitrification) and H2O (nitrification) parameters.

- *Brazil, Tropical South America.* The Brazilian site is situated near Manaus (Figure 4d). Low fertility xanthic ferralsols are under tropical rain forest, an association common in the humid tropics. Organic matter input is high, indicated by the CARBON factor close to 7. The narrow range of annual temperatures is mirrored in the three equally constant functions describing organic matter decomposition. The reference SOD function of Mosier is intermediate at 85, while SOD2 is about 25% higher and SOD3 about 15% lower. Total precipitation is very high at about 2200 mm/year, exceeding the PE of 1675 mm/year. The constancy in temperature produces comparatively constant PE in the range of about 120-150 mm/month; although there are no very arid months, precipitation peaks at 225-300 mm/month from January to May and declines slowly to a low of 64 mm in August. Because the water storage capacity is relatively high and some precipitation continues during the dry season, the soil remains moist much of the year. More aerobic conditions in the soil are reflected in lower values of the O2 (denitrification) and H2O (nitrification) parameters for September through November. Increased soil aeration precedes declining anaerobicity by a month. Annual N2O values from the three temperature functions are very similar; Mosier (SOD1) and Fung (SOD3) are both 54 while Parton (SOD2) is higher at 61, consistent with differences expected in this temperature range (Figure 2).

Experiments 1, 2 and 6 result in N2O values slightly below the reference case. Excluding the factors accounting for organic matter input (CARBON) and decomposition (SOD), removes features that contribute to high N_2O production. Experiment 6, which evaluates the effect of the preceding month's soil water on N2O, shows slightly lower values only for the first half of the year. The remaining experiments all increase N2O estimates in this tropical site, by larger values than the declines of the other experiments: removal of the (low) fertility factor increases the annual total by about 12%; the same increase is produced in experiment 5 in which the low fertility of 2 is prescribed at 3 and the CARBON factor is excluded. The largest response, an increase of ~20%, comes from experiment 4 in which the role of denitrification was excluded. Although this location has considerable organic matter turnover, it has low fertility well-drained soils characteristic of tropical forest regions. Furthermore, the high annual precipitation is sufficiently seasonal to reduce considerably N_2O production capability for about half the year.

2.4.4 *Comparison With Field Measurements*

Modeled monthly N2O is compared with measured N_2O fluxes in various natural ecosystems (Table 8). This comparison allows a general evaluation of the model and data bases in a variety of environments, and provides a framework for translating nondimensional N2O values into N_2O

fluxes. Several observations are excluded from the comparison because site descriptions are incomplete, or the climate, soil conditions or vegetation types from the 1° data bases corresponding to measurement locations differ from those reported. In some cases, the mean of N2O values of a surrounding pair of cells with conditions essentially identical to the measurement site is compared to the measured flux.

The reference case using the SOD1 function (Figure 5) yields the best fit to the measured fluxes (r^2 for least squares fit to a quadratic function = 0.55). SOD2 and SOD3 (not shown) result in r^2 of 0.38 and 0.24, respectively. The r^2 for quadratic fits are 0.16, 0.48, 0.55, 0.43, 0.42, 0.60, and 0.33 for experiments 1-7, respectively.

Relative to the reference case, exclusion of the temperature effect (experiment 1) causes a large reduction in r^2 from 0.55 to 0.16, a dramatic increase in N2O in temperate sites (particularly in temperate climates during autumn, winter and spring), and a moderate decline in N2O values for tropical sites. In temperate summers with sufficient soil moisture, however, temperatures may be comparable with or even higher than those in the tropics. Hence, the effect of excluding SOD in summer is less important than in temperate winters.

As shown in section 4.2, the factor CARBON provides additional variability within climatic and soil regions. However, for the measurement sites considered, the exclusion of CARBON (experiment 2) reduces r^2 from 0.55 in the reference case to 0.48. Using only CARBON to predict N_2O production (experiment 7) gives r^2 of 0.33 and a large increase in N2O particularly in temperate ecosystems.

Although soil fertility may be the principal cause of variation in N_2O fluxes regionally, exclusion of soil-fertility variations globally (experiment 3) has no major effect on the comparison except at temperate sites with high soil fertility (e.g., sites 1 and 2 in Figure 5). Experiment 3 suggests that the soil fertility scale used here does not resolve sufficiently the fertility variations found in natural conditions.

Excluding the oxygen limitation of soils (experiment 4) results in a value for r^2 of 0.43. Globally uniform soil texture, drainage and fertility (experiment 5) reduces r^2 from 0.55 to 0.42, suggesting that variations in these controls capture important information on N_2O flux variations on a global scale. Exclusion of the factor O2 (experiment 4) overpredicts N_2O production in well drained soils. Experiment 6 considers the status of soil water and oxygen of the current month only, and results in an r^2 of 0.60. The change is mainly caused by a decrease in N2O in December for one site near Manaus, Brazil (Table 8, site 12) (Keller et al., 1986). This and minor shifts for some other points causes a slightly improved correlation. The r^2 for the reference case and experiment 6 are similar, but since the major difference is a shift of only one point, the reference case was maintained.

2.4.5 *Estimate of the Global Preagricultural N_2O Emission*

A regression relationship is obtained between measured N_2O fluxes and modeled (reference case) N_2O production indices (N2O). This relationship is applied to the global distributions of monthly N_2O indices to estimate monthly fluxes. The global emission thus calculated is 6.8 Tg N_2O-N y^{-1}. The tropics (± 30° of the equator) contribute 5.4 Tg N_2O-N y^{-1} and the emission from extra-tropical regions (poleward of 30°) is 1.4 Tg N_2O-N y^{-1}.

The role of the world's wetlands in the nitrous oxide budget is uncertain. For tropical and temperate wetlands, partially represented by organic soils (group 23), the model may overestimate the potential for N_2O production. In these wetlands anaerobicity resulting from high water tables may block nitrification; this may limit denitrification rates. Furthermore, under very wet conditions, nitrous oxide can be consumed to serve as an electron acceptor. Therefore, wetlands could act as a sink for N_2O during part(s) of the year. One problem is that many wetlands mapped by Matthews and Fung (1987), ~525 × 10^6 ha globally, do not coincide with the organic soils

Table 8. Summary of measurements on nitrous oxide fluxes from natural ecosystems and the corresponding site information

Vegetation type / Ecosystem	Pa†	Ta†	Soil Texture	DC†	pH	Nitrification rate	Flux Range	Mean Flux	Month of measurement	Soil information and location; where known FAO/Unesco (1974) and USDA (1975) classification are given (USDA in parentheses)	Reference ‡
						µg N/d/g soil	µg N m^{-2} h^{-1}				
TEMPERATE REGIONS, NON WETLAND SITES											
Grasslands											
Native shortgrass prairie	300		fine loamy	wd§				9.6	V-IX	Xerosols (ustollic Haplargids), Col., USA	1¶
Native shortgrass prairie	300		15% clay (slope);	wd§				0.9	1 yr	Xerosols (ustollic Haplargid) (slope), Colorado, USA	2¶
Native shortgrass prairie	300		28% clay(bottom)	wd§				1.8	1 yr	Kastanozems (aridic Argiustoll) (swale), Colorado, USA	2¶
Grass			loam	wd§			0.5-2.5	1.5	> 1 yr	loess soil, Mainz, FRG	3
Native prairie	232#		loamy sand	wd§	6.7		0.4-1.8	0.7§	IX-III	Wisconsin, USA	4¶
Prairie	232#		sand	wd§	6.2		0.4-2.1	0.5§	IX-III	Arenosols (typic Udipsamments), Wisconsin, USA	4
Prairie	274#		silt loam	wd§	7.3		0.4-3.2	0.7§	IX-III	lithic Chernozems(lithic Haplustolls), Wisconsin ,USA	4¶
Tall grass prairie, burned	600-800§	10§			6.0		2.2	2.2*	VI-X/IV-X	loess over glacial till, Wisconsin, USA	5
Tall grass prairie	600-800§	10§			6.0		2.5	2.2*	VI-X/IV-X	loess over glacial till, Wisconsin, USA	5¶
Grass (meadow)			sand				2.0-13.0		> 1 yr	Mainz, FRG	3
Grass (wet meadow)	600-800§	10§		pd§	6.1		30.6	30.6*	VI-X/IV-X	Wisconsin, USA	5
Deciduous forests											
Mixed forest				wd§			1.0-3.0		> 1 yr	loess soils, Mainz, FRG	3¶
Deciduous forest	900§	13§						10.3	1 yr	mineral soil, New York, USA	6
Hardwood forest	1300	6-13#				13 kg ha^{-1}y^{-1}		2	summer	soils in glacial till, New Hampshire, USA	7
Oak			sandy loam	wd§	3.7††		4.5-10.5‡‡	8.5	1 yr	brown sandy soil; 7.2%C, Mainz, FRG	8
Oak			sandy loam	wd§	3.6††		3.5-9.5‡‡	7.8	1 yr	Podzols (Spodosols) 1-3cm litter; 2.8% C, Mainz, FRG	8
Oak-hornbeam			sandy loam	wd§	3.7††		5.5-7.5‡‡	7.7	1 yr	Podzols (Spodosols) 1-2cm litter;2.4%C, Mnz,FRG	8
Oak-hornbeam			sandy loam	pd§	3.4††		5.5-7.5‡‡	10.6	1 yr	pseudogley, 1-3 cm litter; 3.4%C, Mainz, FRG	8
Hornbeam-oak			sandy loam	wd§	4.7††		1.5-3.5‡‡	3.0	1 yr	eutric Cambisols (typic Dystrochrepts), 2.8%C,	8¶
Hornbeam-oak			sandy clay loam	wd§	6.7††		2.5-4.5‡‡	3.8	1 yr	eutric/dystric Cambisols (Typic/Dystric Eutrochrepts) 3.6% C, Mainz, FRG	8¶
Black oak					5.0		5.4	15.5*	VI-X/IV-X	Wisconsin, USA	5
Black oak, 80 yr			sandy loam	wd	sl. acid§		-0.01-0.56	0.27	1 yr	Podzols/Cambisols (entic Haplorthods), stony; 5.2-7.8 cm litter, Mass., USA	9¶
Coniferous forests											
Red pine, 62 yr			sandy loam	wd	sl. acid§		-0.08-0.42	0.17	1 yr	Podzols/Cambisols (entic Haplorthods), stony; 4.3-4.9 cm litter, Mass., USA	9
White pine, 40 yr	600-800§	10§			4.5		27.7	36.4*	VI-X/IV-X	Wisconsin, USA	5

Table 8. (continued)

Vegetation type / Ecosystem	Pa†	Ta†	Soil Texture	DC†	pH	Nitrification rate µg N/d/g soil	Flux Range µg N m^{-2} h^{-1}	Mean Flux	Month of measurement	Soil information and location; where known FAO/Unesco (1974) and USDA (1975) classification are given (USDA in parentheses)	Reference ‡
TEMPERATE REGIONS, WETLAND SITES											
Forest, organic soil			organic	pd				11.4	1 yr	Florida, USA, Everglades	6¶
Marsh (sedge meadow)			organic	pd	7.0		0.7 1.1*		VI-X/IV-X	Wisconsin, USA	5
Salt marsh (sp. alterniflora)			organic	pd				3.5	1 yr	Louisiana, USA	10¶
Brackish marsh (sp. patens)			organic	pd				5.4	1 yr	Louisiana, USA	10¶
Fresh marsh (Panicum hemitomon)			organic	pd				6.3	1 yr	Louisiana, USA	10¶
Open water, salt								1.1	1 yr	Louisiana, USA	10
Open water, brackish								2.4	1 yr	Louisiana, USA	10
Open water, fresh								3.9	1 yr	Louisiana, USA	10
Marsh drained	600-800§	10§	organic	wd	6.1		64.8 148.7*		VI-X/IV-X	Wisconsin, USA	5
TROPICAL REGIONS, NON WETLAND SITES											
Savannas											
Savanna (grassland/woodland)	1000	15-20§	sandy loam/sand	wd§	<6††		2.0-6.4	4	II	Venezuela; dry season	11¶§§
Savanna (grassland/woodland)	1000	15-20§	sandy loam/sand	wd§	<6††			17	II	Venezuela; watered soil	11¶§§
Undisturbed forests											
Terra firme moist forest	2000	25.8		wd§				43	IV	Ferralsols (Oxisols), Manaus, Brazil	7¶
Terra firme moist forest	2000	25		wd§				13	XII	Ferralsols (Oxisols), Manaus, Brazil	12¶
Terra firme moist forest	2000	24		wd§				31	III	Ferralsols (Oxisols), Manaus, Brazil	12¶
Primary tropical moist forest	4300	22		wd§				8	VII	Tena, Ecuador	12¶
Elfin cloud forest		19		wd§				11	IV	Puerto Rico	12¶
Colorado/Palm forest		20		pd				3	IV	Puerto Rico	12
Primary forest	4100	25				0.16-2.14	29-52	40	X	Old alluvial, La Selva, Costa Rica	13¶
Primary forest	4100	25				0.14-2.10	7-25	16	X	Old alluvial, La Selva, Costa Rica	13¶
Primary forest	4100	25				1.42-2.16	14-32	23	X	Soils formed in basalt, La Selva, Costa Rica	13¶
Primary forest	4100	25				1.37-1.67	41-83	62	X	Soils formed in basalt, La Selva, Costa Rica	13¶
Primary forest	4100	25				2.44-3.62	12-24	18	X	Soils formed in basalt, La Selva, Costa Rica	13¶
Primary forest	4100	25				2.21-2.73	37-61	49	X	Soils formed in basalt, La Selva, Costa Rica	13¶
Primary forest (75 yr)	2700	22		wd§		1.88-24.6	9-43	26	VII	Andosols (Andepts), Turrialba, Costa Rica	13¶
Lowland forest, undisturbed	2200	25		wd§			15-35		1 yr	Ferralsols (Oxisols), Manaus, Brazil	14¶
Terra firme forest	1770	26	clay	wd§	4.0	1.88-2.44	9.4-12.6	11.0	VIII	Ferralsols/Acrisols (Oxisols/Ultisols), ridges	15¶
Terra firme forest	1770	26	clay	wd§	4.0	1.42-1.66	8.7-13.3	11.0	VIII	Ferralsols/Acrisols (Oxisols/Ultisols), slopes	15¶
Campinarana forest	1770	26	sand§	pd§	3.7	0.49-0.73	8.3-11.7	10.0	VIII	Podzols/Arenosols (Spodosols/Psamments); Amazonia, valley	15
Terra firme forest	2200	25	clay	wd§		1.02-1.20		19	IV-V	Ferralsols (Oxisols), Manaus, Brazil	16¶
Terra firme forest	2200	25	sand	wd§		0.05-0.09		3	IV-V	Arenosols (Psamments), Manaus, Brazil	16

Table 8. (continued)

Vegetation type / Ecosystem	Pa†	Ta†	Soil Texture	DC†	pH	Nitrification rate µg N/d/g soil	Flux Range µg N m^{-2} h^{-1}	Mean Flux µg N m^{-2} h^{-1}	Month of measurement	Soil information and location; where known FAO/Unesco (1974) and USDA (1975) classification are given (USDA in parentheses)	Reference ‡
Primary montane forest	2200	16		wd§		-0.04		0	I	Andosols (Andepts) 200 y, Hawaii	13
Primary montane forest	2200	16		wd§		-0.04	0.4-2.0	1.2	I	Andosols (Andepts), 200 y, Hawaii	13
Primary montane forest	3500	16		wd§		0.06-0.18	0.5-1.7	1.1	I	Andosols (Andepts), 1000 y, Hawaii	13
Primary montane forest	3500	16		wd§		-0.04-0.1	1.3-3.9	2.6	I	Andosols (Andepts), 1000 y, Hawaii	13
Primary montane forest	4500	16		wd§		-0.11-1.07	1.0-2.6	1.8	I	Andosols (Andepts), 4000 y, Hawaii	13
Secondary forest (logged)	2000	26		wd§				23.1	VII-VIII	Ferralsols (Oxisols), Manaus, Brazil	18¶
Dry tropical forest	748	25		wd§				9.1	IX	Cambisols (Entisols), Chamela, Mexico	17¶
Dry tropical forests	748	25		wd§				2.0	III	Cambisols (Entisols), Chamela, Mexico	17¶
Disturbed forests											
Secondary forest, 20 yr	4200	25				1.92-2.56	4-52	28	X	Recent alluvial, La Selva, Costa Rica	13
Secondary tropical moist forest		22		wd§				7.4	VII	Ecuador	12
Tabanuco forest (secondary)		23		wd				42.5	IV	Acrisols (Ultisols), Puerto Rico	12
Mahogony plantation		30		wd§				7.9	IV	Puerto Rico	12
Lowland forest, cleared, burned	2200			wd§			15-35		1 yr	Ferralsols (Oxisols), Manaus, Brazil	14
TROPICAL REGIONS, WETLAND SITES											
Varzea	2200	25		pd		0.26-0.34		1	IV-V	Gleysols (?), Manaus, Brazil; waterlogged	16¶

† Pa, mean annual precipitation (mm); Ta, mean annual temperature (°C); DC, drainage class (wd = well drained; spd = somewhat poorly drained; pd = poorly drained).

‡ 1, Mosier et al. (1981); 2, Parton et al. (1988); 3, Seiler and Conrad (1981); 4, Cates and Keeney (1987); 5, Goodroad and Keeney (1984a); 6, Duxbury et al. (1982); 7, Keller et al. (1983); 8, Schmidt et al. (1988); 9, Bowden et al. (1990); 10, Smith et al. (1983); 11, Hao et al. (1988); 12, Keller et al. (1986); 13, Matson and Vitousek (1987); 14, Luizao et al. (1989); 15, Livingston et al. (1988); 16, Matson et al. (1990); 17, Vitousek et al. (1990); 18, Keller et al. (1988).

§ = interpretations: A.F. Bouwman.

¶ = measurements used in the comparison of measurements and modeled N2O scalar for the corresponding sites.

= during measurements.

* = first value refers to summer autumn 1979; second value to March-November 1980; no annual figures available.

†† = soil pH measured in KCl solution.

‡‡ = 50% probability with 95% confidence range.

§§ = for the dry season the measurement data were compared with the modeled N2O value averaged for May-October, for the rainy season the measurement data for the watered soil were compared with the modeled N2O values averaged for November-May.

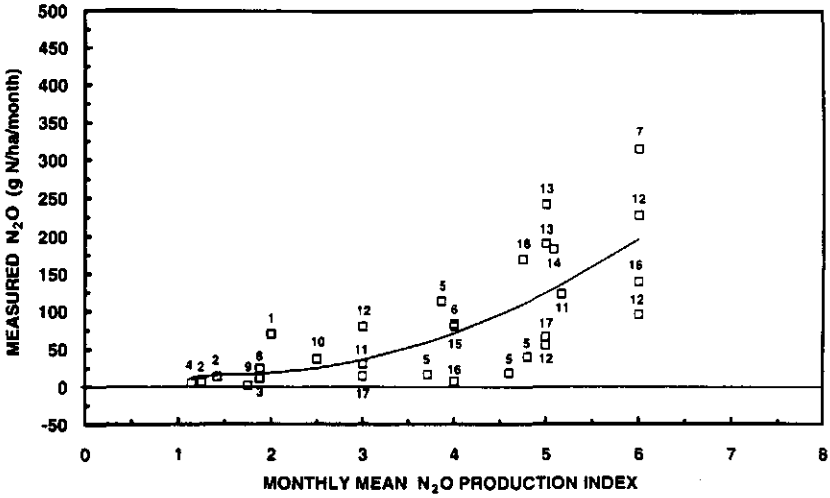

Figure 5. Relationship between measured N_2O fluxes and modeled N2O from the reference case for a series of field sites. Labels refer to sites listed in Table 8. The solid line representes the best quadratic fit: for N2O > 1.5, flux = 9.01 x $(N2O)^2$ - 27.87 x (N2O) + 38.22; for N2O $\leq$ 1.5, flux = 11.3 x (N2O).

identified in the soils data base used here. We repeated the above calculation excluding areas designated as wetlands by Matthews and Fung (1987). In this calculation, emission and consumption of N_2O in wetlands are assumed to be equal. Wetlands cover only about 4% of the ice-free land area, and their exclusion from the analysis causes a decline in the modeled global N_2O emission of a few percent. We do not know whether N_2O emission from tropical wetlands is overestimated here, but the analysis suggests that the global contribution of wetlands to the N_2O budget is minor.

2.5. CONCLUSIONS

The model results show that spatial and seasonal distributions of N_2O production are very similar to climate patterns. Low winter temperatures limit N_2O production in temperate regions while in the tropics, temperature does not limit the processes involved in N_2O production. In the wet tropics, where organic matter inputs are high and both soil moisture conditions and temperatures are favorable, N_2O indices (N2O) are among the highest. This is consistent with high emissions reported for tropical forests (Table 8). While it is tempting to conclude that climate is the most important control factor, we note that C/N ratios and lignin content of the litter are not included explicitly as controls in the model. However, as their geographic variations mirror, to some extent, those of climate, we conclude that the pattern of N_2O production is determined, to first order, by the pattern of climate variation. Together with physical soil conditions, climate determines oxygen limitation and water availability. The factors FERT and O2 introduce variability among sites, without producing major impacts on the level of the N2O values.

The lack of major differences in predicted N2O between the reference case and experiment 3, in which the role of soil fertility is investigated, suggests that the model does not adequately reflect fertility levels among soils. In practice, only broad properties for soil groups are used and information to distinguish fertility differences among soil types within groups is lacking. Exclusion of the fertility index increases the variability of the N_2O potential, reflecting the simplicity of the scale used.

The variability in N_2O production (expressed as the percentage of the standard deviation of the N_2O index relative to the mean within each soil group) is highest in ecosystems with overall low N2O values, i.e., in moist temperate climates, and in both temperate and tropical environments under arid and semiarid regimes. Apparently, where climate (soil water and/or temperature) is most limiting, variability in other factors results in higher percent standard deviations than those in regions with climatic conditions favorable for N_2O production; absolute standard deviations are highest in areas with high N_2O production potential.

Comparison with measurement data shows that predicted levels of N_2O production are in general agreement with measured fluxes, even though responses to long-term mean climate are compared with responses to local weather conditions prevailing during measurement periods. The ~30 flux measurements used for the comparison represent only six different ecosystems (wetlands, temperate forests, steppe, tropical savannas, tropical dry forests, and tropical rain forest). It is difficult to draw conclusions about the relative importance of various parameters on the basis of this limited data set. In addition, it is difficult to assess published measurements of nitrous oxide fluxes. In many measurement reports, site descriptions are incomplete: climate data are often absent, or provided only as annual mean precipitation and temperature; imprecise descriptions of conditions during the period of measurements are common; complete descriptions of chemical and physical soil characteristics, such as pH, texture, structure, drainage condition and soil classification, are rarely reported.

Because of the simplicity of the model and the sparcity of data, the estimate of global N_2O emission from natural ecosystems based on the regression analysis is extremely uncertain. To improve models of this type, more measurements are needed in those places expected to have high N_2O fluxes, and in regions or ecosystems which may be important contributors to the global production of N_2O due to their large area. Globally, only about 30 measurements have complete enough information to allow comparison; these do no include any data for Asia, Africa or Australia. We do not know how well the model performs in these areas with different environmental conditions and soils.

The role of soil sinks needs to be addressed in future. Consumption of N_2O has been reported (e.g., Ryden, 1981, 1983). If this is a common phenomenon, it may have important implications for the N_2O global budget (Cicerone, 1989). Data on consumption are too scarce to describe this process in a global model such as this one.

The 1° resolution of the soil data base does not reflect the detail of the 1:5 M soil map (FAO/Unesco, 1974-1981) used in its compilation. In digitizing the soil map, only the dominant soil type was recorded (Zobler, 1986) so that many of the associated or included soils are not represented. These minor soils may be of importance for trace gas emissions. In particular, wetlands and hydromorphic soils are poorly represented because they occur frequently as linear and scattered features that do not dominate at the 1° resolution. Even at the 1:5M scale of the FAO/Unesco soil map, they are underrepresented (Van Diepen, 1985). Parallel to the need for greater spatial resolution is the need for greater information resolution about soil fertility, drainage, and texture. In the model, only five levels of these properties are resolved, although there are >100 soil types. To improve the model, soil data bases including a number of soil characteristics vital to evaluating fertility are required. More spatial and temporal detail in data bases of vegetation (NPP) and litter composition would allow a better description of the temporal and geographic variability of controls on N_2O fluxes and, possibly, episodic pulses of N_2O.

The latitudinal and seasonal distributions of N_2O fluxes modeled here may be used as inputs to two- and three-dimensional models of the atmosphere to test hypotheses about the N_2O budget.

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APPENDIX A

The primary gridded data sets used in the model, commented upon briefly in the text, are discussed more fully below.

A.1. *Climate*

Shea (1986) produced climatologies of monthly surface air temperature and precipitation, at 2.5° resolution for the globe, from station observations. Lacking a global climatology for soil temperatures, we used surface air temperatures. This may introduce phase errors, of up to a season in middle and high latitudes, in the seasonality of N_2O production.

A.2. *Soil Type, Texture, Fertility, and Drainage*

The global distribution of soil properties was obtained from the data base of Zobler (1986), compiled in digital form at 1° resolution from the FAO/Unesco (1974-1981) Soil Map of the World. The 1:5M FAO/Unesco soil maps distinguish dominant soil units, associated soils and inclusions, topsoil texture of the dominant soil and slope. On the maps, additional information on phases is indicated with special symbols. The full digital data base of Zobler (1986) includes distributions of major and associated soil units, soil texture, slope, and phase. The data sets used in this study are major soil units and soil texture.

The FAO/Unesco (1974-1981) soil classification system has two levels: soil groups and soil units. There are 26 soil groups encompassing 106 soil units. The soil groups and soil units are classified on the basis of diagnostic horizons and diagnostic properties. Descriptions of the soil groups and soil units can be found in the works by FAO/Unesco (1974-1981), Fitzpatrick (1983) and Driessen and Dudal (1991). Two of the soil units, gelic Planosols, and ferric Podzols, do not occur in the digital data base because they never dominate at 1° resolution.

For this study, soil units are aggregated into 23 new groups. Table 1 lists the 23 groups and their areas, presented under 10 higher-level soil clusters (I-X). The global distribution of the 23 new groups is shown in Plate 1. Soils are formed through the impact of climate, vegetation, fauna (including humans) and topography on the parent material. The relative influence of these soil-forming factors varies among sites, explaining the considerable variety found in soils. Globally, climate is the major soil-forming factor and under similar climatic conditions, soil development is sufficiently consistent to form a basis for soil classification. Such zonal soils are represented in Table 1 by clusters II-IV and VI-VII. Variations occur where parent rock (clusters VIII and X), physiographic position (IX) or other local features dominate soil formation. These soils are intrazonal and azonal soils with no clear relation to vegetation and climate.

In this model, individual gelic soil types are combined to form the permafrost soil cluster. In addition, several soils are combined following the new terminology of FAO/Unesco (1988): all shallow soils (Lithosols, Rankers, and Rendzinas) are grouped as Leptosols (group 21), and weakly-developed soils under arid and semiarid moisture regimes, Xerosols and Yermosols, are grouped as desert soils (group 14).

The data base of soil units provides the basis for deriving information on two factors in the model: fertility and drainage. Indices for these factors, ranging from one to five, are associated with each of the 106 soil units as shown in Table 1 and discussed in section 3.

The reliability of the FAO/Unesco soil map is not spatially uniform. Reliability is particularly low in the Amazon basin, the former Soviet Union, parts of eastern Africa, and Europe. Additional information has become available since compilation of the FAO/Unesco soil maps in the 1960s, reflected in the revised legend of FAO/Unesco (1988).

Topsoil texture is defined as the relative proportions of clay (particles < 2 µm), silt (2-50 µm), and sand (50-2000 µm) in the top 30 cm of the soil. The FAO/Unesco soil maps have three broad texture classes for the topsoil: coarse, medium, and fine. In addition to these texture classes, the digital data set identifies several texture combinations; the organic class is for nonmineral topsoils (Table 2). Topsoils with coarse textures have <18% clay and >65% sand and include sands, loamy sands and sandy loams. The medium texture class, with <35% clay and <65% sand, includes sandy loams, loams, sandy clay loams, silt loams, silts, silty clay loams, and clay loams. Fine-textured topsoils have >35% clay and include clays, silty clays, sandy clays, clay loams, and silty clay loams. The reader is referred to the texture triangle in FAO/Unesco (1974-1981) and to U.S. Department of Agiculture (1975) for further information on characteristics of texture classes.

A.3. *Vegetation*

The satellite-derived normalized difference vegetation index (NDVI) is a measure of the primary productivity of vegetation (Box et al., 1989). The NDVI is calculated as the difference between radiances in the visible (0.58-0.68 µm) and near infrared (0.725-1.1 µm) portions of the spectrum normalized by the sum of the radiances. The Advanced Very High Resolution Radiometer (AVHRR) on board the NOAA series of polar-orbiting satellites (Tarpley et al., 1984) routinely measures the radiances. The weekly maximum NDVI's at ~25 km resolution obtained from NOAA for 1984 were further sampled and averaged for this study. First, the maximum monthly NDVI at 25 km resolution was retained from the weekly maxima. These dates were then averaged to 1° resolution. The digital data base of vegetation types compiled by Matthews (1983) is used for verifying measurement site characteristics and for analysis of model results.

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Direct emission of nitrous oxide from agricultural soils

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Chapter 3

DIRECT EMISSION OF NITROUS OXIDE FROM AGRICULTURAL SOILS

Abstract. This analysis was based on a review of published measurements of nitrous oxide (N_2O) emission from fertilized fields. From the literature data selections were made to analyze the importance of factors that regulate N_2O production, including soil conditions, type of crop, nitrogen (N) fertilizer type and soil and crop management. Reported N_2O losses from anhydrous ammonia and organic forms of N fertilizers or combinations of organic and synthetic N fertilizers are higher than those for other types of N fertilizer. However, the management and environmental conditions represented by the set of measurement data is too limited to be used for estimating emission factors for each fertilizer type individually. The literature data are appropriate for estimating the order of magnitude of emissions. The fertilizer-induced N_2O emission is higher for measurements covering longer periods than for measurements which represent short periods. Therefore, a simple method to estimate the total annual direct N_2O emission from fertilized fields was based on those measurements covering periods of one year, resulting in the following equation: N_2O emission (kg N $ha^{-1}yr^{-1}$) = 1 + 1.25 ± 1% of the N application (kg N $ha^{-1}yr^{-1}$). The relation is independent of the type of fertilizer. Although the above regression equation includes considerable uncertainty, it may be appropriate for global analyses.

3.1. INTRODUCTION

Nitrous oxide (N_2O) plays an important role in the atmospheric radiative balance and in the stratospheric ozone chemistry. A great number of major and minor sources have been identified, yet there is considerable uncertainty in the source strengths. Part of the uncertainty arises from the paucity of measurements of N_2O fluxes. Another part stems from the difficulty of extrapolating measurements of biogenic fluxes from soils and aquatic sources to larger scales, because of their extreme heterogeneity both in space and time. For abiogenic sources, such as fossil fuel combustion and industrial processes, political, economic and cultural factors are major uncertainties in making extrapolations. Khalil and Rasmussen (1992) recently presented a global N_2O budget indicating that the uncertainty for most N_2O sources amounts to at least a factor 2.

There is also considerable uncertainty in the estimates of N_2O emission from the world's cultivated fields. During recent years few new measurements of N_2O fluxes in agricultural fields have been published, despite the interest in increasing concentrations of greenhouse gases in the atmosphere. Many assessments rely on flux measurements carried out during the period 1980-1990. For example, attempts have been made to estimate the N_2O emission caused by synthetic nitrogen (N) fertilizers (Eichner, 1990) and synthetic and organic fertilizers (Bouwman, 1990) based on literature reviews. Synthesis of current knowledge resulted in an estimated annual emission from cultivated fields of 0.03 - 3 Tg N_2O-N (Watson et al., 1992).

The direct efflux of N_2O from agricultural fields is possibly only part of the emission caused by N fertilization. Nitrogen may leach from the soil and form a source of N_2O fluxes from groundwater by degassing or from surface waters. Nitrogen taken up by plants may be consumed by humans or animals. Denitrification of the nitrogen in their excreta may, once in the environment, form a source of N_2O.

Many reviews have been published on the microbial processes responsible for N_2O production, nitrification and denitrification (e.g. Firestone and Davidson, 1989). The release of N_2O may be a by-product of nitrifiers that denitrify nitrite (NO_2^-) under oxygen stress (Poth and Focht, 1985). Under moist and oxygen depleted conditions denitrification is generally the major source of N_2O, and both the rate of denitrification and the conditions that influence the ratio N_2 / N_2O determine the N_2O emission (Davidson, 1991). Many factors regulate nitrification and denitrification. A review of these factors is presented by Bouwman (1990). In summary they include:

- Soil moisture and temperature, regulating microbial processes;
- Soil oxygen availability, a control of denitrification; oxygen supply is determined by the soil water content and the rate of microbial respiration;

- Concentrations of NO_3^- and $NH4^+$, influencing the reaction rates; obviously the plant roots play a role here (i) by consuming the nutrients and (ii) as a source of nutrients from residues or exudates;
- Organic carbon as energy source for denitrifiers;
- Soil reaction or pH, influencing nitrification and denitrification and the ratio N_2/N_2O.

The method proposed by Eichner (1990) to calculate N_2O emission from different fertilizer types was adopted by the IPCC for making country estimates (OECD, 1991). The wide range of uncertainty in this estimate is, however, not appropriate in global inventories.

Another method for estimating the N_2O emission from fertilized fields was based on N application, weather conditions, soil properties, soil, crop and water management with the complex DNDC model (Changsheng Li et al., 1992a; 1992b). Other more simple approaches are the mechanistic model developed by Mosier and Parton (1985) for Colorado grasslands, and the model relating N_2O emission to the water filled pore space (Davidson, 1991). These models describe conditions for N_2O production reasonably well. They were developed and validated for the conditions of a single site. Extrapolation of emissions should be based on validation for a wide variety of conditions. This requires soil data and daily weather data that are not available at the global scale.

The aim of this analysis is to develop an approach to estimate global direct emissions of N_2O from fertilized agricultural fields. Measurement data of N_2O emission in relation to N fertilization from various studies were collected from the literature. Section 3.2 briefly discusses a number of the regulating factors of N_2O production on the basis of this data set. Another important aspect that will be discussed is the length of the period covered by the flux measurements and their frequency. On the basis of this analysis and comparison with earlier estimates a method to estimate annual emission from fertilized fields will be described in section 3.3.

3.2. COMPARISON OF EXPERIMENTS

The measurement data collected from the literature presented in the Appendix include experiments in cropped and unplanted plots with different soils fertilized with different types of N fertilizers, ranging from organic fertilizers and combinations of synthetic and organic fertilizers, and measurements in unfertilized control plots. The emission of N_2O is presented as:

- The *total N_2O emission* during the period covered by the measurements.
- The *fertilizer-induced N_2O emission*, calculated as the difference in emission between the fertilized and the control plot, presented as a percentage of the fertilizer N applied. The fertilizer-induced N_2O emission ranges between 0.0% and 6.8% of the N application for 87 experiments for mineral soils in the Appendix that included a control plot.
- The total N_2O emission as a percentage of the N applied. The total N_2O emission from 180 experiments for mineral soils in the Appendix ranges between 0.0 to 7.8% of the N application.

The flux measurement technique, duration of measurements and sampling frequency is indicated for all experiments in the Appendix. Details on the techniques used can be found in the individual reports listed. Gas collection chambers on the soil surface are commonly used to quantify the N_2O flux from the soil to the atmosphere. Reviews of the theoretical and practical problems which cause variability in gas flux measurements using chambers are presented by Mosier (1989).

Several factors regulate the production, consumption and emission of N_2O. A number of these will be discussed briefly on the basis of the data in the Appendix. Another important aspect that will be discussed is the length of the period covered by the flux measurements and their frequency.

- *Duration of measurements*. The length of the period covered by the measurements may influence the amount of N_2O stemming from fertilizers that is captured. The average fertilizer-induced N_2O emission is 0.7 ± 1.1% of the N application based on all experiments for mineral soils (Appendix). The average fertilizer-induced N_2O emission is 0.8 ± 1.2% for experiments with a duration of measurements of > 30 days, 1.1 ± 1.4% for experiments covering > 100 days, and 1.6 ± 0.4% for experiments covering > 200 days. This suggests that if measurements of N_2O fluxes from fertilized fields are extended over longer periods, the observed N_2O emission is higher. This also suggests, that it is necessary to measure fluxes during prolonged periods to account for all the fertilizer-induced emission.

- *Frequency of measurements.* Brumme and Beese (1992) observed that N_2O flux measurements done once per week tend to overestimate the total emission estimate relative to daily observations by 20%. In many studies the frequency of measurements is once per day or once in 2 or 3 days, particularly in periods of high fluxes shortly after fertilizer application (Appendix). In some studies the measurements were done once per week. These differences in frequency of flux measurements may form another source of uncertainty.
- *Presence and type of crop.* Many studies included fertilized but unplanted fields (Appendix). Since there is no uptake of nitrogen from the soil, denitrification and associated N_2O emission may be higher than in cropped fields. The mean fertilizer-induced N_2O emission for unplanted fields is 0.9 ± 1.5% of the N application, while the mean for fields with crops or grass is 0.4 ± 0.6%.

Ungrazed grassland plots (0.4 ± 0.7, N = 17) showed somewhat lower N_2O emission than cropped fields (0.4 ± 0.6, N = 28). Grasses have a longer growing season than crops, leading to more N uptake and less denitrification in grasslands than in cropped fields. This is not confirmed by the data, possibly because most measurements covered only the spring and summer period and not the full year.

For most experiments in the Appendix it is impossible to conclude whether differences are caused by the type of crop, the amount and type of N fertilizer or the management practices. However, in some experiments the crop or the combined effect of crop and management clearly determine the N_2O emission, i.e. wetland rice and leguminous crops.

Wetland rice in experiment 15 and 36 showed low N_2O fluxes, and the N_2O emission from dryland rice fields was somewhat higher (experiment 25). This may be caused by the low availability of oxygen, which is unfavourable for nitrification. Moreover, low oxygen availability may lead to a low N_2O / N_2 ratio in denitrification products. However, Byrnes et al. (1993) showed that drainage of wetland rice fields may give rise to significant N_2O emission. As drainage of rice fields was not considered in experiments 15 and 36, the reported N_2O emissions may be underestimated.

Fields with legumes showed high N_2O emission. As leguminous crops usually receive little or no N fertilizer, these high N_2O emissions may be attributed to N inputs from symbiotic N fixation. The only examples in the Appendix are alfalfa (2.3-4.2 kg N_2O-N $ha^{-1}yr^{-1}$, experiment 17), soybeans (0.34-1.97 kg N_2O-N $ha^{-1}yr^{-1}$, experiment 41) and clover (experiment 14). The measurements in the clover fields did not result in high fluxes, perhaps because N fertilizer added in this experiment prevented N fixation. Unfortunately the measurement period was not reported.

- *Crop residues.* The data in the Appendix indicate that N coming from decomposition and mineralization of crop residues may also contribute to N_2O fluxes. The effect of crop residues can best be illustrated by comparing experiments in Iowa on typic Haplaquolls (experiments 5 and 6). For both the control and the fertilizer treatment experiment 6 showed much higher N_2O emission than experiment 5. In experiment 5 maize residues were incorporated in the surface layer, while in experiment 6 soybean residues were left on the surface to decompose.

Experiment 20 included plots with rye grown as a cover crop after harvest of the previous crop. The rye crop was incorporated before sowing tobacco and resulted in lower N_2O emission than plots with manure or alfalfa residue.

- *Tillage*. Surface application of N fertilizers to residues in plots with minimum or reduced tillage leads to high N_2O emission (experiment 20). This is consistent with experiments 8 and 13 that showed lower N_2O emission from ploughed plots cropped to winter wheat fertilized with NH_4NO_3 than unploughed, directly sown plots.
- *Source and amount of nitrogen*. The variability in N_2O fluxes is extremely high for all N fertilizer types and all application levels (Figure 1). Fluxes ranging between 0 and 30 kg N_2O-N $ha^{-1}yr^{-1}$ were observed in plots with mineral soils. The results for the unfertilized control plots (Appendix) range between -0.6 and 4.2 kg N_2O-N ha^{-1} (avarage 0.8, standard deviation 1 kg N ha^{-1}, N = 55). The variability may be caused by many different factors, of which the history of fertilization and management may be important ones. For none of the fertilizer types there is a clear relation between N application and N_2O emission, but some forms of N show consistently higher N_2O emissions than other types.

Fluxes of N_2O from plots amended with combinations of organic and synthetic fertilizers are generally high. It should be noted that the literature reports listed in the Appendix presented the N in organic fertilizers as total N, including mineral nitrogen and organic nitrogen. This indicates that there is uncertainty in the amount of N applied, because part of the organic N is not directly available, while the volatilization of NH_3 after application of organic fertilizers was not accounted for. Emissions from fields fertilized with NO_3^--based fertilizers and combinations of organic and NO_3^- fertilizers from experiment 31 were relatively high compared to results of other experiments. Measurements in experiment 31 were carried out immediately after irrigation and rainfall events, and this have caused an overestimation of both denitrification and N_2O emission extrapolated over the growing season.

Within the group of synthetic fertilizers, anhydrous ammonia induces high N_2O fluxes. This may, however, not be the result of the type of fertilizer, but merely of the mode of application (see below).

- *Mode of fertilizer application*. Some experiments indicated an important effect of the mode of fertilizer application. Most fertilizers were broadcast onto the soil surface and incorporated by tillage. Injection is the customary method of applying anhydrous ammonia. This may produce highly alkaline soil zones of high ammonium concentration (Breitenbeck and Bremner, 1986a) that may lead to high N_2O production (Bouwman, 1990). Experiments 4, 5, 6 and 10 showed that deeper injection of anhydrous ammonia lead to higher N_2O emission than shallow injection. Another example is experiment 36 where urea drilled into the soil caused higher N_2O emission than top-dressed urea for the high N application of 180 kg N ha^{-1}.

It is difficult to explain why deeper injection resulted in higher N_2O emission. The N loss by NH_3 volatilization from applied anhydrous ammonia is probably lower for deep than for shallow injection. However, if the ammonia is injected deeper, the transport of the N_2O formed is over a longer distance, which increases possibilities for further N_2O reduction.

- *Timing of fertilizer application*. The set of data does not include sufficient experiments studying the effect of timing of fertilizer application. Application in periods when the crop actually needs nutrients will reduce N losses by denitrification and leaching, thereby also reducing N_2O losses (Mosier, 1993).
- *Soil type and properties*. In experiments 4 and 6 different soils were included to measure the effect of different N fertilizers on N_2O emission. Unfortunately the authors of the report did not explain the differences. A possible explanation may be the soil texture. In experiments 6 and 8 similar soils with different textures were included. The heavy textured soils showed higher N_2O emission than the soil with lighter texture. In contrast, in experiment 4 the light textured soils showed higher emissions than heavier textured soils.

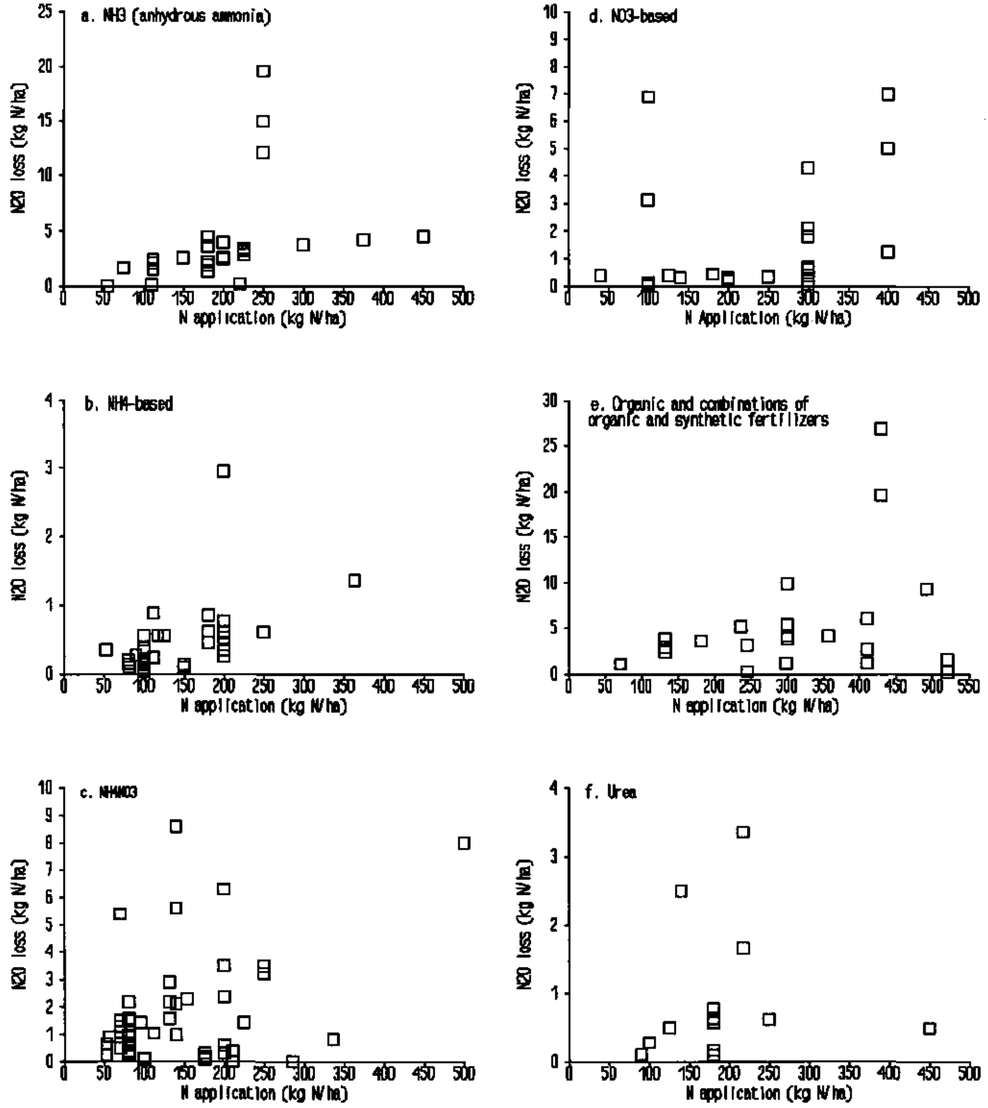

Figure 1a-f. Relation between N fertilizer application and N_2O emission from mineral soils for experiments listed in the Appendix, independent of the period covered by the measurements, presented for (a) anhydrous ammonia (NH_3); (b) ammonium (NH_4)-based fertilizers; (c) ammonium nitrate (NH_4NO_3); (d) nitrate (NO_3)-based fertilizers; (e) organic fertilizers and combinations of organic and synthetic fertilizers; (f) urea.

Drained organic soils showed high N_2O emission of up to 100 kg N_2O-N $ha^{-1}yr^{-1}$ (experiments 17 and 43). Although the fields studied were not fertilized, the mineralization of the organic soil material is a significant source of N. Terry et al. (1981) estimated that mineralization of nitrogen may amount to up to 1300 kg N $ha^{-1}yr^{-1}$ (see Appendix). The observed N_2O emission from these soils constitutes a fraction of < 1 to > 10% of N mineralization (Appendix).

Another soil property that may affect N_2O emission is the soil reaction or pH. The pH may affect nitrification, denitrification and the N_2O reduction. Generally it is thought that N_2O reduction is inhibited at low pH (various references quoted in Bouwman, 1990; Bouwman et al., 1993). Plots with identical soils of different pH established in 1962 gave no measurable differences in N_2O emission (experiment 20). This may be due to adaptation of denitrifiers to soil pH (Parkin et al., 1985).

- *Soil drainage.* Experiment 11 concentrated on drainage of a poorly drained soil with stagnant water (stagnogley). Draining the soil caused a decrease in the N_2O emission. For all experiments the soil was classified as well drained, poorly drained or moderately well drained, based on data given in the reports or on the soil taxonomic class or soil description. For example, Paleudalfs are considered well drained, while the name Calciaquolls suggests hydromorphic properties and poor drainage. However, there was no clear relation found between soil drainage and N_2O emission for the experiments listed.

3.3. METHOD TO ESTIMATE THE DIRECT N_2O EMISSION FROM FERTILIZED FIELDS

The method presented by Eichner (1990) accounts for the fertilizer-induced emission, i.e. the emission from a fertilized plot minus that from a control plot, determined during the measurement period. Eichner (1990) calculated the fertilizer-induced N_2O emission as a percentage of N fertilizer applied specified for a number of fertilizer types (Table 1). There are a number of uncertainties in this method:

- The data sets used by Eichner (1990) and in this study represent only a limited number of climatic, soil and management conditions. For example, Eichner (1990) based the median and range of N_2O emission induced by anhydrous NH_3 on only a few experiments, mostly carried out in Iowa (experiments 3-7). The highest fertilizer-induced N_2O emission (6.8%, experiment 6) was observed in fields where soybean residues were left on the surface to decompose. This may not be representative of worldwide practices in fields where anhydrous ammonia is applied.
- Addition of observations to the dataset of Eichner (1990) can result in changes in the calculated average N_2O losses caused by fertilization. This study includes 14 measurements for anhydrous ammonia that were not reviewed by Eichner (1990), causing a 30% lower fertilizer-induced emission (Table 1). This has important consequences for the estimated emission from the application of anhydrous ammonia, which contributes about 45% to the global N_2O emission from fertilizers based on Eichner's method. The greatest difference is found for urea, where the N_2O emission resulting from this study exceeds the estimate of Eichner (1990) by a factor of 3 caused by only 7 additional measurements.
- The concept of the fertilizer-induced N_2O emission does not yield an estimate of the *total* annual emission. Most measurements listed in the Appendix cover the crop season or shorter periods. Most of the N_2O is usually released during a period of a month after fertilizer application, after which emissions decline to a "background" level (Breitenbeck and Bremner, 1986). This is consistent with most of the literature reports in the Appendix. Although the background emission may be low, the contribution to the annual flux may not be negligible. Moreover, it is uncertain whether this background emission level is influenced by the fertilization and soil management during previous years. As the observed N_2O emission from fertilized lands in the Appendix is significantly higher for longer measurement periods than for shorter measurement periods, the background emission should be included in estimates of the annual emission from fertilized lands.

Table 1. Average and standard deviation of the fertilizer-induced N_2O emission† for different types of N fertilizer reported by Eichner (1990) compared with results from this study

Type	Eichner (1990)‡			This study		
	N§	Average	S.D.¶	N§	Average	S.D.¶
Anhydrous ammonia	12	2.3	2.0	23	1.6	1.6
Ammonium nitrate	8	0.3	0.3	10	0.3	0.3
Salts of ammonium	17	0.1	0.1	20	0.1	0.1
urea	7	0.1	0.0	14	0.3	0.6
Salts of nitrate	15	0.2	0.5	16	0.2	0.4
Organic / combinations of organic and synthetic fertilizers				5	1.5	0.5

† The fertilizer-induced emission is calculated as emission from the fertilized plot minus that from the control plot, presented as percentage of N fertilizer application.

‡ Recalculated from the datatables in Eichner (1990) including N-applications > 250 kg N ha^{-1}, and with corrections in the data from Seiler and Conrad (1981), Conrad et al. (1983) and Christensen (1983).

§ N = number of experiments

¶ S.D. = standard deviation

A simple method is proposed here to calculate the *total annual* N_2O emission from fertilized fields, independent of crop, management, soil conditions and fertilizer type. As noted above the length of the measurement period seems to be important to determine the total of N_2O emission. Figure 1 shows the relation between N-fertilizer application and N_2O emission for all experiments on mineral soils in the Appendix. Clearly, there is no correlation between N application level and N_2O emission if the duration of measurements is not taken into account. For experiments with a duration of N_2O flux measurements of a full year, the correlation is much better. Data presented in Figure 2 for cropped fields and ungrazed grass plots include a variety of different fertilizers (including synthetic, organic and combinations of organic and synthetic N fertilizers), weather conditions and soils. The results from experiment 2 were excluded, because of reported abnormal low precipitation. The experiments for leguminous crops (experiment 17 and 41) were also excluded because the input from N fixation was not reported.

Least squares fitting of the data in Figure 2 to a linear function resulted in equation (1) with r^2 of 0.8:

$$N_2O\text{-flux (kg N ha}^{-1}\text{yr}^{-1}) = 1 + 0.0125 * \text{N application} \tag{1}$$

This relationship was based on 20 different measurements only, and its global applicability is highly uncertain. The *background* emission of 1 kg N_2O-N $ha^{-1}yr^{-1}$ was based on only 5 measurements for unfertilized plots, with a range of emissions of -0.6 to + 3.2 kg N_2O-N $ha^{-1}yr^{-1}$ (experiment 30 and 19, respectively). It is, however, consistent with the average of the 33 measurements covering more than 100 days in unfertilized control plots of 1.2 ± 1.1 kg N ha^{-1}.

The *fertilizer-induced N_2O emission* of 1.25% is close to the calculated 1.1% (± 1.4) fertilizer-induced N_2O emission based on 43 experiments with a duration of measurements of > 100 days where a control plot was included. The 1.25% fertilizer-induced emission is also consistent with the estimate of 1% of Mosier (1993), and with the 0.5-2% N_2O emission from fertilizers estimated by Bolle et al. (1986).

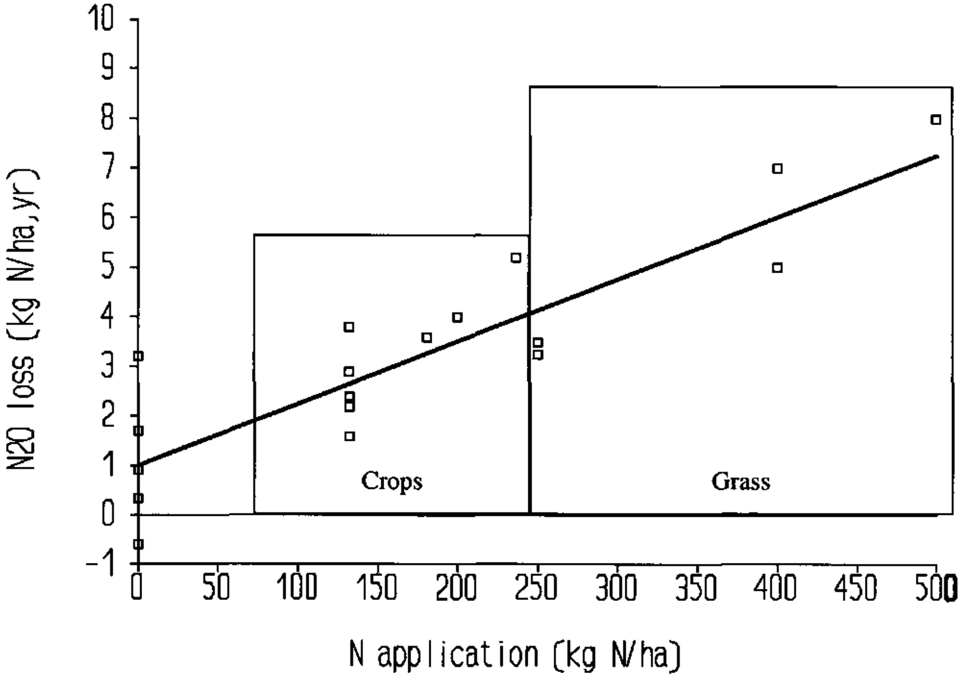

Figure 2. Relation between N fertilizer application and N_2O emission for experiments on plots with mineral soils with a coverage of measurements of 1 year and for N application rates < 500 kg N ha^{-1} yr^{-1}, excluding results for experiment 2 and measurements for leguminous crops (Appendix). The boxes indicate measurements in cropped fields and those in ungrazed grasslands.

3.4. DISCUSSION AND CONCLUSIONS

Although the individual factors that control N_2O production are known, it is impossible to describe their interaction under field conditions on the basis of the data in the Appendix. The processes of nitrification and denitrification and the controls of the reduction of N_2O to N_2 have their specific optimum conditions. These conditions may change from one year to another, and the importance of the different N_2O producing processes may also change as a consequence. The variability in the data is caused by a variety of factors related to management, such as timing of N additions, timing and frequency of irrigation or precipitation, presence or absence of crops, type of crop, history, mode and timing of fertilizer application, and soil management. It is also caused by factors such as local rainfall and temperature that are not manageable.

Byrnes et al. (1990) concluded that N_2O emissions may be more closely related to soil properties than to the N source applied. The comparison in Table 1 suggests that there may be differences in N_2O emission caused by the fertilizer type. However, the addition of a few measurements can drastically change the calculated emission factor for a fertilizer type, as was shown for e.g. anhydrous ammonia. Therefore, the set of data presented in the Appendix is too limited to calculate the N_2O emission specific for each fertilizer type, and it is not likely that in the coming years sufficient new data will be generated. However, the available data are adequate to estimate the order of magnitude of emissions.

A simple approach was developed, based on a background emission of 1 kg N_2O-N $ha^{-1}yr^{-1}$ plus a fertilizer-induced N_2O emission of 1.25% of the N application. This method applies to all fertilizer types, and may not be adequate to estimate emissions for local conditions or specific crops. The range of uncertainty for the fertilizer-induced N_2O emission is 0.25 - 2.25% based on the full set of data, but excluding the extremes (A.R. Mosier, 1994, personal communication). The method may be adequate for global analyses. Assuming that the global N fertilizer use in 1990 of 80 Tg N yr^{-1} (FAO, 1991) is applied exclusively to arable fields and that no organic fertilizers are used, the background emission calculated for the global arable land area of $1440x10^6$ ha is 1.4 x 10^{12} g N_2O-N yr^{-1} and the fertilizer-induced emission is 1 x 10^{12} g N_2O-N yr^{-1}.

This estimate does not include N_2O emissions from leguminous crops. These crops usually receive little or no N fertilizer. The N_2O emissions from fields with leguminous crops may be considerable. These high N_2O emissions may be attributed to N inputs from symbiotic N fixation. The global area of leguminous crops is 145 Mha (FAO, 1991), about 10% of the total arable land. This area does not include legumes grown as green manures not reported by the FAO (1991), and legumes in grasslands and N fixing grass species. The N inputs from legumes to agricultural systems may be of the same order of magnitude as global synthetic N fertilizer use (Duxbury et al., 1993), indicating the potential importance for the N_2O cycle.

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APPENDIX

Ref.†	Location	Soil classification‡	Texture/other properties	Drainage§	Crop/treatment	Fertilizer type¶	N-Appl. rate (kg N/ha)	N_2O loss (kg N/ha)	Length of exp. (days)	Method#	Freq.††	Fertilizer induced N_2O loss (% of N-appl.)‡‡ I	Fertilizer induced N_2O loss (% of N-appl.)‡‡ II	Remarks
MINERAL SOILS														
1	Reading, UK		loamy sand	w	unplanted	NO_3	200	0.30	135	c -	d		0.2	weed free
1	Reading, UK		clay loam	w	unplanted	NO_3	200	0.22	135	c -	d		0.1	weed free
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass	NH_4	117	0.56	63	c -	d/w		0.5	intensive management
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass	NH_4	82	0.10	63	c -	d/w		0.1	intensive management
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass		0	0.13	105	c -	d/w			intensive management
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass	NH_4	112	0.23	63	c -	d/w		0.2	intensive management
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass	NH_4	52	0.35	63	c -	d/w		0.7	intensive management; also presented in Hutchinson & Brams (1992)
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass		0	0.30	63	c -	d/w			low management
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass		0	0.07	63	c -	d/w			low management
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass		0	0.08	105	c -	d/w			low management
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass	NH_4	112	0.24	63	c -	d/w		0.2	low management
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass		0	0.20	63	c -	d/w			low management; also presented in Hutchinson & Brams (1992)
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass	NH_4'	363	1.37	365	c -	d/w		0.4	intensive management; sum 365 days
2	Texas, USA	glossarenic Paleudalfs	sandy loam, 1.7 %C	w	grass	NH_4	112	0.89	365	c -	d/w		0.8	low management; sum 365 days
3	Iowa, USA	typic Calciaquolls	clay loam; 4.9% C	p	unplanted		0	0.33	96	c -	3-7 d			
3	Iowa, USA	typic Calciaquolls	clay loam; 4.9% C	p	unplanted	urea	125	0.50	96	c -	3-7 d	0.1	0.4	
3	Iowa, USA	typic Calciaquolls	clay loam; 4.9% C	p	unplanted	urea	250	0.62	96	c -	3-7 d	0.1	0.2	
3	Iowa, USA	typic Calciaquolls	clay loam; 4.9% C	p	unplanted	NH_4	125	0.56	96	c -	3-7 d	0.2	0.4	
3	Iowa, USA	typic Calciaquolls	clay loam; 4.9% C	p	unplanted	NH_4	250	0.61	96	c -	3-7 d	0.1	0.2	
3	Iowa, USA	typic Calciaquolls	clay loam; 4.9% C	p	unplanted	NO_3	125	0.38	96	c -	3-7 d	0.0	0.3	
3	Iowa, USA	typic Calciaquolls	clay loam; 4.9% C	p	unplanted	NO_3	250	0.36	96	c -	3-7 d	0.0	0.1	
4	Iowa, USA	typic Haplaquolls	loam; 2.5%C, pH 7.7	p	unplanted		0	0.65	140	c -	3-7 d			
4	Iowa, USA	typic Haplaquolls	loam; 2.5%C, pH 7.7	p	unplanted	NH_3	180	4.40	140	c -	3-7 d	2.1	2.4	AA injected at 20 cm
4	Iowa, USA	typic Haplaquolls	loam; 2.5%C, pH 7.7	p	unplanted	NH_4 (aq. ammonia)	180	0.86	140	c -	3-7 d	0.1	0.5	
4	Iowa, USA	typic Haplaquolls	loam; 2.5%C, pH 7.7	p	unplanted	urea	180	0.77	140	c -	3-7 d	0.1	0.4	
4	Iowa, USA	typic Calciaquolls	silty clay loam; 4.6%C, pH 7.9	p	unplanted		0	0.38	140	c -	3-7 d			
4	Iowa, USA	typic Calciaquolls	silty clay loam; 4.6%C, pH 7.9	p	unplanted	NH_3	180	1.92	140	c -	3-7 d	0.9	1.1	AA injected at 20 cm
4	Iowa, USA	typic Calciaquolls	silty clay loam; 4.6%C, pH 7.9	p	unplanted	NH_4 (aq. ammonia)	180	0.45	140	c -	3-7 d	0.0	0.3	
4	Iowa, USA	typic Calciaquolls	silty clay loam; 4.6%C, pH 7.9	p	unplanted	urea	180	0.57	140	c -	3-7 d	0.1	0.3	
4	Iowa, USA	typic Calciaquolls	silty clay loam; 4.6%C, pH 7.9	p	unplanted	NO_3	180	0.44	140	c -	3-7 d	0.1	0.2	
4	Iowa, USA	typic Haplaquolls	clay loam; 2.7%C, pH 6.9	p	unplanted		0	0.51	140	c -	3-7 d			
4	Iowa, USA	typic Haplaquolls	clay loam; 2.7%C, pH 6.9	p	unplanted	NH_3	180	2.17	140	c -	3-7 d	0.9	1.2	AA injected at 20 cm
4	Iowa, USA	typic Haplaquolls	clay loam; 2.7%C, pH 6.9	p	unplanted	NH_4 (aq. ammonia)	180	0.62	140	c -	3-7 d	0.1	0.3	
4	Iowa, USA	typic Haplaquolls	clay loam; 2.7%C, pH 6.9	p	unplanted	urea	180	0.64	140	c -	3-7 d	0.1	0.4	
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated		0	0.45	116	c -	3-7 d			
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	75	1.67	116	c -	3-7 d	1.6	2.2	AA injected at 20 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	150	2.58	116	c -	3-7 d	1.4	1.7	AA injected at 20 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	225	3.17	116	c -	3-7 d	1.2	1.4	AA injected at 20 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	300	3.75	116	c -	3-7 d	1.1	1.3	AA injected at 20 cm

APPENDIX

Ref.†	Location	Soil classification‡	Texture/other properties	Drainage§	Crop/treatment	Fertilizer type¶	N-Appl. rate (kg N/ha)	N_2O loss (kg N/ha)	Length of exp. (days)	Method#	Freq.††	Fertilizer induced N_2O loss (% of N-appl.)‡‡ I	II	Remarks
MINERAL SOILS														
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	375	4.26	116	c -	3-7 d	1.0	1.1	AA injected at 20 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	450	4.54	116	c -	3-7 d	0.9	1.0	AA injected at 20 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated		0	0.71	156	c -	3-7 d			
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	112	1.52	156	c -	3-7 d	0.7	1.4	AA injected at 10 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	112	2.10	156	c -	3-7 d	1.2	1.9	AA injected at 20 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	112	2.39	156	c -	3-7 d	1.5	2.1	AA injected at 30 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	225	2.82	156	c -	3-7 d	0.9	1.3	AA injected at 10 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	225	3.25	156	c -	3-7 d	1.1	1.4	AA injected at 20 cm
5	Iowa, USA	typic Haplaquolls	clay loam; 3.8% C, pH 6.9	p	unplanted; maize residues incorporated	NH_3	225	3.44	156	c -	3-7 d	1.2	1.5	AA injected at 30 cm
6	Iowa, USA	typic Calciaquolls	silty clay loam; 4.6%C, pH 7.9	p	unplanted; soybean plants left to decompose		0	1.70	139	c -	w			
6	Iowa, USA	typic Calciaquolls	silty clay loam; 4.6%C, pH 7.9	p	unplanted; soybean plants left to decompose	NH_3	250	15.00	139	c -	w	5.3	6.0	AA injected at 20 cm
6	Iowa, USA	typic Haplaquolls	clay loam; 2.7%C, pH 6.9	p	unplanted; soybean plants left to decompose		0	2.50	139	c -	w			
6	Iowa, USA	typic Haplaquolls	clay loam; 2.7%C, pH 6.9	p	unplanted; soybean plants left to decompose	NH_3	250	19.60	139	c -	w	6.8	7.8	AA injected at 20 cm
6	Iowa, USA	typic Haplaquolls	loam; 2.5%C, pH 7.7	p	unplanted; soybean plants left to decompose		0	2.00	139	c -	w			
6	Iowa, USA	typic Haplaquolls	loam; 2.5%C, pH 7.7	p	unplanted; soybean plants left to decompose	NH_3	250	12.10	139	c -	w	4.0	4.8	AA injected at 20 cm
7	Iowa, USA	typic Haplaquolls	clay loam; 3.7% C, pH 6.8	p	unplanted; maize residues incorporated		0	0.62	355	c -	3-7 d			
7	Iowa, USA	typic Haplaquolls	clay loam; 3.7% C, pH 6.8	p	unplanted; maize residues incorporated	NH_3	180	3.62	355	c -	3-7 d	1.7	2.0	AA injected at 18 cm in fall
7	Iowa, USA	typic Haplaquolls	clay loam; 3.7% C, pH 6.8	p	unplanted; maize residues incorporated		0	0.43	167	c -	3-7 d			
7	Iowa, USA	typic Haplaquolls	clay loam; 3.7% C, pH 6.8	p	unplanted; maize residues incorporated	NH_3	180	1.37	167	c -	3-7 d	0.5	0.8	AA injected at 18 cm in spring
8	Oxon, UK	typic Haplaquepts	clay, 3.2-3.9%C	p	wheat, winter, ploughed	NH_4NO_3	70	0.90	212	c -	w		1.3	Nov.'77-June '78
8	Oxon, UK	typic Haplaquepts	clay, 3.2-3.9%C	p	wheat, winter, direct drilled	NH_4NO_3	70	5.40	212	c -	w		7.7	Nov.'77-June '78
8	Oxon, UK	typic Haplaquepts	clay, 3.2-3.9%C	p	oilseed rape, ploughed	NH_4NO_3	140	5.60	212	c -	w		4.0	Nov.'78-June '79
8	Oxon, UK	typic Haplaquepts	clay, 3.2-3.9%C	p	oilseed rape, direct drilled	NH_4NO_3	140	8.60	212	c -	w		6.1	Nov.'78-June '79
8	Oxon, UK	typic Haplaquepts	clay loam, 2-2.1% C	p	wheat, winter, ploughed	NH_4NO_3	70	0.50	212	c -	w		0.7	Nov.'77-June '78
8	Oxon, UK	typic Haplaquepts	clay loam, 2-1.2%C	p	wheat, winter, direct drilled	NH_4NO_3	70	1.50	212	c -	w		2.1	Nov.'77-June '78
8	Oxon, UK	typic Haplaquepts	clay loam, 2-2.1% C	p	oilseed rape, ploughed	NH_4NO_3	140	1.00	212	c -	w		0.7	Nov.'78-June '79
8	Oxon, UK	typic Haplaquepts	clay loam, 2-1.2%C	p	oilseed rape, direct drilled	NH_4NO_3	140	2.10	212	c -	w		1.5	Nov.'78-June '79
9	Madison, USA	typic Hapludalfs		w	maize	organic/NH_4NO_3/urea	237	5.20	365	c -	7-30 d	2.1	2.2	168/13/56 manure/NH_4NO_3/urea; prev. maize residues incorporated
9	Madison, USA	typic Hapludalfs		w	maize	organic/NH_4NO_3	181	3.60	365	c -	7-30 d	1.8	2.0	168/13/56 manure/NH_4NO_3/urea; prev. maize residues incorporated
9	Madison, USA	typic Hapludalfs		w	grass		0	0.34	365	c -	7-30 d			
10	Washinghton,USA	ultic Haploxerolls	silt loam	w	unplanted		0	0.03	35	o -	2 d			24 hour cont. measurement per 2 days
10	Washinghton,USA	ultic Haploxerolls	silt loam	w	unplanted	NH_3	55	0.05	35	o -	2 d	0.0	0.1	AA injected at 15 cm
10	Washinghton,USA	ultic Haploxerolls	silt loam	w	unplanted	NH_3	110	0.10	35	o -	2 d	0.1	0.1	AA injected at 15 cm
10	Washinghton,USA	ultic Haploxerolls	silt loam	w	unplanted	NH_3	220	0.23	35	o -	2 d	0.1	0.1	AA injected at 15 cm
11	Oxon, UK	typic Haplaquepts	clay, undrained	p	wheat, winter, direct drilled	NH_4NO_3	53	0.65	30	c 1	w		1.2	
11	Oxon, UK	typic Haplaquepts	clay, undrained	p	wheat, winter, direct drilled		0	0.07	28	c 1	w			
11	Oxon, UK	typic Haplaquepts	clay, undrained	p	wheat, winter, direct drilled	NO_3	100	3.12	31	c 1	w		3.1	
11	Oxon, UK	typic Haplaquepts	clay, undrained	p	wheat, winter, direct drilled		0	0.07	28	c 1	w			
11	Oxon, UK	typic Haplaquepts	clay, drained	m	wheat, winter, direct drilled	NH_4NO_3	96	1.44	30	c 1	w		1.5	
11	Oxon, UK	typic Haplaquepts	clay, drained	m	wheat, winter, direct drilled	NH_4NO_3	53	0.22	31	c 1	w		0.4	

APPENDIX

Ref.†	Location	Soil classification‡	Texture/other properties	Drainage§	Crop/treatment	Fertilizer type¶	N-Appl. rate	N_2O loss	Length of exp.	Me-thod#	Freq.††	Fertilizer induced N_2O loss (% of N-appl.)‡‡		Remarks
							(kg N/ha)		(days)			I	II	
MINERAL SOILS														
11	Oxon, UK	typic Haplaquepts	clay, drained	m	wheat, winter, direct drilled		0	1.49	31	c 1	w			
11	Oxon, UK	typic Haplaquepts	clay, drained	m	wheat, winter, direct drilled		0	0.07	30	c 1	w			
12	Oxon, UK	typic Haplaquepts	clay, undrained	p	wheat, winter, direct drilled	NH_4NO_3/NO_3	153	2.30	57	c 1	2-3 d		1.5	
13	Oxon, UK	typic Haplaquepts	clay, 3.7% C	p	wheat, winter, ploughed	NH_4NO_3	70	1.10	242	c 1	w		1.6	
13	Oxon, UK	typic Haplaquepts	clay, 3.7% C	p	wheat, winter, direct drilled	NH_4NO_3	70	1.30	242	c 1	w		1.9	
13	Oxon, UK	typic Haplaquepts	clay, 3.7% C	p	grass	NH_4NO_3	210	0.40	242	c 1	w		0.2	
14	Mainz, Germany	loess, pararendzina	sandy clay loam	w	grass	NO_3	100	0.07		c -	d	0.1		
14	Mainz, Germany	loess, pararendzina	sandy clay loam	w	grass	NH_4	100	0.05		c -	d	0.1		
14	Mainz, Germany	loess, brown soil	sandy loam	w	unplanted (beet field, plants removed)	NO_3	100	0.02		c -	d	0.0		
14	Mainz, Germany	loess, brown soil	sandy loam	w	unplanted (beet field, plants removed)	NH_4	100	0.15		c -	d	0.2		
14	Mainz, Germany	loess, brown soil	sandy loam	w	unplanted (beet field, plants removed)	NH_4	100	0.22		c -	d	0.2		
14	Mainz, Germany	loess	sandy clay loam	w	grass	NO_3	100	0.01		c -	d	0.0		
14	Mainz, Germany	loess	sandy clay loam	w	grass	NH_4	100	0.03		c -	d	0.0		
14	Mainz, Germany	loess	sandy clay loam	w	grass	NO_3	100	0.07		c -	d	0.1		
14	Mainz, Germany	loess	sandy clay loam	w	grass	NH_4	100	0.38		c -	d	0.4		
14	Mainz, Germany	loess	sandy clay loam	w	clover	NO_3	100	0.00		c -	d	0.0		
14	Mainz, Germany	loess	sandy clay loam	w	clover	NH_4	100	0.07		c -	d	0.1		
14	Mainz, Germany	loess	sandy loam	w	grass	NO_3	100	0.02		c -	d	0.0		
14	Mainz, Germany	loess	sandy loam	w	grass	NH_4	100	0.07		c -	d	0.1		
14	Mainz, Germany	loess	sandy loam	w	grass	NH_4	100	0.08		c -	d	0.1		
15	Australia		clay		rice, wetland	NO_3	40	0.38	18	o -	cont		1.0	
16	New York, USA	glossoboric Hapludalfs	silt loam, 1% C, pH 6.9	w	maize		0	0.30	85	c 1	d			
16	New York, USA	glossoboric Hapludalfs	silt loam, 1% C, pH 6.9	w	maize	NO_3	140	0.30	85	c 1	d	0.0	0.2	starter dosis 20 NH_4NO_3
16	New York, USA	glossoboric Hapludalfs	silt loam, 1% C, pH 6.9	w	maize	urea	140	2.50	85	c 1	d	1.6	1.8	starter dosis 20 NH_4NO_3
17	New York, USA	glossoboric Hapludalfs	silt loam	w	maize	organic/NH_4NO_3	132	2.40	365	c -	d		1.8	112/20 organic/NH_4NO_3; 1979/80
17	New York, USA	glossoboric Hapludalfs	silt loam	w	maize	organic/NH_4NO_3	132	2.90	365	c -	d		2.2	112/20 organic/NH_4NO_3; 1980/81
17	New York, USA	glossoboric Hapludalfs	silt loam	w	maize	organic/NH_4NO_3	132	3.80	365	c -	d		2.9	112/20 organic/NH_4NO_3; 1980/81
17	New York, USA	glossoboric Hapludalfs	silt loam	w	timothy weeds		0	0.90	365	c -	d			1980/81
17	New York, USA	glossoboric Hapludalfs	silt loam	w	timothy weeds		0	1.70	365	c -	d			1979/80
17	New York, USA	glossoboric Hapludalfs	silt loam	w	maize	NH_4NO_3/urea	132	1.60	365	c -	d		1.2	1979/80
17	New York, USA	glossoboric Hapludalfs	silt loam	w	maize	NH_4NO_3/urea	132	2.90	365	c -	d		2.2	1980/81
17	New York, USA	glossoboric Hapludalfs	silt loam	w	maize	NH_4NO_3/urea	132	2.20	365	c -	d		1.7	1980/81
17	New York, USA	glossoboric Hapludalfs	silt loam	w	alfalfa		0	2.30	365	c -	d			1980/81
17	New York, USA	glossoboric Hapludalfs	silt loam	w	alfalfa		0	4.20	365	c -	d			1979/80
18	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass	organic	1230	3.25	314	g	d/w		0.3	1978-1979
18	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass		0	0.45	273	g	d/w			1979-1980
18	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass	NO_3	400	1.25	273	g	d/w		0.3	1979-1980
18	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass	organic	298	1.10	273	g	d/w		0.4	1979-1980
18	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass		0	3.55	314	g	d/w			1978-1979
18	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass	NO_3	100	6.90	314	g	d/w		6.9	1978-1979
19	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass	NO_3	700	13.40	365	o 2	2-3 d/w		1.9	
19	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass	organic	700	3.30	365	o 2	2-3 d/w		0.5	
19	Edinburgh, UK	stagnogley, 4.1% C	sandy loam over clay loam	p	grass		0	3.20	365	o 2	2-3 d/w			

APPENDIX

Ref.†	Location	Soil classification‡	Texture/other properties	Drainage§	Crop/treatment	Fertilizer type¶	N-Appl. rate	N_2O loss	Length of exp.	Method#	Freq.††	Fertilizer induced N_2O loss (% of N-appl.)‡‡		Remarks
							(kg N/ha)		(days)			I	II	
MINERAL SOILS														
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.16% C	w	tobacco	NH_4NO_3/straw	80	0.70	253	c -	w		0.9	80 kg N from NH_4NO_3; unknown amount of N from straw incorporated; 1981
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.27% C	w	tobacco	NH_4NO_3/straw	80	0.30	190	c -	w		0.4	80 kg N from NH_4NO_3; unknown amount of N from incorporated cover crop (rye); 1981
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.31% C	w	tobacco	organic/NH_4NO_3	245	0.30	190	c -	w		0.1	80 NH_4NO_3 + 165 kg N/ha from alfalfa; 1981
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.72% C	w	tobacco	organic/NH_4NO_3	410	2.70	252	c -	w		0.7	80 NH_4NO_3 + 330 kg N/ha from manure; 1981
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 6.7; 1.61% C	w	tobacco	NH_4NO_3	80	1.00	210	c -	w		1.3	1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 5.1; 1.56% C	w	tobacco	NH_4NO_3	80	0.90	210	c -	w		1.1	1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 1.56% C	w	tobacco	NH_4NO_3	80	1.50	206	c -	w		1.9	1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.16% C	w	tobacco	NH_4NO_3/straw	80	2.20	249	c -	w		2.8	80 kg N from NH_4NO_3; unknown amount of N from straw incorporated; 1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.27% C	w	tobacco	NH_4NO_3/straw	80	1.60	202	c -	w		2.0	80 kg N from NH_4NO_3; unknown amount of N from incorporated cover crop (rye); 1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.31% C	w	tobacco	organic/NH_4NO_3	245	3.20	202	c -	w		1.3	80 NH_4NO_3 + 165 kg N/ha from alfalfa; 1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.72% C	w	tobacco	organic/NH_4NO_3	410	6.10	257	c -	w		1.5	80 NH_4NO_3 + 330 kg N/ha from manure; 1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 5.8; 2.72% C	w	barley	organic/NH_4NO_3	520	1.60	215	c -	w		0.3	80 NH_4NO_3 + 440 kg N/ha from sludge; 1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 6.8; 1.74% C	w	maize	NH_4NO_3	200	6.30	190	c -	w		3.2	reduced tillage; 1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 6.8; 1.74% C	w	maize	NH_4NO_3	200	3.50	190	c -	w		1.8	reduced tillage; 1980
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 6.7; 1.56% C	w	vegetables	NH_4NO_3	80	0.20	160	c -	w		0.3	1979
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 5.1; 1.56% C	w	vegetables	NH_4NO_3	80	0.20	160	c -	w		0.3	1979
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 1.56% C	w	vegetables	NH_4NO_3	80	0.40	160	c -	w		0.5	1979
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 4.7; 2.72% C	w	vegetables	organic/NH_4NO_3	410	1.20	160	c -	w		0.3	80 NH_4NO_3 + 330 kg N/ha from manure; 1979
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 5.8; 2.72% C	w	barley	organic/NH_4NO_3	520	0.20	152	c -	w		0.0	80 NH_4NO_3 + 440 kg N/ha from sludge; 1979
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 6.8; 1.81% C	w	maize	NH_4NO_3	200	0.30	157	c -	w		0.2	reduced tillage; 1979
20	Wisconsin, USA	typic Argiudolls	silt loam; pH 6.8; 1.74% C	w	maize	NH_4NO_3	200	0.60	157	c -	w		0.3	reduced tillage; 1979
21	Colorado, USA	aridic Argiustoll	clay (montmorillonitic)	w	maize	NH_3	200	2.60	128	c -/m	w		1.3	irrigated maize
21	Colorado, USA	aridic Argiustoll	clay (montmorillonitic)	w	maize	NH_3	200	4.00	365	c -/m	w		2.0	irrigated maize; flux based on extrapolation
22	Ontario, Canada	gray brown Luvisol	sandy loam	w	maize		0	0.10	80	c -	w			Estimated from Figure 1, p. 434
22	Ontario, Canada	gray brown Luvisol	sandy loam	w	maize	NH_4NO_3	336	0.85	80	c -	w	0.2	0.3	About 3 measurements/month
23	Konosu, Japan	alluvial soil		w	rape	NH_4	150	0.09	38	c -	2 h		0.1	Fig. 4 (p. 24) shows 2 h intervals of measurement

APPENDIX

Ref.†	Location	Soil classification‡	Texture/other properties	Drainage§	Crop/treatment	Fertilizer type¶	N-Appl. rate	N_2O loss	Length of exp. (days)	Me-thod#	Freq.††	Fertilizer induced N_2O loss (% of N-appl.)‡‡		Remarks
							(kg N/ha)					I	II	
MINERAL SOILS														
23	Konosu, Japan	alluvial soil		w	wheat	NH_4	80	0.14	186	c -	2 h		0.2	
23	Tsukuba, Japan	andosols		w	wheat	NH_4	80	0.19	186	c -	2 h		0.2	
23	Tsukuba, Japan	andosols		w	rape	NH_4	100	0.34	56	c -	2 h		0.3	
23	Tsukuba, Japan	andosols		w	rape	NH_4	150	0.14	38	c -	2 h		0.1	
23	Tsukuba, Japan	andosols		w	carrot	NH_4	200	0.52	116	c -	2 h		0.3	
23	Konosu, Japan	alluvial soil		w	carrot	NH_4	200	0.62	116	c -	2 h		0.3	
23	Konosu, Japan	alluvial soil		w	rice, dryland	NH_4	100	0.33	120	c -	2 h		0.3	
23	Tsukuba, Japan	andosols		w	rice, dryland	NH_4	100	0.55	120	c -	2 h		0.6	
23	Mito, Japan	andosols		w	rice, dryland	NH_4	90	0.27	139	c -	2 h		0.3	
24	Tsukuba, Japan	gray lowland soil, 2.5% C		p	carrot	NH_4	200	0.25	116	c -	3-10 d	0.1	0.1	nitrification inhibitor added
24	Tsukuba, Japan	gray lowland soil, 2.5% C		p	carrot	NH_4	200	0.60	116	c -	3-10 d	0.3	0.3	
24	Tsukuba, Japan	gray lowland soil, 2.5% C		p	carrot		0	0.08	116	c -	3-10 d			
24	Tsukuba, Japan	gray lowland soil, 2.5% C		p	carrot	NH_4	200	0.34	116	c -	3-10 d	0.1	0.2	
25	Colorado, USA	aridic Argiustolls	clay (montmorillonitic)	w	maize	NH_3	200	2.50	123	c -/m	d/w		1.3	irrigated maize; AA injected
26	Colorado, USA	ustic Torriorthents		w	barley	organic (N-mineralized)	356	4.19	153	c -	3 d/w	1.0	1.2	total N added is 1436 kg
26	Colorado, USA	ustic Torriorthents		w	barley	organic (N-mineralized)	71	1.09	153	c -	3 d/w	0.8	1.5	total N added is 287 kg
26	Colorado, USA	ustic Torriorthents		w	barley		0	0.82	153	c -	3 d/w			N-mineralized is 69 kg
26	Colorado, USA	ustic Torriorthents		w	barley	NH_4NO_3	224	1.43	153	c -	3 d/w	0.4	0.6	
26	Colorado, USA	ustic Torriorthents		w	barley	NH_4NO_3	112	1.04	153	c -	3 d/w	0.5	0.9	
26	Colorado, USA	ustic Torriorthents		w	barley	NH_4NO_3	56	0.93	153	c -	3 d/w	0.7	1.7	
26	Colorado, USA	ustic Torriorthents		w	barley		0	0.52	153	c -	3 d/w			
27	Colorado, USA	aridic Argiustolls	clay loam	w	maize		0	2.23	120	c 2	2-4 p.d/3 p.w			irrigated maize
27	Colorado, USA	aridic Argiustolls	clay loam	w	maize	NH_4	200	2.95	120	c 2	2-4 p.d/3 p.w		1.5	irrigated maize
27	Colorado, USA	aridic Argiustolls	clay loam	w	barley		0	0.45	86	c 2	2-4 p.d/3 p.w			irrigated barley
27	Colorado, USA	aridic Argiustolls	clay loam	w	barley	NH_4	200	0.76	86	c 2	2-4 p.d/3 p.w		0.4	irrigated barley
28	California, USA	typic Xerorthents	loam	w	unplanted	NO_3/organic	300	9.90	16	c 2	d/w		3.3	unknown amount of N from organic fert.; controlled soil moisture; summer exp.
28	California, USA	typic Xerorthents	loam	w	unplanted	NO_3/organic	300	5.40	16	c 2	d/w		1.8	unknown amount of N from organic fert.; controlled soil moisture; summer exp.
28	California, USA	typic Xerorthents	loam	w	unplanted (ryegrass 4 months before exp.)	NO_3	300	4.30	16	c 2	d/w		1.4	controlled soil moisture; summer exp.
28	California, USA	typic Xerorthents	loam	w	unplanted (ryegrass 4 months before exp.)	NO_3	300	1.80	16	c 2	d/w		0.6	controlled soil moisture; summer exp.
28	California, USA	typic Xerorthents	loam	w	unplanted	NO_3	300	2.10	16	c 2	d/w		0.7	controlled soil moisture; summer exp.
28	California, USA	typic Xerorthents	loam	w	unplanted	NO_3	300	0.60	16	c 2	d/w		0.2	controlled soil moisture; summer exp.
28	California, USA	typic Xerorthents	loam	w	unplanted	NO_3/organic	300	4.20	16	c 2	d/w		1.4	unknown amount of N from organic fert.; controlled soil moisture; winter exp.

APPENDIX

Ref.†	Location	Soil classification‡	Texture/other properties	Drainage§	Crop/treatment	Fertilizer type¶	N-Appl. rate (kg N/ha)	N_2O loss (kg N/ha)	Length of exp. (days)	Method#	Freq.††	Fertilizer induced N_2O loss (% of N-appl.)‡‡ I	II	Remarks
MINERAL SOILS														
28	California, USA	typic Xerorthents	loam	w	unplanted	NO_3/organic	300	3.80	16	c 2	d/w		1.3	unknown amount of N from organic fert.; controlled soil moisture; winter exp.
28	California, USA	typic Xerorthents	loam	w	unplanted (ryegrass 4 months before exp.)	NO_3	300	0.70	16	c 2	d/w		0.2	controlled soil moisture; winter exp.
28	California, USA	typic Xerorthents	loam	w	unplanted (ryegrass 4 months before exp.)	NO_3	300	0.10	16	c 2	d/w		0.0	controlled soil moisture; winter exp.
28	California, USA	typic Xerorthents	loam	w	unplanted	NO_3	300	0.40	16	c 2	d/w		0.1	controlled soil moisture; winter exp.
28	California, USA	typic Xerorthents	loam	w	unplanted	NO_3	300	0.20	16	c 2	d/w		0.1	controlled soil moisture; winter exp.
29	Berkshire, UK	Ochraqualfs	loam over clay; 3.5 % C	p	grass	NH_4NO_3	250	3.25	365	o -	d/3 p.w		1.3	
30	Berkshire, UK	Ochraqualfs	loam over clay; 3.5 % C	p	grass		0	-0.60	365	o 1	2-3 p.w.			
30	Berkshire, UK	Ochraqualfs	loam over clay; 3.5 % C	p	grass	NH_4NO_3	500	8.00	365	o 1	2-3 p.w.		1.6	
30	Berkshire, UK	Ochraqualfs	loam over clay; 3.5 % C	p	grass	NH_4NO_3	250	3.50	365	o 1	2-3 p.w.		1.4	
31	California, USA	pachic Haploxerolls	fine loamy	w	vegetables	NO_3	620	41.80	210	o 1	d/2-3 d		6.7	lettuce-celery, irrigated
31	California, USA	pachic Haploxerolls	fine loamy	w	vegetables	NO_3	620	20.20	210	o 1	d/2-3 d		3.3	lettuce-celery, irrigated
31	California, USA	pachic Haploxerolls	fine loamy	w	vegetables	NO_3	620	26.40	210	o 1	d/2-3 d		4.3	lettuce-celery, irrigated
31	California, USA	pachic Haploxerolls	fine loamy	w	vegetables	organic/NO_3	430	19.60	210	o 1	d/2-3 d		4.6	144/286 organic/NO_3; artichokes, irrigated
31	California, USA	pachic Haploxerolls	fine loamy	w	vegetables	organic/NO_3	430	26.90	210	o 1	d/2-3 d		6.3	144/286 organic/NO_3; artichokes, irrigated
31	California, USA	pachic Haploxerolls	fine loamy	w	vegetables	NO_3	680	26.80	210	o 1	d/2-3 d		3.9	cauliflower, irrigated
31	California, USA	pachic Haploxerolls	fine loamy	w	vegetables	NO_3	680	29.20	210	o 1	d/2-3 d		4.3	cauliflower, irrigated
32	California, USA	pachic Haploxerolls	fine loamy	w	vegetables	NH_4/urea/NH_3	335	7.68	123	o 1	d/w		2.3	celery, irrigated; 12-18% of denitrification of 51.2 kg N/ha as N2O (p.117)
33	Mainz, Germany	loess loam	sandy l.-clay loam, 0.8% C, pH 7.4	w	grass		0	0.02	49	c -	d			
33	Mainz, Germany	loess loam	sandy l.-clay loam, 0.8% C, pH 7.4	w	grass	NO_3	100	0.07	49	c -	d	0.05	0.1	
33	Mainz, Germany	loess loam	sandy l.-clay loam, 0.8% C, pH 7.4	w	grass	NH_4	100	0.09	49	c -	d	0.07	0.1	
33	Mainz, Germany	eolian sand	sand	w	weeds		0	0.13	71	c -	d			
33	Mainz, Germany	eolian sand	sand	w	weeds	NO_3	100	0.14	71	c -	d	0.01	0.1	
33	Mainz, Germany	eolian sand	sand	w	weeds	NH_4	100	0.22	71	c -	d	0.09	0.2	
33	Mainz, Germany	loess	sandy loam, 2-2.6% C	w	grass		0	0.02	32	c -	d			authors refer to crop as "meadow"
33	Mainz, Germany	loess	sandy loam, 2-2.6% C	w	grass	NO_3	100	0.03	32	c -	d	0.01	0.0	authors refer to crop as "meadow"
33	Mainz, Germany	loess	sandy loam, 2-2.6% C	w	grass	NH_4	100	0.05	32	c -	d	0.03	0.0	authors refer to crop as "meadow"
34	Mainz, Germany	eolian sand	sand	w	weeds		0	0.04	72	c -	2d/m			estimated from Figure 2, p.556
34	Mainz, Germany	eolian sand	sand	w	weeds	NH_4	100	0.13	72	c -	2d/m	0.1	0.1	
34	Mainz, Germany	eolian sand	sand	w	weeds	NO_3	100	0.05	72	c -	2d/m	0.0	0.1	
34	Mainz, Germany	eolian sand	sand	w	weeds	NH_4NO_3	100	0.09	72	c -	2d/m	0.1	0.1	
35	Andalusia, Spain		loamy sand	w	grass		0	0.00	10	c -	d			estimated from Figure 2, p. 165
35	Andalusia, Spain		loamy sand	w	grass	NH_4NO_3	100	0.08	10	c -	d	0.1	0.1	estimated from reported % N2O loss; plot had received additional 75 kg N earlier in the year
35	Andalusia, Spain		loamy sand	w	unplanted, soybean residues incorporated		0	0.10	28	c -	d			estimated from 15x10^-6 g N2O-N loss/m^2/h.

APPENDIX

Ref.†	Location	Soil classification‡	Texture/other properties	Drainage§	Crop/treatment	Fertilizer type¶	N-Appl. rate	N_2O loss	Length of exp.	Me-thod#	Freq.††	Fertilizer induced N_2O loss (% of N-appl.)‡‡		Remarks
							(kg N/ha)	(kg N/ha)	(days)			I	II	
MINERAL SOILS														
35	Andalusia, Spain		loamy sand	w	unplanted, soybean residues incorporated	NH_4NO_3	100	0.14	28	c -	d	0.0	0.1	estimated from reported 0.04 % N2O loss from NH_4NO_3
35	Andalusia, Spain		loamy sand	w	unplanted, soybean residues incorporated	urea	100	0.28	28	c -	d	0.2	0.3	estimated from reported 0.18 % N2O loss from urea
36	Louisiana, USA	typic Albaqualfs	silt loam; 0.7% C; pH 6	p	rice, wetland		0	0.07	105	c -	w			
36	Louisiana, USA	typic Albaqualfs	silt loam; 0.7% C; pH 6	p	rice, wetland	urea	90	0.11	105	c -	w	0.0	0.1	urea drilled
36	Louisiana, USA	typic Albaqualfs	silt loam; 0.7% C; pH 6	p	rice, wetland	urea	180	0.17	105	c -	w	0.1	0.1	urea drilled
36	Louisiana, USA	typic Albaqualfs	silt loam; 0.7% C; pH 6	p	rice, wetland	urea	90	0.11	105	c -	w	0.0	0.1	urea top dressed
36	Louisiana, USA	typic Albaqualfs	silt loam; 0.7% C; pH 6	p	rice, wetland	urea	180	0.09	105	c -	w	0.0	0.1	urea top dressed
37	UK		clay loam, 4% C	p	grass	NO_3	400	7.00	365	c -	2 p.d/w	1.5	1.8	mean of reported emission of 6.0-8.0 kg N/ha
37	UK		silt loam, 2.3% C	w	grass	NO_3	400	5.00	365	c -	2 p.d/w	1.0	1.3	mean of reported emission of 4.0-6.0 kg N/ha
37	UK		silt loam, 2.3% C	w	grass		0	0.90	365	c -	2 p.d/w			mean of reported emission of 0.8-1.0 kg N/ha
38	Denmark		sandy loam, 1.9%C, pH 5.3	w	grass	NH_4NO_3	200	2.38	100	o -	h	0.9	1.2	two applications
38	Denmark		sandy loam, 1.9%C, pH 5.3	w	grass	organic	492	9.35	100	o -	h	1.8	1.9	two applications; cow slurry, 50% of N inorganic
38	Denmark		sandy loam, 1.9%C, pH 5.3	w	grass		0	0.67	100	o -	h			
39	Scotland		loam; pH 6.6; 4.4% C	w	barley, winter, ploughed	NH_4NO_3	210	0.01	8	c -	d		0.0	
39	Scotland		loam; pH 6.6; 4.4% C	w	barley, winter, ploughed	NH_4NO_3	285	0.01	8	c -	nk		0.0	
39	Scotland		loam; pH 6.6; 4.4% C	w	barley, winter, direct drilled	NH_4NO_3	210	0.01	8	c -	nk		0.0	
39	Scotland		loam; pH 6.6; 4.4% C	w	barley, winter, direct drilled	NH_4NO_3	285	0.01	8	c -	nk		0.0	
39	Scotland		loam; pH 6.5; 3.3% C	w	barley, winter, ploughed	NH_4NO_3	210	0.01	8	c -	nk		0.0	
39	Scotland		loam; pH 6.5; 3.3% C	w	barley, winter, ploughed	NH_4NO_3	285	0.00	8	c -	nk		0.0	
39	Scotland		loam; pH 6.5; 3.3% C	w	barley, winter, direct drilled	NH_4NO_3	210	0.01	8	c -	nk		0.0	
39	Scotland		loam; pH 6.5; 3.3% C	w	barley, winter, direct drilled	NH_4NO_3	285	0.01	8	c -	nk		0.0	
40	Colorado, USA	Aridic Argiustolls	clay loam, 1.1% C, pH 7.2	w	maize		0	0.12	97	c -	3 p.w			irrigated maize
40	Colorado, USA	Aridic Argiustolls	clay loam, 1.1% C, pH 7.2	w	maize	urea	218	3.36	97	c -	3 p.w	1.5	1.5	irrigated maize
40	Colorado, USA	Aridic Argiustolls	clay loam, 1.1% C, pH 7.2	w	maize		0	0.11	97	c -	3 p.w			irrigated maize
40	Colorado, USA	Aridic Argiustolls	clay loam, 1.1% C, pH 7.2	w	maize	urea	218	1.65	97	c -	3 p.w	0.7	0.8	irrigated maize
41	Iowa, USA		sandy loam, 0.9%C, pH 7.9	p	soybeans		0	0.34	365	c -	3d/21d			
41	Iowa, USA		silty clay loam, 5.4%C, pH 7.5	p	soybeans		0	0.65	365	c -	3d/21d			
41	Iowa, USA	typic Calciaquolls	clay loam, 3.6%C, pH 8.1	p	soybeans		0	1.35	365	c -	3d/21d			
41	Iowa, USA		sandy loam, 1.3%C, pH 6.7	p	soybeans		0	1.05	365	c -	3d/21d			
41	Iowa, USA	typic Haplaquolls	loam, 2.9%C, pH 6.9	p	soybeans		0	1.97	365	c -	3d/21d			
41	Iowa, USA		loam, 2.5%C, pH 6.5	p	soybeans		0	1.87	365	c -	3d/21d			
42	New York, USA				wheat		0	0.09	46	nk	nk			
42	New York, USA				wheat	NH_4NO_3	175	0.30	46	nk	nk	0.1	0.2	
42	New York, USA				wheat	NH_4NO_3	175	0.16	46	nk	nk	0.0	0.1	
44	Colorado, USA	ustollic Haplargids	fine sandy loam	w	grass	urine		0.14	62	c -	3d			
44	Colorado, USA	ustollic Haplargids	fine sandy loam	w	grass	urine	450	0.49	62	c -	3d	0.1	0.1	

APPENDIX

Ref.†	Location	Soil classification‡	Texture/other properties	Drainage§	Crop/treatment	Fertilizer type¶	N-Appl. rate (kg N/ha)	N_2O loss (kg N/ha)	Length of exp. (days)	Method#	Freq.††	Fertilizer induced N_2O loss (% of N-appl.)‡‡ I	II	Remarks
ORGANIC SOILS														
17	Florida, USA		organic	w	onions	NH_4NO_3	170	85.00	365	c -	d		50.0	
17	Florida, USA		organic	w	onions	NH_4NO_3	170	72.00	365	c -	d		42.4	
17	Florida, USA		organic	w	maize	NH_4NO_3	170	76.00	365	c -	d		44.7	
17	Florida, USA		organic	w	maize	NH_4NO_3	170	152.00	365	c -	d		89.4	
17	Florida, USA		organic	w	sugarcane		0	48.00	365	c -	d			
17	Florida, USA		organic	w	sugarcane		0	7.00	365	c -	d			
17	Florida, USA		organic	w	grass		0	97.00	365	c -	d			
17	Florida, USA		organic	w	grass		0	16.00	365	c -	d			
17	Florida, USA		organic	w	unplanted		0	165.00	365	c -	d			
17	Florida, USA		organic	w	unplanted		0	59.00	365	c -	d			
43	Florida, USA	euic lithic Medisaprists	organic	p	unplanted		0	164.90	365	c -	3d			estimated N-mineralization 1300 kg N/ha/yr
43	Florida, USA	euic lithic Medisaprists	organic	p	grass		0	96.80	365	c -	3d			estimated N-mineralization 1300 kg N/ha/yr
43	Florida, USA	euic lithic Medisaprists	organic	p	sugarcane		0	48.40	365	c -	3d			estimated N-mineralization 1300 kg N/ha/yr

† 1, Armstrong (1983); 2, Brams et al. (1990); 3, Breitenbeck et al. (1980); 4, Breitenbeck & Bremner (1986a); 5, Breitenbeck & Bremner (1986b); 6, Bremner et al. (1981a); 7, Bremner et al. (1981b); 8, Burford et al. (1981); 9, Cates & Keeney (1987); 10, Cochran et al. (1980); 11, Colbourn & Harper (1987); 12, Colbourn et al. (1984a); 13, Colbourn et al. (1984b); 14, Conrad et al. (1983); 15, Denmead et al. (1979); 16, Duxbury & McConnaughey (1986); 17, Duxbury et al. (1982); 18, Eggington & Smith (1986); 19, Eggington & Smith (1986); 20, Goodroad et al. (1984); 21, Hutchinson & Mosier (1979); 22, McKenney et al. (1980); 23, Minami (1987); 24, Minami (1990); 25, Mosier & Hutchinson (1981); 26, Mosier et al. (1982); 27, Mosier et al. (1986); 28, Rolston et al. (1978); 29, Ryden (1981); 30, Ryden (1983); 31, Ryden & Lund (1980); 32, Ryden et al. (1979); 33, Seiler & Conrad (1981); 34, Conrad & Seiler (1980); 35, Slemr et al. (1984); 36, Smith et al. (1982); 37, Webster & Dowdell (1982); 38, Christensen (1983); 39, Arah et al. (1991); 40, Bronson et al. (1992); 41, Bremner et al. (1980); 42, Duxbury (personal communication), quoted in Eichner (1990); 43, Terry et al. (1981); 44, Mosier et al. (1981).

‡ Reported soil classification according to USDA (1975) or general description.

§ w = well drained; m = moderately well drained; p = poorly drained.

¶ NH_3 = anhydrous ammonia; NH_4 = salts of ammonia; NO_3 = salts of nitrate; NH_4NO_3 = ammonium nitrate; organic = various forms of organic fertilizers.

c = closed chamber method; o = open chamber method; g = soil N_2O gradient method; m = micrometeorological method; - = only N_2O measured; 1 = N_2 and N_2O measured (C_2H_2 inhibition); 2 = ^{15}N labelling.

†† Freq. = frequency of sampling; d = once per day, w = once per week, m = once per month, 3-7 d = once per 3-7 days, 2 p.d or 2 p.w = twice per day/week, cont = continuous, d/w or other combinations indicate higher frequency at high and lower frequency at low flux rates.

‡‡ I = flux from fertilized plot minus flux from unfertilized control plot, presented as % of N-application.

II = flux from fertilized plot presented as % of N-application.

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Uncertainties in the global source distribution of nitrous oxide

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Chapter 4

UNCERTAINTIES IN THE GLOBAL SOURCE DISTRIBUTION OF NITROUS OXIDE

Abstract. Inventories with 1°x1° resolution were compiled of nitrous oxide (N_2O) emissions from fertilized arable land, animal excreta, post-clearing effects on soil emissions, fossil fuel and fuelwood combustion and industrial N_2O sources. For other sources of N_2O, including soils under natural vegetation, oceans and biomass burning, published inventories were used. From these inventories the annual N_2O emission was calculated for four broad latitudinal zones covering the globe. Uncertainties were assessed by comparing variants of inventories with source estimates inferred from inverse modeling techniques. Major uncertainties occur in the tropics, where small errors in both soil and oceanic emission estimates may have large repercussions for the zonal distributions. Although there may still be many poorly known and unidentified N_2O sources, the analysis has resulted in improved understanding of some sources i.e.: (i) the oceanic N_2O emission may be more important than assumed in recent global N_2O budgets, with a major portion stemming from the 30°-90°S zone; (ii) the N_2O emission from animal excreta forms a significant global source; (iii) most of the N_2O from arable lands and grasslands, including effects of synthetic fertilizers and animal excreta, comes from the northern hemisphere; accounting for only the synthetic-fertilizer effect on N_2O emission leads to an underestimation of the emission from arable lands; (iv) fossil fuel combustion and industrial N_2O sources are dominant in the 30°-90°N zone, while N_2O from fuelwood combustion is mainly produced in the 0°-30°N zone; (v) the estimation of enhanced N_2O soil emission following tropical forest clearing that has accounted for gradually declining N_2O fluxes, along with aging of the clearing leads to a global emission that is significant but lower than previous estimates; (vi) most of the N_2O from coastal marine and freshwater systems, and soil N_2O emission resulting from N deposition probably comes from the northern hemisphere.

4.1. INTRODUCTION

Despite its low current concentration of about 310 ppbv, nitrous oxide (N_2O) is regarded as one of the main greenhouse gases (Wang et al., 1976) because of its long atmospheric lifetime (~150 years; Khalil and Rasmussen, 1992) and its radiative forcing of about 200 times that of CO_2 (Shine et al., 1990). In the troposphere N_2O is inert, but in the stratosphere N_2O is destroyed by photolysis, and a minor part by reaction with excited oxygen atoms. The latter reaction is the primary source of nitric oxide, which contributes to ozone depletion (Crutzen, 1970). The average N_2O concentration in the northern hemisphere is 0.75-1.0 ppbv higher than in the southern hemisphere (Prinn et al., 1990; Weiss, 1981; Butler et al., 1989).

The atmospheric concentration of nitrous oxide (N_2O) is increasing. The rate of increase of 0.25-0.31% yr^{-1} (Prinn et al., 1990) observed in the 1980s resulted from 30% larger sources than sinks and a 50% growth in N_2O emissions since pre-industrial times (Prinn et al., 1990; Khalil and Rasmussen, 1992; Graedel and Crutzen, 1993). The atmospheric N_2O concentration started to increase rapidly in the 1940s and was faster during the late 1980s than during the mid-1970s (Khalil and Rasmussen, 1992). In 1992 the rate of increase was about half that in the previous decade (Swanson et al., 1993). This was perhaps a result of the observed global cooling associated with aerosols emitted by Mt. Pinatubo in 1991 (Dutton and Christy, 1992) - causing lower N_2O production in soils (Kreileman and Bouwman, 1994) and oceans. The growth rate of atmospheric N_2O started to increase again in late 1993 (J. Elkins, personal communication).

The growth in atmospheric N_2O is caused by a large number of poorly known minor sources, including agricultural soils, soils under natural vegetation, aquatic sources, biomass burning, fossil fuel combustion, industrial sources and traffic (Khalil and Rasmussen, 1992). Recently also atmospheric chemistry, sewage treatment, land disturbance, animal manure, waste incineration and even global warming effects on N_2O emission from biogenic sources have been identified as potential N_2O sources (Banin, 1986; Khalil and Rasmussen, 1992). Our knowledge of the individual source and sink candidates is still poor (Watson et al., 1992).

The aim of this study is to develop a priori grid-based estimates of N_2O sources that can be used in 3-dimensional transport models of the atmosphere. Before analysis with atmospheric models, we need to investigate disagreements between the a priori estimates and the current knowledge of the regional distribution of annual emissions. We also need to assess the uncertainties of the regional distribution of the different sources.

In section 2 we will present preliminary gridded inventories of N_2O emission comparing results from the literature with our results for soils under natural vegetation, grasslands and animal excreta, arable lands and fertilizer use, biomass burning, post-savanna burning and post-clearing effects, oceans, combustion and industrial sources. Section 3 will review other poorly quantified source candidates. In section 4 the comparison of the different estimates with inverse modeling results presented by Prinn et al. (1990) will be discussed.

4.2. GRID-BASED INVENTORIES OF N_2O EMISSIONS

Published inventories on a 1°x1° grid of monthly N_2O emission from soils under natural vegetation, oceans and biomass burning were used. Grid-based inventories of annual emissions were compiled for a number of other sources listed in the global N_2O budget of Khalil and Rasmussen (1992). All inventories are considered representative of the situation before the 1991-1993 decline in the atmospheric N_2O growth. Annual N_2O emissions were calculated for four broad latitudinal zones from the grid-based estimates for comparison with estimates inferred from inverse modeling presented by Prinn et al. (1990). The uncertainty in the annual emission from the four latitudinal zones was addressed by comparing variants of the emission inventories.

4.2.1. *Soils under Natural Vegetation*

The emission from soils under natural vegetation was calculated using the simple global model developed by Bouwman et. al. (1993). This model describes the spatial and temporal variability of the major controlling factors of N_2O production in soils. The basis for the model is the strong relation between N_2O fluxes and the amount of nitrogen (N) cycling through the soil-plant-microbial biomass system (Matson and Vitousek, 1987). The model includes indices for five major regulators of N_2O production. The first index ranks the input of organic matter into the soil. The second, soil fertility, indicates the quality and N content of the organic matter. The effects of soil moisture status and temperature (indices 3 and 4) rank the rate of soil decomposition, mineralization and nitrification. The mineralization of N is indirectly determined by the combination of indices 1-4. The fifth index is the soil oxygen status, which describes conditions for denitrification. The monthly potential for N_2O production - the N2O index - was calculated from the above five indices.

Kreileman and Bouwman (1994) slightly modified the model developed by Bouwman et al. (1993). Bouwman et al. (1993) used the normalized difference vegetation index as a correlate for organic matter input, while Kreileman and Bouwman (1994) used net primary production. The Bouwman et al. (1993) model version was based on the climate database compiled by Shea (1986), whereas Kreileman and Bouwman (1994) used the 0.5° resolution database from Leemans and Cramer (1991). Contrary to Shea (1986), the more recently compiled climate database includes corrections for altitude. Comparison of the monthly N2O index calculated by both model versions with reported measurements for about 30 locations in six ecosystems yielded values for r^2 of close to 0.6 (Bouwman et al., 1993; Kreileman and Bouwman, 1994). The resulting regression equations were used to calculate emissions on a 1°x1° resolution. Bouwman et al. (1993) warned of the considerable uncertainty in the emission estimates. There is a lack of flux measurement data for a number of important ecosystems that have not been sampled, and data for Asia, Africa and Australia are not available. It is not known how the model performs in these areas. The uncertainty in relation to other sources is discussed in section 5.

Table 1. Areas and estimated annual nitrous oxide emission, mean, and standard deviation of N_2O emission for broad groups of current land cover

		NS1†			NS2‡		
		Annual N_2O Emission§			Annual N_2O Emission§		
Type	Area, Mha	Total, Tg N	Mean, kg N ha^{-1}	s.d.¶, %	Total, Tg N	Mean, kg N ha^{-1}	s.d.¶, %
Closed tropical forest	1681	2.3	1.4	36	2.3	1.4	31
Open tropical forest	1028	1.0	1.0	49	1.0	1.0	48
Temperate forest	2246	0.5	0.2	87	0.5	0.2	81
Grassland #	3147	1.5	0.5	91	1.4	0.4	94
Arable land #	1438	1.0	0.7	76	0.9	0.6	76
Other land	3723	0.7	0.2	91	0.5	0.1	97
Ice (excluding Antarctica)	228	0.0	0.0	-	0.0	0.0	-
Total	13490	7.0			6.6		

† NS1 = Bouwman et al. (1993).

‡ NS2 = Kreileman and Bouwman (1994).

§ The emissions were calculated by overlaying the emission inventories from two different studies (NS1 and NS2) with the land cover database of Olson et al. (1983).

¶ s.d. = standard deviation, representing the spatial variability within ecosystems, not the uncertainty of the estimate.

The estimated emissions for arable and grassland represent background emissions, excluding effects of anthropogenic N inputs.

- *Results*. We will compare model results with estimates from the literature. The results of both models (NS1 = Bouwman et al. [1993]; NS2 = Kreileman and Bouwman [1994]) were grouped for broad classes of land cover types based on Olson et al. (1983) (Table 1). The calculated global emission from soils under natural vegetation - excluding arable lands and grasslands - is 4.3-4.5 Tg N_2O-N yr^{-1} (Tg = teragram; 1 Tg = 10^{12} g). The resulting 2.3 Tg N yr^{-1} from 1681 Mha (Mha = 10^6ha = 10^{10} m^2) of closed tropical forests (Table 1) corresponds with the 2.4 Tg N yr^{-1} calculated by Matson and Vitousek (1990) for 1500 Mha of "moist and wet lowland tropical forests". The estimated 1 Tg N yr^{-1} from 1000 Mha of open tropical forests is within the range (0.4-1.3 Tg N yr^{-1}) given by Vitousek et al. (1989) for 1000 Mha of "dry tropical forests". The 0.5 Tg N yr^{-1} obtained for temperate forests is consistent with the range of other estimates (Bowden et al., 1990; Schmidt et al., 1988; Watson et al., 1992).

The models also produce results for current agricultural areas (Table 1). These emission estimates do not account for anthropogenic N inputs and may be used as so-called "background" emission from unfertilized grasslands and arable lands.

4.2.2. *Grasslands and Animal Excreta*

The complex of grasslands (Table 1) includes 2390 Mha of Mediterranean grazing areas, warm grass/shrub and cool grass/shrub complexes given in land cover database of Olson et al. (1983). The 2195 Mha agricultural land (Olson et al., 1983) includes 1438 Mha of arable land (FAO, 1991). The 757 Mha complement was assumed to be part of the grassland complex and the resulting global area of grasslands is thus 3147 Mha (Table 1). FAO (1991) presented an estimate for the global permanent grassland area of 3402 Mha for 1990. Because of definitional differences correction of our estimate was not considered feasible.

- *Results*. The global background emission calculated for grasslands is 1.4-1.5 Tg N yr^{-1} (Table 1). In this estimate the use of synthetic fertilizers in grasslands was neglected because only 6% of global N fertilizer use of 80 Tg N yr^{-1} is applied to 5-8% of the world's grasslands (IFA/IFDC/FAO, 1992). Furthermore, in the background emission estimate the N input from symbiotic N fixation was not considered because of the uncertainty on the occurrence of legumes in grasslands and their contribution to N_2O emission.

We did account for N_2O emission from animal excreta. This N_2O source was recently identified by Khalil and Rasmussen (1992), who estimated a global emission of 0.2-0.6 Tg N yr^{-1} from cattle sheds and barns. Our estimate of N excretion for different animal categories (Table 2) is from Van der Hoek (1994). We assumed that animal excreta are allocated exclusively to grasslands because it is not known to what extent animal excreta are used on global scale to fertilize arable land or for other purposes (e.g. fuel). We assumed that the omission of animal excreta applied as fertilizer to crop lands is compensated by the underestimation of fertilizer use in grasslands. The gridded animal densities of Lerner et al. (1988) were updated with country statistics of the animal populations from FAO (1991) to produce grid-based estimates for 1990. The N excretion from chickens, for which no inventory was available, was distributed over arable land cells from Olson et al. (1983). The N_2O emission may be less than 0.5% of the N in sheep urine (Sherlock and Goh, 1983) to 0.6% for cattle urine (Mosier et al., 1981). Considerable N_2O emissions have been observed from manure-treated soils (Christensen, 1983; Bouwman, 1994) and animals kept in stables (Khalil and Rasmussen, 1992).

The N_2O emission was assumed to be 1% of the N in animal excreta. Although this estimate may be conservative (Bouwman, 1994), it results in a global emission of 1.0 Tg N_2O-N yr^{-1} (Table 3) for the global N excretion from animals of ~100 Tg N yr^{-1} (Table 2). The volatilization of NH_3 from animal excreta was not accounted for since most atmospheric NH_3 is transported over short

Table 2. Animal populations (heads) for 1990 (FAO, 1991) for two world regions, estimated nitrogen excretion and total N in animal excreta. Source: Van der Hoek (1994).

Category	Heads 1990 (x 10^6) Region		N excretion (kg N per head per yr) Region		Total N in excreta (Tg N per yr) Region	
	I†	II‡	I†	II‡	I†	II‡
Dairy cattle	103	125	80	60	8	8
Non-dairy cattle	287	778	45	40	13	31
Buffalo	1	138	45	45	0	6
Camels	0	19	55	55	0	1
Horses	16	45	45	45	1	2
Sheep	528	687	10	10	5	7
Goats	27	560	9	9	0	5
Pigs	339	516	11	11	4	6
Chickens	4427	6335	0.5	0.5	2	3
Total					33	69

† I = Europe, the former USSR, North America, Australia and New Zealand, Israel and Japan.

‡ II = Latin America, Oceania excl. Australia and New Zealand, Africa and Asia excl. former USSR.

Table 3. The fraction of total N_2O emission in each latitudinal zone from animal excreta, biomass burning, post-clearing effects following deforestation, and oceans, and the global emission from each of these sources

Latitudinal zone	Animal excreta	Biomass burning (direct)	Agric. waste burning	Post-clearing effects of deforestation	Oceans†	
					O1	O2
30°-90°N	0.42	0.00	0.50	0.00	0.18	0.21
0 - 30°N	0.37	0.47	0.39	0.59	0.25	0.22
0 - 30°S	0.15	0.53	0.09	0.40	0.27	0.13
30°-90°S	0.06	0.00	0.03	0.00	0.30	0.44
Global emission (Tg N yr^{-1})	1.0	0.1	0.1	0.4	2.0	3.6

† Two different estimates are used (O1 and O2) for ocean.

distances (Langford et al., 1992). Hence, we assumed that volatilized NH_3-N is deposited in the same grid box and that 1% is converted to N_2O in soils.

Our estimates for the background emission from grasslands of 1.4-1.5 Tg N_2O-N yr^{-1} and 1 Tg yr^{-1} from animal excreta are consistent with Duxbury et al. (1993), who proposed a global emission of 1.6 Tg N yr^{-1} for "natural" grasslands and 0.7 Tg N yr^{-1} for "anthropogenic" grasslands.

4.2.3. *Arable Land*

Global estimates of the N_2O emission from fertilized fields show a great uncertainty despite the large number of reported measurements (Bouwman, 1994). Two different inventories of N_2O emissions from arable lands were compiled.

- *Results.* The first inventory (AR1) was based on Eichner (1990), who defined the fertilizer-induced N_2O loss as the difference between the N_2O emission from a fertilized plot and that from an unfertilized control plot, presented as a percentage of the N fertilizer application. The calculated fertilizer-induced N_2O-N emission rates ranged from 0.0% to 1.6% of the N application, depending on the type of fertilizer. These rates represent the N_2O emission during the period covered by the measurements. Eichner (1990) proposed to double the emission factors to include emissions beyond the measurement period and emissions of N_2O from N leaching from fertilizer. Therefore, in AR1 we doubled Eichner's emission rates. This resulted in a global emission rate of 0.5% (0.2-2.2%) from N application for the global mix of N fertilizer types used in 1990 (IFA, 1992; FAO, 1991) and a global emission of 0.4 (0.1-1.7) Tg N_2O-N yr^{-1} (Table 4). There are a number of uncertainties in AR1:

- Mosier (1993) and Bouwman (1994) concluded that the measurement data set currently available from the literature was too limited to calculate the N_2O emission for each fertilizer type. For example, anhydrous ammonia contributes ~45% to the global N_2O emission of AR1. This estimate would be reduced by 40% by adding data for 14 measurements of anhydrous ammonia (Bouwman, 1994). The available data may, however, be adequate to estimate the order of magnitude of emissions.

Table 4. Areas, estimated total N_2O emission and fractions of total emission released in each latitudinal zone for NS1 and NS2 for natural soils and grasslands, and AR1 and AR2 for arable land

Latitudinal zone	Soils under natural vegetation	Grasslands	Arable land	
			AR1†	AR2‡
		Area (Mha)		
30°-90°N	3954	1380	786	786
0° - 30°N	2486	758	436	436
0° - 30°S	1993	773	148	148
30°-90°S	244	236	68	68
Total	8677	3147	1438	1438
NS1§				
	Fraction of total emission released in each zone			
30°-90°N	0.13	0.23	0.81	0.51
0° - 30°N	0.37	0.29	0.16	0.35
0° - 30°S	0.48	0.39	0.02	0.11
30°-90°S	0.02	0.09	0.01	0.03
		Emission (Tg N yr^{-1})		
Background	4.5	1.5	0.0	1.0
Fertilizer-induced	0.0	0.0	0.4	1.0
Total	4.5	1.5	0.4	1.9
NS2¶				
	Fraction of total emission released in each zone			
30°-90°N	0.12	0.18	0.81	0.50
0°-30°N	0.40	0.37	0.16	0.38
0°-30°S	0.47	0.39	0.02	0.10
30°-90°S	0.01	0.06	0.01	0.02
		Emission (Tg N yr^{-1})		
Background	4.3	1.4	0.0	0.9
Fertilizer-induced	0.0	0.0	0.4	1.0
Total	4.3	1.4	0.4	1.8

† AR1 = 2x Eichner's fertilizer-induced emission, independent of background emission and identical for NS1 and NS2.

‡ AR2 = background emission + 1.25% of N-fertilizer application

§ NS1 = Bouwman et al. (1993)

¶ NS2 = Kreileman and Bouwman (1994)

- Most reported measurements cover the crop season or shorter periods. Most of the N_2O is usually released during a period of a month after fertilizer application (Breitenbeck and Bremner, 1986), after which emissions decline to a "background" level. Bouwman (1994) showed that observed N_2O emission from fertilized lands in the literature is significantly higher for longer measurement periods than for shorter measurement periods. This indicates that the background emission should also be included in estimates of the annual emission from arable lands.

Therefore, the second inventory for arable lands (AR2), presented in Table 4, includes estimates for both the background emission and the fertilizer-induced emission - independent of fertilizer type. The background N_2O emission was derived from grid-based model estimates NS1 (Bouwman et al., 1993) and NS2 (Kreileman and Bouwman, 1994). The global background N_2O emission calculated for arable lands given by Olson et al. (1983) is 0.9-1.0 Tg N_2O-N yr^{-1} (Tables 1 and 4). This estimate includes variability caused by climate and soils but does not account for effects of management. The available one-year zero-fertilizer experiments show a wide range of N_2O emissions (Bouwman, 1994); even net annual uptake of N_2O was observed by Ryden (1981; 1983). This variability may be caused by the combined effect of management and environmental conditions (Mosier, 1993; Bouwman, 1994).

The estimated fertilizer-induced N_2O emission for AR2 was based on Bouwman (1994), who selected measurements covering periods of one year from the data sets presented by Eichner (1990), Bouwman (1990) and additional reports. The resulting N_2O emission represented a linear function of the N fertilizer application independent of fertilizer type and soil conditions. The regression line indicated N_2O losses of 1.25% of the N applied. Although the r^2 calculated was 0.8, there is considerable uncertainty in this N_2O emission rate because of the limited number of available one-year measurements. However, the percentage of N_2O emission is within the range of the 0.5-2% estimated by others (Bolle et al., 1986; Skiba et al., 1992; Velthof and Oenema, 1993). The N_2O emission rate of 1.25% and the ~80 Tg N yr^{-1} of N fertilizer use in 1990 yields a global fertilizer-induced emission of 1.0 Tg N_2O-N yr^{-1}. For AR2 the sum of background emission and fertilizer-induced N_2O emission is 1.8-1.9 Tg N yr^{-1} (Table 4).

For allocating fertilizer application we used a 1°x1° resolution country database (Lerner et al., 1988) and the 1990 N fertilizer mix for 90 countries from IFA (1992), representing > 98% of the ~80 Tg N yr^{-1} global synthetic N fertilizer consumption. For 60 other countries we used the total 1990 N fertilizer consumption from FAO (1991), assuming that this fertilizer mix was identical to the regional data from IFA (1992). For 34 minor countries fertilizer data are not reported in IFA (1992) and FAO (1991). The average country N application level was calculated as the ratio of country N fertilizer use / area arable land + permanent crops (FAO, 1991). For AR1 this was calculated for each fertilizer type, and for AR2 the total country N fertilizer consumption was used. The use of mean N fertilizer application levels for all arable land within a country is not realistic (IFA/IFDC/FAO, 1992). Unfortunately, grid-based data on application levels, and crop and management levels (e.g. irrigated and rainfed) are not available on the global scale.

The N_2O emissions were simply assigned to the arable grid boxes given by Olson et al. (1983). We made corrections using country data from FAO (1991) for arable land and permanent crops by using country-specific correction factors. The difference between the estimated 1438 Mha of arable area (Table 1) and the 1444 Mha reported by FAO (1991) occurs because a number of minor countries are not represented in the country database of Lerner et al. (1988).

4.2.4. *Biomass Burning*

Several global estimates have been made of emissions from biomass burning during forest clearing, natural forest fires, savanna burning, shifting cultivation and agricultural residue burning. Crutzen and Andreae (1990) reported a N_2O emission from the burning of 0.1-0.3 Tg N yr^{-1} based on N_2O release of 0.7±0.3% of the N content of the material burned, with N/C ratios of 0.01 for forests, 0.006 for savannas and 0.01-0.02 for agricultural waste. Lobert et al. (1990) measured a fraction of N_2O-N/N in biomass of 0.77±0.21%. Other authors used ratios of N_2O-N/C-burned of 0.01-0.03% (Hao et al., 1990; Cofer et al., 1991; Andreae, 1991) yielding global emissions of 0.5-2.2 Tg N yr^{-1}. The most recent global N_2O budget includes 0.2-1 Tg N yr^{-1} for biomass burning (Watson et al., 1992). We used N_2O emission factors presented by Crutzen and Andreae (1990).

- *Results.* Emissions were distributed on the basis of the 5°x5° grid distributions for savanna burning and deforestation, compiled by Hao et al. (1990) (Table 3). The estimated global emission (excluding fuelwood and agricultural waste burning) is 0.1 Tg N yr^{-1}.

For agricultural waste burning we used regional estimates of Andreae (1991) and emission factors from Crutzen and Andreae (1990). For the global agricultural waste burning of ~900 Tg C yr^{-1} the calculated emission of 0.1 Tg N yr^{-1} was distributed over the arable land cells of each region (Table 3).

4.2.5. *Tropical Forest Conversion*

Human activity in tropical forests changes the emission rates of biogenic trace gases (Keller et al., 1986). Accelerated decomposition and mineralization of litter, root material and soil organic matter in the first years after forest clearing may cause a pulse of N_2O emissions. The annual N_2O emission from a Brazilian pasture converted from tropical forest 4-5 years beforehand, exceeded the emission from a forest site growing on identical soils by a factor of 3 (Luizao et al. (1989). Keller et al. (1993) found emissions from 2-10 year-old pastures that exceeded forest soil emissions of N_2O by a factor of 5-8. Enhanced emissions are generally not observed in older clearings (Robertson and Tiedje, 1988; Sanhueza et al., 1990). This may be caused by the gradual decreasing content of easily decomposable soil organic matter along with the age of the clearing (Keller et al., 1993). In Costa Rica Keller et al. (1993) observed lower emissions (1/3 to 1/2) from old pastures (>12 years) than from adjacent forest sites with identical soils and a gradual decrease in emissions with the aging of pastures.

- *Results.* Matson and Vitousek (1990) estimated a global emission of 0.7 Tg N yr^{-1} for 220 Mha of forest converted after 1950 to pasture. This estimate does not consider declining N_2O emission along with pasture age. To include this phenomenon, applying to forest conversion to grassland and arable land, we assumed the N_2O increase to last for 10 years (as in Keller et al., 1993). The emission is assumed to be five times the average emission for a tropical rainforest of 1.4 kg N $ha^{-1}yr^{-1}$ (Table 1) in year 1 after clearing (Table 5). Between the years 2 and 10 after the clearing there is a linear decrease in N_2O emission to the average values for arable land or grassland estimated for NS2 at 0.6 and 0.4 kg N ha^{-1} yr^{-1}, respectively (Table 1).

Tropical forest conversion rates were taken as 2.4 Mha yr^{-1} for pastures and 12.6 Mha yr^{-1} for arable land. These rates were updated from Houghton et al. (1987) with 1990 deforestation estimates from FAO (1993). The calculated global emission of 0.4 Tg N yr^{-1} (Table 5) was distributed on the basis of the deforestation inventory of Hao et al. (1990) (Table 3). The calculated N_2O emission for grassland after 4-5 years (Table 5) is comparable to the 5.7 kg N $ha^{-1}yr^{-1}$ measured by Luizao et al. (1989) for a 4-5 year old pasture, but much lower than the emissions reported by Keller et al. (1993), which shows the uncertainty.

4.2.6. *Enhanced Soil N_2O Emissions Following Savanna and Grassland Burning*

Burning makes N and other nutrients available in the residual material (Sanchez, 1982). During the rains following the fires that usually occur at the end of the dry season (Hao et al., 1990), this may lead to prolonged enhancement of N_2O emissions (Anderson et al., 1988; Anderson and Poth, 1989). The global total amount of residual N on the surface after savanna burning may be 0.6-2.4 Tg N yr^{-1} (based on Crutzen and Andreae, 1990). Even if we assume that a high 5% of this amount is released as N_2O after mineralization and nitrification, the resulting N_2O emission would be < 0.12 Tg N yr^{-1}. Measurements of enhanced N_2O emissions following biomass burning are too scarce to verify this. In our a priori estimate this source is assumed to be negligible.

Table 5. Effect of annual forest conversion to permanent pasture and arable land on N_2O fluxes

	2.4 Mha yr^{-1} forest conversion to permanent pasture†		12.6 Mha yr^{-1} forest conversion to arable land†	
Years after clearing (yr)	N_2O loss after clearing‡ (kg N $ha^{-1}yr^{-1}$)	Total extra N_2O loss§ (Gg¶ N yr^{-1})	N_2O loss after clearing‡ (kg N $ha^{-1}yr^{-1}$)	Total extra N_2O loss§ (Gg¶ N yr^{-1})
1	7.0	13	7.0	71
2	6.3	12	6.3	62
3	5.5	10	5.6	53
4	4.8	8	4.9	44
5	4.1	6	4.2	35
6	3.3	5	3.4	26
7	2.6	3	2.7	17
8	1.9	1	2.0	8
9	1.1	-1	1.3	-1
10	0.4	-2	0.6	-10
Total		55		303

† Clearing rates updated from Houghton et al. (1987) for early 1980s with data from FAO (1993) for late 1980s.

‡ In the first year after clearing assumed emissions are five times higher than the pre-clearing emission for closed tropical forest of 1.4 kg N ha yr^{-1} (Estimate NS2, Table 1). In years 2-10 emissions decrease linearly to the average level calculated for grasslands (0.4 kg N yr^{-1}) and arable lands (0.6 kg N yr^{-1}), respectively (estimate NS2, Table 1).

§ Calculated as the difference between N_2O emission after clearing and the pre-clearing emission, times the forest conversion rate.

¶ Gg = gigagram; 1 Gg = 10^9 g

4.2.7. *Oceans*

Fluxes from water surfaces cannot be measured directly. Generally, oceanic fluxes are modeled from N_2O supersaturation in the surface water and the water-air gas transfer coefficient. Measurements of N_2O supersaturation cover only a minor part of the area covered by the world's oceans (Butler et al., 1989; Weiss et al., 1992) and it is not clear if measurements include sufficient seasonal differences. The water-air gas transfer coefficient is a function of windspeed and solubility and diffusivity of N_2O in sea water. Given the scarcity of measurements, global estimates are fraught with potential error and the uncertainty in the results may be 50% (Erickson, 1988). Watson et al. (1992) presented an estimate of 1.4-2.6 Tg N yr^{-1} based on Butler et al. (1989), with low emissions attributed to El Niño Southern Oscillation events (ENSO) and higher emissions in non-ENSO years. There is considerable spatial variability. High fluxes were calculated for the Indian Ocean upwelling region (Butler et al., 1989; Law and Owens, 1990a), Kuroshio extension (Butler et al., 1989) and the Antarctic Ocean (Nevison, 1994). Antarctic upwelling water has high N_2O supersaturation throughout the year (Weiss et al., 1992) and there is high turbulence caused by frequent storms (Owens, 1993).

- *Results.* We will compare two different estimates. The first (O1) is 2.0 Tg N yr^{-1} - the mean of the range given by Watson et al. (1992) - homogeneously distributed over ocean surface area in each latitudinal zone as in Prinn et al. (1990) (Table 3). We realize that this distribution does not reflect the observed variability of emissions. The second estimate (O2) is the inventory developed by Nevison (1994), based on ocean cruise data from Butler et al. (1989) and Weiss et al. (1992). Different functions to estimate fluxes resulted in a range of global estimates of 2.8-5.7 Tg N yr^{-1}. We adopt Nevison's "best" estimate resulting from the water-air flux calculation of Erickson (1993). The global emission of 3.6 Tg N_2O-N yr^{-1} for O2 is 0.2 Tg N yr^{-1} lower than the estimate by Nevison (1994). This was caused by the conversion from the original 2.8°x2.8° grid to our 1°x1° grid. Major differences between O1 and O2 are in the 0°-30°S and 30°-90°S zones, where O2 gives high emissions in line with the above discussed observations, while O1 is based on the global mean ocean flux in all zones.

4.2.8. *Fossil Fuel Combustion*

Commercial energy use by stationary combustion is a minor source of N_2O (Khalil and Rasmussen, 1992). Mobile combustion, in particular by cars equipped with catalytic converters, is a more important source and growing fast (Berges et al. (1993). We used emission estimates for 13 world regions (including individual countries such as the USA, Canada, the former USSR and Japan) reported by De Vries et al. (1994). These were based on energy data from IEA (1992). Emission factors for stationary sources were taken from De Soete (1993). We assumed that 100% of the automobiles in the USA and Canada were equipped with new catalytic converters in 1990, for which the estimated emission factor is 6.4 g N_2O-N/GJ (Olivier, 1993). This is a conservative estimate, since aged catalysts may have much higher emission factors. However, many old cars may not be equipped with catalysts. For other regions we assumed that in 1990 vehicles were not equipped with catalytic converters. For these we used emission factors as proposed by OECD (1991).

- *Results*. The global emission from fossil fuels thus calculated is 0.3 Tg N yr^{-1} for 1990, with a range of 0.1-0.6 Tg N yr^{-1}, based on Olivier (1993). This is lower than the range of 0.3-0.9 Tg N yr^{-1} presented by Watson et al. (1992), mainly caused by recently published lower emission factors for stationary sources. Our estimate is consistent with other estimates for stationary combustion (Khalil and Rasmussen, 1992) and mobile sources (Berges et al., 1993). Emissions distributed on the basis of the global database of population densities presented by Lerner et al. (1988) show that ~90% of the global emission from this source comes from the 30°-90°N zone (Table 6).

Table 6. Fraction of the total N_2O emission in each latitudinal zone from fossil fuel combustion, fuelwood combustion and industry (including adipic acid and nitric acid production) and the global emission from each of these sources

Latitudinal zone	Fossil fuel combustion	Fuelwood combustion	Industry	
			AA†	NA‡
30°-90°N	0.90	0.33	0.61	0.71
0 - 30°N	0.09	0.52	0.32	0.24
0 - 30°S	0.01	0.15	0.06	0.04
30°-90°S	0.01	0.01	0.00	0.01
Global emission (Tg N yr^{-1})	0.3	0.1	0.3	0.2

† AA = adipic acid production.
‡ NA = nitric acid production.

4.2.9. *Fuelwood Combustion*

Little data has been published on fuelwood burning and associated emissions of N_2O. Our data is from De Vries et al. (1994), who estimated that fuelwood energy consumption based on FAO (1991) and IEA (1992) data for 13 world regions is ~19 PJ yr^{-1}. This is 6% of the world's energy supply. Other estimates for total global biomass use for energy purposes are as high as 14% (e.g. Scurlock and Hall, 1990). No representative measurement data are available for N_2O emission from fuelwood combustion (Smith et al., 1993).

- *Results.* The emission factor of 6.4 g N_2O-N/GJ estimated by De Vries et al. (1994) yields a global emission of 0.13 Tg N yr^{-1}, with a range of 0.0-0.5 Tg yr^{-1} inferred from Smith et al. (1993) and the uncertainty in global fuelwood consumption. This is consistent with the 0.15 and 0.16 Tg N yr^{-1} estimated by Andreae (1991) and Pepper et al. (1992), respectively. Similar to fossil fuel combustion, we used population densities to distribute emissions. About 70% of the estimated global emission from fuelwood burning stems from the tropics (30°N-30°S) and 30% from the 30°-90°N zone (Table 6).

4.2.10. *Adipic Acid Production*

Recently, adipic acid (AA) manufacturing has been recognized as an important source of N_2O (Thiemens and Trogler, 1991). Watson et al. (1992) estimated that emissions from AA production may be 0.4-0.6 Tg N yr^{-1}. Adipic acid is a compound primarily used for the production of 6,6 nylon (Castellan et al., 1991). Cyclohexane is used to produce a mixture of cyclohexanone and cyclohexanol, which is subsequently oxidized with nitric acid to produce AA. Nitrous oxide is an intrinsic by-product of this oxidation step (Thiemens and Trogler, 1991). Our inventory for this source was based on the production capacity and coordinates of the AA plants given by Castellan et al. (1991), with additional information for Brazil, China and Korea from McCulloch (1993, personal communication). We estimated that the global production capacity of AA in 1990 was 2175 Mg yr^{-1}. The actual global production may be some 20-25% lower. However, we assumed that actual production equals the capacity to compensate for production in the former USSR and Eastern European countries for which no data were available. This estimate is close to Thiemens and Trogler (1991), who reported an estimated global production of 2200 Mg yr^{-1} for 1989.

The unabated N_2O emission rate is 191 g N_2O-N per kg AA produced. A 98% reduction of N_2O emission was achieved by incineration at the Du Pont plant in Victoria, Texas (USA), which accounts for about 15% of the world's AA production capacity (Thiemens and Trogler, 1991). The N_2O abatement at all Du Pont plants - in total about 30% of the world's production capacity - is 53% (Reimer et al., 1992) at present. This corresponds to an emission factor of 4 g N_2O-N kg^{-1} AA (98% abatement) for the Du Pont plant in Victoria and of 191 g N kg^{-1} AA for the other Du Pont plants. The current global abatement is 32% (Reimer et al., 1993). Therefore, we used an emission rate of 147 g N kg^{-1} AA produced for all other manufacturers corresponding to 23% abatement.

- *Results.* The calculated global emission from AA production is 0.3 Tg N_2O-N yr^{-1}. A range of 0.2-0.4 Tg N_2O-N yr^{-1} is obtained for a low actual production of only 80% of production capacity, and by assuming 100% utilization of the production capacity and no abatement. Our range is lower than the 0.4-0.6 Tg N yr^{-1} estimated by Watson et al. (1992) because we used recent information about the abatement of emissions. About 60% of the N_2O from AA production may stem from the 30°-90°N zone and ~90% from the northern hemisphere (Table 6).

4.2.11. *Nitric Acid Production*

Manufacture of nitric acid (HNO_3) was recently identified as a source of N_2O (Watson et al., 1992). Nitric acid (NA) is used as the main feedstock in fertilizer production. Off-gas measurements at Du Pont and other nitric acid producers show that N_2O is an unwanted accidental by-product of oxidation of NH_3 over a catalyst at elevated temperatures (Olivier, 1993). To estimate the global NA production in 1990 we used UN (1993) data. Because data for 1990 were not reported for many countries we extrapolated historical data for about half of the global production. The resulting global NA production could be 7 Tg HNO_3-N yr^{-1}. Global production estimates by the industry range from 11 to 14 Tg HNO_3-N yr^{-1} (Reimer et al., 1992; McCulloch, personal communicaton). Comparison of UN and industry statistics shows inconsistencies for many countries. Therefore, we adopted a current global NA production level of 11 Tg N yr^{-1}. Since the UN (1993) statistics on NA production are incomplete and most NA is used for the production of nitrogen fertilizers, we used the country statistics of N fertilizer production from IFA (1992) to distribute the global 11 Tg N yr^{-1} of NA production among countries. The emissions were spread spatially on the basis of population density as described for fossil fuel combustion.

- *Results*. Reported emission factors for NA production are in the range of 7 to 27 g N_2O-N kg^{-1} HNO_3-N (Olivier, 1993). The global NA production of 11 Tg N yr^{-1} then yields an estimate of 0.2 (0.1-0.3) Tg N yr^{-1}, of which ~70% stems from the 30°-90°N zone (Table 6). Our global estimate corresponds with the 0.1-0.3 Tg N yr^{-1} estimated by Watson et al. (1992).

4.3. OTHER SOURCES AND SINKS OF N_2O

Apart from the sources discussed in section 2, there are other candidate sources and sinks. For some of these it is difficult to provide even the order of magnitude of emissions, while others may be globally insignificant. We realize that the list of sources discussed below may be incomplete.

4.3.1. *Industrial and Chemical Processes*

Processes, particularly those which involve nitrogen oxidation steps in overall reducing conditions, are potential sources of N_2O. Examples include use of nitric acid for other purposes than fertilizer production, production of dioic acids, coking of coal, chlorination of water (e.g. in power plants), cement manufacturing (pre-calciner) and metal treatment processes (Olivier, 1993). No estimates have been reported in the literature for these sources. An upper limit may be estimated on the basis of global total production of basic materials and of the N content.

Other sources of N_2O are the production and use of explosives (Volk, 1978), industrial use and N_2O used for anesthesia (Logan and Farmer, 1989). Granli and Bøckman (1994) estimated that N_2O in fumes from explosives is insignificant (< 0.01 Tg N yr^{-1}). The global N_2O released to the atmosphere from medical and industrial uses may be 0.03 Tg N yr^{-1}. Corona power loss from electric transmission systems generate at least 0.04 Tg N yr^{-1} (Granli and Bøckman, 1994).

4.3.2. *Atmospheric N_2O Formation*

Formation of N_2O from atmospheric NH_3 oxidation by OH may be in the order of 0.3-1.2 Tg N yr^{-1} (Dentener, 1993). This estimate is based on a 10°x10° resolution 3-dimensional model study, showing that a major part of the N_2O production takes place in the tropics. Khalil and Rasmussen (1992) suggested the possibility of N_2O formation in exhaust gases from combustion sources. Similarly, N_2O formation may occur in smoke plumes from biomass burning. To date no estimates for these processes have been published.

4.3.3. *Coastal Waters and Freshwater Systems*

Measurements indicate that eutrophication of aquatic systems may cause increased N_2O production (Seitzinger, 1988). Global estimates of the N_2O emission from aquatic systems contaminated with human waste range from 0.01-1.6 Tg N yr^{-1} (Kaplan et al., 1978; McElroy et al., 1978; Elkins et al., 1979; Hemond and Duran, 1989). Sewage-water treatment has also been identified as a source of N_2O (Banin, 1986) but this may be of local importance, for example, in northern latitudes (Van den Born et al., 1991). Emissions of N_2O from lakes contaminated with N (Labroue et al., 1991), the North Sea (Law and Owens, 1990b) and different estuaries and inland waters (Seitzinger, 1988) are generally higher than oceanic fluxes. This indicates that these systems may be important global N_2O sources as suggested by Butler et al. (1989).

Inputs of N to surface waters from human waste, leaching from soils and N deposition are difficult to quantify (Owens, 1993). In rainfed agriculture in Europe, leaching of N may be an important source of nitrate pollution of groundwater (RIVM/RIZA, 1991). Nitrate leached from N-enriched soils may eventually enter surface waters, where it may be denitrified and form a potential source of N_2O. Conrad et al. (1983) estimated that fertilizer-N leached from soils could contribute 1.1 Tg N yr^{-1} globally.

4.3.4. *N_2O in Soil Solution*

Dissolved N_2O in soil water may leach to the groundwater and be released to the atmosphere by degassing from groundwater entering surface streams or from groundwater used as irrigation water. Ecosystem losses of N_2O via the solution and contributions to global N_2O may be considerable (Dowdell et al., 1979; Bowden and Bormann, 1986; Davidson and Swank, 1990). Measured losses via the solution may be of the same order of magnitude as observed N_2O emissions from agricultural soils to the atmosphere. The losses of N_2O dissolved in drainage water originating from wetland rice paddies may be considerable (Minami and Fukushi, 1984). Ronen et al. (1988) estimated that aquifers contaminated with N may contribute 0.5-1.1 Tg N yr^{-1}.

4.3.5. *N Deposition*

The global terrestrial deposition of N may be 36 Tg NO_x-N yr^{-1} (Jaffe, 1992) and 75 Tg NH_3-N yr^{-1} (Schlesinger and Hartley, 1992), with about half of the NH_3 stemming from animal excreta. This N input may result in enhanced N_2O evolution. High N_2O emissions of close to 6 kg N $ha^{-1}yr^{-1}$ were observed by Brumme and Beese (1992) for acid forest soils receiving an N deposition of 35 kg N $ha^{-1}yr^{-1}$. However, N additions to forest soils did not always lead to these high emissions (Castro et al., 1993). If we exclude the NH_3 from animal excreta (already accounted for) and assume an emission factor of 1%, the N_2O emission induced by N deposition could be 0.8 Tg N yr^{-1}. We cannot give reliable latitudinal estimates, but most of the enhanced N_2O emission may stem from the industrialized countries in the 30°-90°N zone.

4.3.6. *Biological N Fixation*

The models of Bouwman et al. (1993) and Kreileman and Bouwman (1994) do not consider N input from symbiotic and nonsymbiotic N fixation. There are estimates for a limited number of natural ecosystems (Boring et al. 1988), but the available data are not adequate to provide reliable estimates of the amount of N fixation. In addition, it is not clear how these N inputs affect N_2O losses from soils in natural systems. In agricultural systems reported N_2O emissions from fields with leguminous crops may be in the order of 2.3 to 4.2 kg N $ha^{-1}yr^{-1}$ for alfalfa (Duxbury et al.,

1982) to 0.34 to 1.97 kg for soybean (Bremner et al., 1980). As these crops usually receive little or no N fertilizer, these high N_2O emissions may be attributed to N inputs from symbiotic N fixation. At the global scale leguminous crops make up 145 Mha (FAO, 1991), about 10% of the total arable land, excluding legumes grown as green manures not reported by the FAO (1991), and legumes in grasslands and N fixing grass species. The N inputs from legumes to agricultural systems may be of the same order of magnitude as global synthetic N fertilizer use (Duxbury et al., 1993), indicating the potential importance for the N_2O cycle.

4.3.7. *Land Degradation and Conversion to Urban Land*

Degradation of agricultural land may lead to a decline in N_2O emission due to loss of soil fertility. Global land losses due to agricultural mismanagement may be 4 Mha yr^{-1} and conversion of agricultural land to other uses such as urban land and infrastructure 8 Mha yr^{-1} (Buringh and Dudal, 1987). The net result of forest clearing, degradation losses and conversion to other uses is a globally observed increase of pasture of 6 Mha yr^{-1} and a decrease in developing countries of 9 Mha yr^{-1} in the period 1960-1990. There was a net increase of 4 Mha yr^{-1} of arable land during 1960-1990 (FAO, 1991). Comparison with forest clearing rates of 15 Mha yr^{-1} (Table 5) shows the importance of land degradation.

4.3.8. *Consumption of N_2O in Soils*

Although forest soils are net annual sources of N_2O, they may sometimes act as N_2O sinks (Keller et al., 1986; Schmidt et al., 1988; Castro et al., 1993). Ryden (1981; 1983) even observed a net annual consumption of N_2O by grassland soils. Water surfaces may also consume N_2O (Kieskamp et al., 1991; Minami and Fukushi, 1984). Periodic or net annual consumption by soils and water may have important repercussions for the global source and the atmospheric lifetime of N_2O. Cicerone (1989) concluded that a global soil sink of 1.5-3 Tg N yr^{-1} may reduce the atmospheric lifetime by as much as 20%.

4.4. DISTRIBUTIONS OVER LATITUDINAL ZONES

Prinn et al. (1990) estimated that the N_2O emissions from the 30°-90°N, 0°-30°N, 0°-30°S, and 30°-90°S latitudinal zones account for, respectively, 22-34%, 32-39%, 20-29% and 11-15% of the global source. These results were obtained with inversion techniques using a simple 9-box atmospheric model. The uncertainty of the emissions from the four latitudinal zones was interdependent, including both the model uncertainty and the standard errors in atmospheric N_2O observations (Prinn et al., 1990). The emission inventories described in section 2 were compared with the mean of the zonal source estimates and the global source of 13 Tg N_2O-N yr^{-1} inferred by Prinn et al. (1990).

- *Results.* The uncertainty of our emission estimates was assessed for eight cases (Table 7). The overall deviation for each case was calculated as the sum of the absolute values of zonal deviations of our calculated zonal emission from the mean of the range deduced by Prinn et al. (1990).

In cases 1a-4b NS1 and NS2 for soils under natural vegetation and estimates AR1 and AR2 for the world's arable lands are compared using O1 and O2 for the oceans and a priori estimates for the other sources (Tables 3 and 6). For NS2 the deviation is overall lower than for NS1. NS2 is lower than NS1 in the extra-tropical zones and in the 0°-30°S tropical zone and somewhat higher in the 0°-30°N zone (Table 4).

Table 7. Global emissions from individual sources of N_2O (Tg N yr^{-1}), zonal emissions, the deviation of the zonal emission from source estimates inferred from inverse modelling, the overall deviation and ratio of northern/southern hemisphere emissions (NH/SH) for eight cases

	Emission inferred from inverse modeling (Prinn et al., 1990)	Case											
		1a	1b	2a	2b	3a	3b	4a	4b	5	6	7	8
		Global emission†											
Source													
Soils under natural vegetation and grasslands		NS1	NS1	NS1	NS1	(NS2)	(NS2)	(NS2)	(NS2)	(NS2)	(NS2)	(NS2)	(NS2)
Oceans		O1	(O2)	O1	(O2)	O1	(O2)	O1	(O2)	(O2)	(O2)	(O2)	(O2)
Arable land incl. fertilizer use		AR1	AR1	(AR2)	(AR2)	AR1	AR1	(AR2)	(AR2)	(AR2)	(AR2)	(AR2)	(AR2)
Animal excreta		1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	(0.0)	(2.0)	1.0	1.0
Biomass burning		0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	(0.2)	0.1
Agricultural waste burning		0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	(0.2)	0.1
Post-clearing effects deforestation		0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Industry (NA + AA production)		0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	(0.7)
Energy/automobiles		0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	(0.6)
Fuelwood combustion		0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	(0.3)	0.1
		Zonal emission†											
Latitudinal zone													
30°-90°N	3.6 (2.9-4.4)	2.7	3.1	3.4	3.8	2.6	3.0	3.1	3.5	3.1	4.0	3.6	4.0
0° -30°N	4.6 (4.2-5.1)	3.6	3.9	4.2	4.5	3.7	4.0	4.3	4.6	4.2	5.0	4.8	4.7
0° -30°S	3.2 (2.6-3.8)	3.7	3.6	3.9	3.8	3.5	3.4	3.7	3.6	3.5	3.8	3.7	3.6
30°-90°S	1.7 (1.4-2.0)	1.0	1.9	1.0	2.0	0.9	1.9	0.9	1.9	1.8	2.0	1.9	1.9
Total		10.9	12.5	12.5	14.1	10.7	12.3	12.1	13.7	12.6	14.8	14.0	14.2
		Deviation of zonal emission from mean zonal estimates by Prinn et al. (1990)†											
Latitudinal zone													
90°-30°N		-0.9	-0.5	-0.3	0.1	-1.1	-0.7	-0.5	-0.1	-0.5	0.3	0.0	0.3
0° -30°N		-1.0	-0.8	-0.4	-0.1	-0.9	-0.6	-0.3	0.0	-0.4	0.4	0.1	0.1
0° -30°S		0.5	0.4	0.7	0.6	0.3	0.3	0.5	0.4	0.3	0.6	0.5	0.5
30°-90°S		-0.7	0.3	-0.7	0.3	-0.8	0.2	-0.8	0.2	0.1	0.3	0.2	0.2
Overall deviation‡		3.2	2.0	2.1	1.2	3.1	1.7	2.1	0.8	1.3	1.6	0.9	1.1
Ratio NH/SH		1.3	1.3	1.5	1.4	1.4	1.3	1.6	1.5	1.4	1.5	1.5	1.6

† Emissions are rounded off to 0.1 Tg. Emissions for the latitudinal zones for NS1, NS2, O1, O2, AR1 and AR2, and other individual sources, are found in Tables 3-6. Source estimates deviating from case 1 are in parenthesis.

‡ The overall deviation is the sum of the absolute values of the zonal deviations.

For the oceans, O2 gives lower total emissions in the 30°-90°N zone than O1, while in the 0°-30°N zone O2 is higher than O1. In the 0°-30°S zone, the difference between O1 and O2 is only 0.1 Tg yr^{-1}. The major cause of lower overall deviation for O2 for all assumptions on natural and agricultural soil emissions is the estimate for the 30°-90°S zone, where O2 is almost 1 Tg yr^{-1} higher than O1. For AR1 the estimated emissions from all zones except the 0°-30°S region are under the lower end of the range given by Prinn et al. (1990) for both O1 and O2. AR2 results in a lower overall deviation than AR1, in particular for the O2 oceanic estimate. The difference between AR1 and AR2 inventories for the southern hemisphere are small because fertilizer use prevails in the northern hemisphere. Case 4b (based on NS2, O2 and AR2) was used as a basis for cases 5-8 because its overall deviation is lower than for cases 1a-4a.

In case 5 the N_2O emission from animal excreta was reduced to 0, while in case 6 it was doubled relative to case 4b. Comparison of cases 4b, 5 and 6 shows that changing the N_2O emission from animal excreta results in high deviations in the northern hemisphere. Lowering estimates for N_2O from excreta in the southern hemisphere results in more consistency with inverse modeling estimates. In the southern hemisphere the effect of changes in N_2O from animal excreta is much smaller than north of the equator because animal populations are smaller in the southern than in the northern hemisphere (Table 3).

Doubling the direct emission from biomass burning (including agricultural waste burning and fuelwood combustion), relative to case 4b causes a minor increase in all latitudinal zones in case 7. This results in a somewhat higher overall deviation than for case 4b. For case 7 the emissions from the 0°-30°S zone are close to the upper end of the range presented by Prinn et al. (1990).

In case 8 the high global estimates presented in section 2 for energy/automobiles (0.6 Tg yr^{-1}) and industrial sources (0.4 Tg N yr^{-1} from AA and 0.3 Tg N yr^{-1} from NA production) were used, with all other sources identical to case 4b. As fossil fuel combustion and industrial sources are dominant in the northern temperate zone (Table 6), there is an increase of ~0.4 Tg yr^{-1} in this zone for case 8 compared to case 4b.

4.5. DISCUSSION

Recent estimates of the *global* source of 13-16 Tg N yr^{-1} (Khalil and Rasmussen, 1992; Minschwaner et al., 1993) show that the global emission may be higher than the upper end of the range of 14.6 Tg N yr^{-1} estimated by Prinn et al. (1990). The zonal inverse modeling estimates may also be underestimated. Somewhat higher zonal estimates than those calculated from Prinn et al. (1990) would result in lower positive and higher negative zonal deviations.

For temperate *natural soils* the Kreileman and Bouwman (1994) model version yields lower estimates than that of Bouwman et al. (1993) and more agreement with inverse modeling estimates. Differences between the NS1 and NS2 inventories may be caused by differences in the climate databases and in the correlate for organic matter input used. Part of the difference in temperate zones may be explained by temperature corrections for altitude in the climate database used for NS2. Lower temperatures in NS2 for temperate mountainous regions result in lower emissions. It is not clear why NS1 and NS2 differ in tropical areas. NS1 and NS2 do not consider observed significant episodic N_2O emissions in temperate regions in autumn (Duxbury et al., 1982), N_2O emission from snow-covered soils (Sommerfeld et al., 1993) and after melting of soil frost (Dörsch et al., 1993). Although this may increase our estimate for temperate zones, higher emissions in temperate regions are possible within the range noted by Prinn et al. (1990).

The emission from the 0°-30°S zone is close to the upper end of the range of the inverse modeling estimate for all cases. Varying the source strength and distribution of emissions from *arable land* has only a minor effect on the total source in this zone. Low estimates for emissions from *animal excreta* (case 6) and *biomass burning* will correspond better with the 0°-30°S zone.

Higher N_2O emission from biomass burning than the a priori estimate may be realistic because various estimates from literature (see section 2) show that direct emissions from biomass burning may higher than the 0.6 Tg N yr^{-1} used in case 7. Most of the atmospheric NH_3 oxidation by OH to N_2O takes place in the tropics; this source was not accounted for in this analysis. Including the 0.3-1.2 Tg N yr^{-1} atmospheric N_2O formation would further increase the overestimation in the 0°-30°S zone.

Considering that emissions from *biomass burning* may be underestimated and that *atmospheric N_2O formation* was omitted, other sources may be overestimated. The major N_2O sources in the tropics are *soils under natural vegetation* and *oceans*; a small percentage error in these large sources may equal the total global emission from other, minor sources. Case 4b is roughly applicable in the 0°-30°N zone, and a high oceanic N_2O emission from this zone is probable (Butler et al., 1989; Law and Owens, 1990a). If the oceanic source estimate is correct for the 0°-30°S zone, the tropical soil emissions must be overestimated in this zone. However, our estimates of N_2O from tropical forest soils were roughly consistent with literature estimates (section 2). Hence, estimates for case 4b may be consistent with source estimates from inverse modeling for a major part of the world; however, this may be the net result of compensation of errors in different source estimates. The only major source in the 30°-90°S zone is the *oceanic emission*, due to the land area in this region being limited, the much smaller population than in the other zones, and because agricultural activity, fossil fuel use, industrial activity and mobile sources are relatively unimportant (Tables 4 and 6). This indicates that if the zonal distributions deduced by Prinn et al. (1990) are correct, the Antarctic Ocean will be a major source region. The oceanic estimate O2 for the 30°-90°S zone agrees with this result, although it is close to the upper end of the range of Prinn et al. (1990). Hence, O1 based on ocean surface area in each latitudinal zone and a mean global oceanic N_2O emission rate probably underestimates the 0°-30°N and 30°-90°S zones, while O2 is more consistent with Prinn et al. (1990).

The N input to rivers may be related to population density (Cole et al., 1993). The fraction of global population is 48%, 42%, 10% and 1% for the 30°-90°N, 0°-30°N, 0°-30°S and 30°-90°S zones, respectively (Lerner et al., [1988] updated with FAO [1991]). If the N_2O emission from *rivers and estuaries* correlate with N inputs, there may be additional important N_2O emissions in the 30°-90°N, 0°-30°N zones and less so in the 0°-30°S zone. This does not contradict the ranges of zonal distributions of Prinn et al. (1990).

The *industrial and chemical N_2O processes*, *explosives*, *medical use* of N_2O and effects of *N deposition* discussed in sections 2 and 3 suggest that most are located in the 30°-90°N zone. Given the uncertainty in the inverse modeling results, a higher emission from the 30°-90°N zone is not impossible.

From the data presented in this analysis we estimated that emissions from the European land area (excluding the CIS) may be 0.6-0.8 Tg N yr^{-1}, excluding N_2O from freshwater systems. This estimate is close to the European emission of 0.8 Tg N yr^{-1} deduced from atmospheric modeling (Prather, 1988). The ratio of northern hemispheric to southern hemispheric emissions deduced by Prinn et al. (1990) is 1.67. Cicerone (1989) found values of 1.5-1.9. Lower values have also been reported, e.g. a ratio of 1.4 calculated by Khalil and Rasmussen (1983). Our calculated ratio for case 4b of 1.5 suggests a somewhat higher southern hemispheric emission than Prinn et al. (1990).

4.6. CONCLUSIONS

The analysis presented has not reduced the uncertainty of annual N_2O emission estimates from individual sources. All sources include uncertainties, but uncertainties in emissions from soils and oceans, the largest global sources, have major implications for the zonal distributions.

All individual source estimates for the *tropics* are uncertain. Although there is consistency with estimates inferred from inverse modeling for the 0°-30°N zone, the total emission for the 0°-30°S zone is close to the upper end of the uncertainty range of Prinn et al. (1990). As emissions from biomass burning may be underestimated and atmospheric N_2O formation in the tropics was omitted, the N_2O emissions from tropical soils and oceans may be incorrect.

The N_2O emission from *temperate soils* calculated with the Kreileman and Bouwman (1994) model version is lower than with that of Bouwman et al. (1993). The models are almost identical and both are hampered by lack of measurement data in major parts of the world. It cannot be determined which inventory is more realistic and there may also be important omissions in both inventories. They do not account for soil N_2O sinks and episodic emissions that may occur in autumn, during thawing in early spring and from snow-covered soils in temperate regions.

No quantitative estimate of N_2O emissions from *coastal marine* and *freshwater systems, aquifers*, and soils with inputs from *N deposition* could be given. However, since these sources may be related to human population, industrial and agricultural activity, most associated emissions may come from the northern hemisphere. Other N_2O source candidates, such as industrial and chemical processes, medical and industrial use of N_2O and production and use of explosives, are located in the 30°-90°N zone. If these sources are significant, the emission from this zone may be close to or exceed the high estimate of 4.4 Tg N yr^{-1} made by Prinn et al. (1990).

For some sources the analysis has improved the understanding of the latitudinal distribution. The estimate of N_2O emission from *oceans* includes large uncertainties, caused primarily by scarcity of measurement data in large parts of the oceans. However, the global oceanic N_2O emission may be higher than assumed by Watson et al. (1992), and the 0°-30°N and 30°-90°S are major source regions.

Animal excreta form a significant global source of N_2O, with major source regions in the northern hemisphere. The global amount of N in animal excreta about equals the N applied as synthetic fertilizers, indicating a possible contribution to the global N_2O budget of the same order of magnitude. The global *biological N fixation* in agricultural systems may also be similar in magnitude to synthetic N fertilizer use (Duxbury et al., 1993).

The inventory AR2 for N_2O emission from *cultivated lands* calculated as the background emission plus fertilizer-induced emission does not contradict source estimates from inverse modeling. The major part of the N_2O induced by N fertilizer use stems from the northern hemisphere. Estimate AR1 calculated as the difference between N_2O emission from fertilized plots and unfertilized control plots for different fertilizer types underestimates global and regional N_2O emissions. In addition, the range of conditions represented in the available measurement data is too limited to calculate emissions by fertilizer type.

The method used to calculate enhanced N_2O emission following *forest clearing* accounted for a gradual decline of N_2O emission with increasing age of the clearing. This resulted in a lower global emission than the estimate by Matson and Vitousek (1990).

Large uncertainties must still be resolved. A number of unidentified sources may still exist and the knowledge of the strength of soil and aquatic sinks for N_2O is insufficient. Apparent consistency between our estimates of annual emissions and those from inverse modeling does not exclude the possibility of important uncertainties in individual source estimates. Better understanding of regional sources may be achieved by using 3-dimensional atmospheric models, which are dependent on the season, and where the observed seasonality of atmospheric N_2O with model results from prescribed source estimates are compared. First attempts by Taylor (1992) and Nevison (1994) have not resulted in improved source estimates. Better results with models are possible through the recent improvements in measurement techniques for N_2O (Hall et al., 1990) and extension of the existing monitoring network. The seasonal distribution of emissions is described in the inventories presented for soils under natural vegetation, oceans and biomass burning. For the other sources seasonal distributions will have to be approximated.

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Soil nitrogen budget before and after deforestation in the humid tropics of Costa Rica

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Chapter 5

SOIL NITROGEN BUDGET BEFORE AND AFTER DEFORESTATION IN THE HUMID TROPICS OF COSTA RICA

Abstract. The loss of nitrogen from a soil under pasture recently established after forest clearing in the humid tropics of Costa Rica was estimated from observed soil organic carbon loss and C/N ratios. The pathways for loss of the nitrogen mineralized shortly after forest clearing were analyzed by compiling soil nitrogen budgets for a deforestation sequence. Part of the nitrogen is lost via leaching while most of the nitrogen loss occurs through denitrification. The increased wetness in soils under pasture relative to forest soils caused by reduced interception and compaction may cause an increase in the N_2/N_2O ratio of the denitrified N. The nitrogen losses caused by deforestation may be an important source of pollution of the atmosphere and aquatic systems.

5.1. INTRODUCTION

Large amounts of soil organic carbon (SOC) and nitrogen (N) are lost from surface soils following land clearing in the tropics. Fires associated with clearing volatilize N from vegetation and surface litter (Ewel et al., 1981), and in the first years following clearing another major amount of N may disappear from surface soils (Nye and Greenland, 1964). The soil nitrate (NO_3^-) availability may increase in the initial period after forest clearing due to rapid decomposition of soil organic matter, and mineralization and nitrification of the soil organic N (Matson et al., 1987; Robertson, 1989). The NO_3^- formed may be lost via denitrification and leaching. Denitrification rates in recently cleared tropical forest soils show an increase relative to those in the undisturbed forest soils (Robertson and Tiedje, 1988). Nitrate leaching is also stimulated by clearcutting (Nye and Greenland, 1964; Vitousek and Matson, 1984; Mueller-Harvey et al., 1985; Matson et al., 1987).

In the humid tropics the influence of nitrification and denitrification on N losses may differ in a number of ways from that in temperate regions. First, soils dominated by variable-charge clays may conserve NO_3^- at the exchange sites (Robertson, 1989). Second, soil moisture, primary productivity and decomposition rates tend to be high in tropical humid forests. Therefore, soil gas diffusion can be depressed while oxygen demand is high, causing frequent anaerobic microsites. These conditions are favourable for denitrification. Where NO_3^- availability is high, such as in soils with anion exchange capacity, denitrification rates may be further enhanced.

Most of the soil organic matter loss generally occurs within a few years after forest clearing (Davidson and Ackerman, 1993). When easily decomposable soil organic matter is depleted, nitrification, denitrification and associated N_2O fluxes decrease to rates lower than in the undisturbed forest (Lamb, 1980; Robertson, 1984; Robertson and Tiedje, 1988; Keller et al., 1993). The analogy of the patterns of C loss after deforestation observed by Veldkamp et al. (1993) and N_2O and NO_x fluxes measured by Keller et al. (1993) suggests a close link between C and N cycling in the soils studied.

Here we will estimate the different loss terms of N for the sites studied by Veldkamp (1994) and Keller et al. (1993) in the Atlantic zone of Costa Rica. Based on observed losses of soil organic carbon (SOC) and C/N ratios, the N loss from a recently cleared forest soil will be estimated. Budgets of soil N will be compiled for a chronosequence from virgin forest to old pasture to assess the order of magnitude of denitrification and other N losses.

5.2. STUDY SITES

The study area is in the Atlantic lowlands of Costa Rica on the footslopes of the Turrialba volcano near Guácimo (10° 12'N, 83° 32'W). The soil parent material consists of laharic, fluvio-laharic and fluvial deposits, mostly of andesitic composition. The climate is humid tropical with a mean annual temperature of 26°C and mean annual rainfall between 3000 and 6000 mm. Precipitation exceeds evapotranspiration throughout the year.

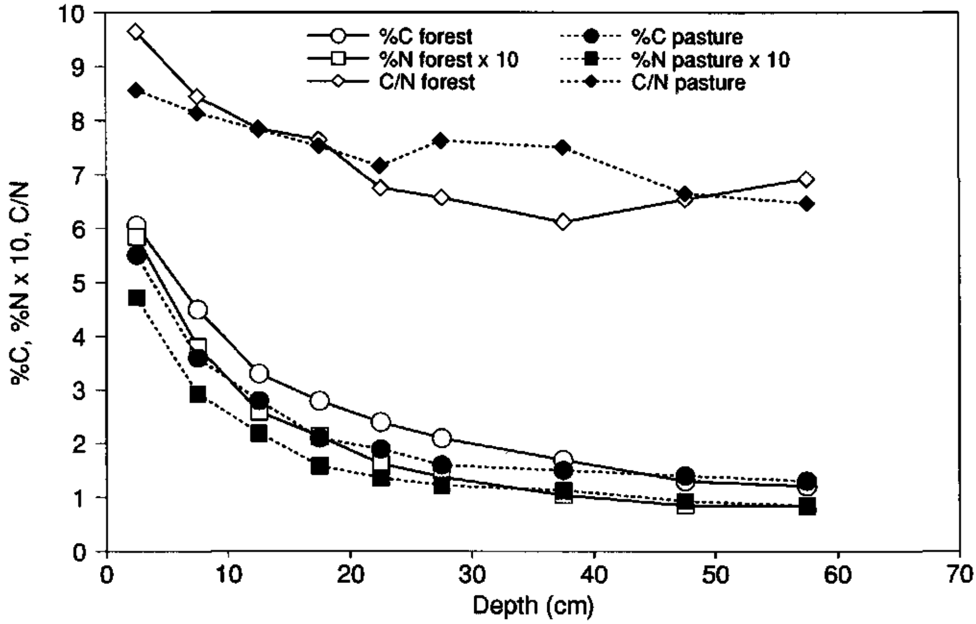

Figure 1. Carbon and nitrogen contents and C/N ratio for different depths in an andic Humitropept under tropical rain forest and under 18-year-old pasture.

The natural vegetation is tropical lowland rain forest, and the land use after forest clearing is mostly low input pasture. Clearing is generally done by small farmers without heavy machinery by removing valuable stems. Usually other stems are cut and left on the sites and stumps are not removed. If burning takes place, it is restricted to piles. Pasture grasses are generally planted directly after the clearing without further land preparation.

Three sites will be compared, including virgin tropical rain forest, and two sites with actively grazed Ischaeum ciliare dominated pasture, that had been cleared respectively 3 and 18 years before sampling. Net primary production (NPP) is 8.5 ton C ha^{-1} yr^{-1} for the forest and 5 ton C ha^{-1} yr^{-1} for the pasture sites (Veldkamp, 1994). The soils are andic Humitropepts, described in detail by Veldkamp (1994).

For this study C/N ratios for samples of soil, grass roots and above ground parts were calculated from total C and N determined with the Fyson Element Analyzer (CHN analyzer type 1108). For the study sites the data from Veldkamp (1994) on SOC dynamics were used as well as reported N_2O fluxes from Keller et al. (1993). We also used data from rain forest sites in the La Selva Biological Station (10° 26'N, 84°W) close to Guácimo with similar vegetation and soil and climatic conditions.

5.3. LOSS OF SOIL NITROGEN

Veldkamp (1994) observed a rapid loss of SOC of about 30 ton C ha^{-1} from the top 0-0.3 m of the andic Humitropept in the first 3 years after forest clearing (Table 1). This is about one-third of the total SOC, consistent with observed C loss caused by forest clearing from similar soils in La Selva (Reiners et al., 1994). Most of this rapid decrease is caused by decomposing litter (25.8 ton C ha^{-1}) and fine root biomass (3.7 ton C ha^{-1}) in the upper 0.3 m (Veldkamp, 1994). The C/N ratio of soil organic matter (excluding litter and roots) in the upper part of the soil under forest is slightly higher than in the pasture soil (Figure 1). Easily decomposable material has a higher C/N ratio than more humified material so that the C/N ratio of the soil organic matter decreases during decomposition, as illustrated by the data collected for the pasture sites. Fresh root and above ground material from different pasture sites (aged 1, 2, 3, 5, 10, 18 and 30 years) have a C/N ratio of 26 ± 4. Under pasture the C/N ratio of the soil organic matter is close to 9 in the top 0.05 m and 7 in deeper soil layers.

We used a C/N ratio of 23 for forest litter observed in La Selva with similar soils and vegetation (P. Vitousek, personal communication, 1992). In their review Vitousek and Sanford (1986) listed a range of C/N ratios of 30-70 for fine (< 6 mm diameter) roots of moist tropical forests. Here the C/N ratio of fine root biomass is taken equal to that of litter (Table 1). This may overestimate the N content, but as the observed mass of fine root material is only 10% of the total SOC loss, this does not have an important effect on the total amount of N mineralized. All litter and fine root biomass is decomposed in the initial 3-year period following clearing (Veldkamp, 1994), so that the N loss from this material is about 420 kg N ha^{-1} yr^{-1} (Table 1). Applying the C/N ratios of soil organic matter in the top 0.3 m of the soil under forest to the soil C-loss (excluding decomposition of litter and roots) reported by Veldkamp (1994) for this layer, yields an estimated loss of soil organic N from the andic Humitropept of about 190 kg N ha^{-1} yr^{-1} during the first 3 years following clearing (Table 1). This estimate accounts for an increase in C and N in the top 0-0.05 m layer of the soil (Table 1), possibly caused by humification of the litter and root biomass remaining after clearing. The total N mineralization rate for the recently cleared site of 610 kg N ha^{-1} yr^{-1} is in agreement with results of estimates based on in situ incubations presented by Matson et al. (1987) for sites in Turrialba, Costa Rica.

5.4. INPUT-OUTPUT BUDGET FOR SOIL NITROGEN

If N mineralization exceeds the uptake by plants and immobilization by microbes, there is an excess of mineral soil N available for nitrification and denitrification, producing N_2 and oxides of nitrogen (N_2O and NO_x). The excess mineral N may also leach from the rooting zone to deeper layers. The assumed annual loss of soil N is 0 for the forest soil and 610 kg N ha^{-1} yr^{-1} for the soil under young pasture (Table 2). The annual SOC loss 18 years after clearing amounts to about 0.5 ton C ha^{-1} yr^{-1} (Veldkamp et al. 1993) and the associated N loss rate is about 50 kg N ha^{-1} yr^{-1} (Table 2). The various soil N fluxes in Table 2 will be discussed briefly.

Table 1. Soil organic carbon (SOC) in the soil under forest (year 0) and under 3-year old pasture, the SOC loss, C/N ratio of soil organic matter in the original forest soil and the calculated N loss

Depth (cm)	SOC forest (ton C ha^{-1})	SOC pasture (ton C ha^{-1})	SOC loss (ton C ha^{-1} yr^{-1})	C/N ratio forest	N loss (kg N $ha^{-1}yr^{-1}$)
Litter + root biomass	29.5	0.0	9.8	23.3	420
0-5	15.9	16.7	-0.3	9.7	-30
5-10	11.0	9.6	0.5	8.4	60
10-15	8.3	6.4	0.6	7.8	80
15-20	6.3	5.8	0.2	7.6	20
20-25	5.5	4.7	0.3	6.8	40
25-30	4.6	4.2	0.1	6.6	20
Total	81.1	47.4	11.2		610

Table 2. Budget of soil N for andic Humitropept under rain forest, young pasture (3-year-old) and old pasture (18-year-old).

N flux	Forest	Young pasture	Old pasture
		(kg N ha^{-1} yr^{-1})	
N precipitation	5	5	5
N fixation	25	0	0
Animal excreta	0	80	80
Animal production	0	-10	-10
Litterfall & rhizodeposition	400	110	110
Plant production	-400	-200	-200
NH_3 volatilization	-4	-15	-10
Leaching	-15	-100	-8
Denitrification†	-11	-480	-18
Net loss of soil N	0	-610	-50

Negative numbers indicate losses of N from the soil system.

† *Denitrification balances the budget. Denitrified N is lost from the soil* and is, therefore, indicated as a negative flux.

- *N precipitation.* Precipitation inputs of N have been measured at two sites in Costa Rica. Hendry et al. (1984) observed a total N precipitation rate of 5 kg N ha^{-1} yr^{-1}, while Parker (1985) measured precipitation of inorganic N of 2 kg N ha^{-1} yr^{-1} in La Selva. Vitousek and Sanford (1986) listed N precipitation measurements of 2 - 21 kg N ha^{-1} yr^{-1} for tropical rain forest in various parts of the world. In the soil N budgets a rate of 5 kg N ha^{-1} yr^{-1} is assumed for all sites (Table 2).
- *N fixation.* It is likely that symbiotic N-fixation is more important in tropical forests than in most temperate forests (Boring et al., 1988). This explains why most tropical rain forests are not N-limited (Vitousek, 1984; Vitousek and Sanford, 1986). The heterogeneous spatial distribution of symbiotic N-fixing species in tropical forests makes it difficult to quantify a species contribution of fixed N over a given land area (Boring et al., 1988). In addition, symbiotic N fixation is strongly influenced by time-dependent biological and environmental factors. Nitrogen fixation may vary during successional development in response to changes in stand structure and N availability. It is generally low in mature stands and higher in early successional forests. Indirect evidence for high rates of N fixation from increasing soil N contents during early succession was given by Jenny (1950) and Greenland and Nye (1959). Sylvester-Bradley et al. (1980) estimated rates of symbiotic N fixation of up to 200 kg N ha^{-1} yr^{-1} in fertile varzea floodplain forest soils with high P contents, intermediate rates of 20 kg N ha^{-1} yr^{-1} in moderately fertile soils, and low rates of 2 kg N ha^{-1} yr^{-1} for infertile, P-limited soils in Amazonia.

Inputs from non-symbiotically fixed N in undisturbed tropical rain forests may be in the order of 15 kg N ha^{-1} yr^{-1} (Jordan et al., 1983). This is much higher than rates observed in temperate ecosystems, owing to high C availability combined with favorable climate (Boring et al., 1988).

In the absence of root nodules observed at similar forest sites in La Selva (W. Reiners, personal communication, 1991), and assuming that the original vegetation is a mature forest, the total N fixation rate is taken to be 25 kg N ha^{-1} yr^{-1} (Table 2). After clearing N fixation is assumed to cease.

- *Animal excreta and export of N via animal production.* Unfortunately no data are available on the grazing intensity, and N consumption and excretion by animals at the study sites. To assess the importance of grazing, we assumed a grazing intensity of two nondairy cows ha^{-1} for the young and old pasture sites. Based on an assumed growth rate of 0.5 kg per head per day the annual production is about 400 kg ha^{-1} yr^{-1}; for an N content of 25 g N kg^{-1}, the N accumulation is about 10 kg ha^{-1} yr^{-1} for the cattle in both the young and old pasture (K.W. Van der Hoek, personal communication, 1994). The N excretion by two nondairy cows is 80 kg N ha^{-1} yr^{-1}, based on an annual excretion of 40 kg N per head (Bouwman et al., 1994). The nitrogen consumption by the animals is, therefore, 90 kg N ha^{-1} yr^{-1}. These assumptions result in a nitrogen use efficiency of about 10%. The N in pasture production is about 200 kg N ha^{-1} yr^{-1} for the NPP of 5 ton C ha^{-1} yr^{-1} and the observed C/N ratio of 26. Hence, the N consumption by the animals would be about 50% of the NPP.

- *Litterfall, rhizodeposition and plant uptake of N.* It is assumed that neither the forest site nor the pasture sites have a net annual biomass increment. The N in litterfall and rhizodeposition in the undisturbed forest is assumed to equal the N uptake. For the forest NPP of 8.5 ton C ha^{-1} yr^{-1} and a C/N ratio of 23 (P. Vitousek, personal communication, 1992) these terms add to about 400 kg N ha^{-1} yr^{-1} (Table 2), in agreement with the mean amount of N in litterfall and root turnover for tropical evergreen forests presented by Vogt et al. (1986).

The pasture NPP of 5 ton C ha^{-1} yr^{-1} with an observed C/N ratio of 26 for root and above ground material, gives a total amount of about 200 kg N ha^{-1} yr^{-1}. Since the estimated consumption of N by the animals is 90 kg N ha^{-1} yr^{-1}, the flux of N in litterfall and rhizodeposition is only 110 kg N ha^{-1} yr^{-1} for the pasture sites. The NPP may have decreased in the old pasture as a result of loss of soil nutrients in the course of time. Because there are no observations of NPP as a function of nutrient losses, we assumed equal NPP, litterfall and rhizodeposition, and plant uptake for the young and old pastures.

- *Ammonia volatilization.* During grazing about 10% of the N excreted by the animals may be volatilized as NH_3 (Van der Hoek, 1994). The NH_3 loss from mineralized N is assumed to be 1% of the total N mineralization (including N from litterfall and rhizodeposition), based on the NH_3 emission rate for injected anhydrous ammonia (Asman, 1992).

- *N leaching.* From a regional study Salati et al. (1982) estimated that the N loss through leaching from soils in the Amazon Basin is 4-6 kg N ha^{-1} yr^{-1}. This is somewhat lower than the 18 kg N ha^{-1} yr^{-1} leaching loss measured by Parker (1985) for undisturbed forest in porous-cup soil water samplers at 70 cm depth, i.e. below a soil layer containing 90% of the root mass. Higher N leaching loss in La Selva may be caused by precipitation rates of close to 4000 mm yr^{-1} causing higher water percolation rates than in the Amazon Basin. In the year following forest clearing, the water percolation and the NO_3^- concentration strongly increased and the N leaching was about 100 kg N ha^{-1} yr^{-1} (Parker, 1985). As the conditions in La Selva are similar to those in the study sites, an annual NO_3^- leaching rate of 15 kg N ha^{-1} yr^{-1} is assumed for intact forest, and 100 kg N ha^{-1} yr^{-1} for the young pasture (Table 2). The total amount of N cycling through the old pasture soil is about half that in the undisturbed forest soil. Therefore, we assumed that the leaching from the old pasture soil is 50% of the N leaching from the forest soil (Table 2). These estimates exclude dissolved organic nitrogen compounds and N_2O in the solution, which may account for a small part of total leaching loss of N after forest disturbance (Bowden and Bormann, 1986).

- *Denitrification.* Knowledge about denitrification is less than about any other major N flow in humid tropical ecosystems. This is largely due to the difficulty of measuring N gas fluxes in soil. The available methods are technically difficult, laborious, provide results that are difficult to interpret, and require instrumentation which is not easily transported in rain forest environments. Robertson and Tiedje (1988) measured denitrification in primary forest sites using the acetylene inhibition technique on intact soil cores from sites in La Selva. The annual flux calculated from the measurements over a 14-month period amounted to 8-21 kg N ha^{-1} yr^{-1}, with highest rates at sites with andic Humitropepts. In recently cleared forest with soils classified as oxic Dystropepts the denitrification rates were up to a factor 3 higher, but these high rates persisted for a few months only. Denitrification losses of about 20 kg N ha^{-1} yr^{-1} were estimated for the Amazon Basin from a regional study (Salati et al., 1982).

In Table 2 denitrification balances the soil N budget. For the undisturbed forest site this resulted in a denitrification rate of 11 kg N ha^{-1} yr^{-1}, increasing to 480 kg N ha^{-1} yr^{-1} for the recently cleared forest site (Table 2). In the old pasture site the calculated denitrification rate remains somewhat higher than in the undisturbed forest.

5.5. CONCLUDING REMARKS

Rapid loss of 30% of the SOC following forest clearing found in this study is in agreement with the conclusions of a review of recent studies by Davidson and Ackerman (1993). Most of the SOC loss from the andic Humitropepts was in the form of litter and root biomass (Veldkamp, 1994). The estimated rate of mineralization of soil organic N associated with such soil organic matter loss in the young pasture is consistent with in situ incubation measurements. Most of the mineralized soil organic N in the recently cleared site is lost by denitrification. The soils have variable charge properties and can retain NO_3^- on their exchange sites, which may explain why only about 15% of the flush of mineralized soil N is lost from the young pasture soil by leaching. The other loss mechanisms, including the N export in the form of animal production and NH_3 volatilization, are probably relatively unimportant in the young pasture. In the old pasture site the N losses are much lower than in the young pasture site. The N losses by animal production, NH_3 volatilization, leaching and denitrification may be all of the same magnitude in the old pasture.

Keller et al. (1993) observed N_2O fluxes close to 6 kg N ha^{-1} yr^{-1} for the forest soils, while in recently cleared forest the N_2O fluxes were 35-50 kg N ha^{-1} yr^{-1}. The N_2O fluxes in the 18 year old pasture sites were about 2 kg N ha^{-1} yr^{-1}. At the sites under study there are no measurements of the molar N_2/N_2O ratio of denitrified N. At an N_2O emission rate of about 6 kg N_2O-N ha^{-1} yr^{-1}, and assuming a total denitrification rate of 8-21 kg N ha^{-1} yr^{-1}, as observed in similar forest sites, the molar N_2/N_2O ratio is 0.3-2.5. For the estimated denitrification rate of 480 kg N ha^{-1} yr^{-1} and N_2O fluxes of 35-50 kg N_2O-N ha^{-1} yr^{-1} observed in the young pasture site, the N_2/N_2O molar ratio would be about 10. Similar high N_2/N_2O molar ratios result from the budget for the old pasture. Such high ratios were also seen by Weier et al., (1993) who found highest molar N_2/N_2O ratios at the highest available C contents and at the highest soil moisture contents. The water percolation through the pasture soils is about a factor 1.7 higher than through the original forest soil (Parker, 1985). Hence, the pastures are much wetter and the anaerobicity may even increase by compaction of the topsoil as observed in the old pasture soils (Veldkamp, 1994; Reiners et al., 1994).

Extrapolation of the results to other sites is difficult, because the SOC and associated N losses depend on the amount of litter and root biomass remaining after forest clearing, soil properties and the productivity of the pasture. For example, the observed C loss was lower in soils of volcanic origin due to the strong stabilization of soil organic matter in Al-organic matter complexes; the C loss was also lower for pastures with higher productivity than for pastures with lower productivity (Veldkamp et al., 1993; Veldkamp, 1994). It is, therefore, difficult to assess the global implications of forest clearing. Bouwman et al. (1994) estimated that the enhanced N_2O emission for the current global rate of deforestation of 15 Mha^{-1} yr^{-1} (FAO, 1993) is about 0.4×10^{12} g N_2O-N yr^{-1}, which is about 10% of the annual atmospheric increase of N_2O in the atmosphere. Based on the estimates for leaching and the N_2/N_2O molar ratio of denitrified N, the total N loss via leaching and denitrification may be a factor 10 higher.

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Testing high resolution nitrous oxide emission estimates against observations using an atmospheric transport model

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Chapter 6

TESTING HIGH RESOLUTION NITROUS OXIDE EMISSION ESTIMATES AGAINST OBSERVATIONS USING AN ATMOSPHERIC TRANSPORT MODEL

Abstract. Global monthly estimates of N_2O emissions were used to prescribe a three-dimensional atmospheric transport model. The simulated N_2O surface concentrations in the northern hemisphere are in general agreement with atmospheric observations, about 1 ppb higher than in the southern hemisphere. The results suggest that N_2O concentrations over strong source regions in continental interiors are much higher than those over oceans. Simulated N_2O concentrations in the northern hemisphere vary strongly with the season, while measured concentrations oscillate strongly without seasonality. The modelled results for the southern hemisphere show much less seasonal variation, which is more consistent with atmospheric measurement data. The emission inventory does not account for soil N_2O consumption in N-limited temperate ecosystems, and N_2O emissions during autumn, thaw periods and from snow covered soils. These potential errors and possible underestimation of N_2O emissions from combustion sources may exaggerate the simulated seasonal trends. Most atmospheric N_2O monitoring stations are not well situated for verifying the simulated seasonality in atmospheric N_2O. There is only one continental site. In addition, the precision of N_2O measurements may not be adequate to distinguish seasonal trends, while atmospheric N_2O observations from different studies are sometimes contradictory.

1. INTRODUCTION

Nitrous oxide (N_2O) plays an important role in the atmospheric radiative balance and in stratospheric ozone chemistry. The atmospheric N_2O concentration is rising but the rate of increase is not constant. Trends have ranged from 0.5 ± 0.2 ppb yr^{-1} to 1.2 ± 0.1 ppb yr^{-1} over three-year periods, averaging 0.8 ± 0.2 ppb yr^{-1} over 1977-1988 (Khalil and Rasmussen, 1992). In 1992 the increase was about half that in the 1980s (Swanson et al., 1993). The concentrations of the other greenhouse gases with major biospheric sources and sinks, CH_4 and CO_2, also increased less in 1992. The average N_2O concentration in the northern hemisphere is shown to be about 1 ppb higher than in the southern hemisphere (Butler et al., 1989), reflecting higher emissions in the northern than in the southern hemisphere. Many major and minor sources have been identified, yet there is considerable uncertainty in the source strengths. Part of the uncertainty arises from the paucity of measurements of N_2O fluxes. Another problem stems from the difficulty in extrapolating measurements of biogenic fluxes from soils and aquatic sources because of their extreme spatial and temporal heterogeneity. For the anthropogenic sources political, economic and cultural factors are major uncertainties in making extrapolations. Khalil and Rasmussen (1992) recently presented a global N_2O budget, indicating that the uncertainty for most N_2O sources amounts to at least a factor of 2.

Seasonal variations of emissions have been predicted using global models used to assess N_2O emissions from soils (Bouwman et al., 1993) and oceans (Nevison, 1994). Since soils and oceans are the dominant N_2O sources in many parts of the world (Bouwman et al., in press), Taylor (1992) speculated that there may be similar seasonality in the atmospheric N_2O concentration. Emissions of N_2O from soils and aquatic systems may change from year to year along with climate variability. In addition, oceanic emissions are influenced by El Niño Southern Oscillation (ENSO) events, with lower global emission in ENSO years than in non-ENSO years (Butler et al., 1989). This may explain some of the observed variability in the atmospheric N_2O increase.

An indirect approach to assess sources and sinks of N_2O involves the use of atmospheric transport models that describe N_2O transport and destruction. This can be done by forward model simulations or by inverse methods. In forward simulations predicted concentrations - based on specific hypotheses for the sources and sinks of a trace gas - can be compared with observations. This allows surface fluxes to be determined that give the best correlation with observations. Studies of the CH_4 cycle using forward runs of three-dimensional atmospheric models by Fung et al. (1991) and Taylor et al. (1991) show that constraints imposed by atmospheric observations can be satisfied on the basis of different assumptions for the CH_4 sources. Emissions of CH_4 from soil and land-use related sources could not be determined uniquely because of lack of flux measurements and atmospheric observations (Fung et al., 1991).

Inverse techniques use observations of atmospheric concentrations as the input to solving the problem of producing flux estimates. For N_2O, the inverse method was recently applied in a simple box model by Prinn et al. (1990). The inverse technique has been used for $CFCl_3$ in a three-dimensional atmospheric chemical transport model (Hartley and Prinn, 1993). Inverse runs can also be used to optimally locate future additional observation sites based on a specified hypothesis for the sources and sinks of the trace gas to be tested.

Currently there are 10 monitoring stations wherever N_2O is measured in the world. These stations are part of the Atmospheric Lifetime Experiment and its successor the Global Atmospheric Gases Experiment (ALE/GAGE) (Prinn et al., 1994) and Climate Monitoring and Diagnostics Laboratory (CMDL) (Montzka et al., 1992) networks as shown in Table 1. They were established in locations where the air is thought to be thoroughly mixed, away from major source regions in order to establish global trends in atmospheric trace gas concentrations. The mixing of air smoothes the signal from individual sources (Keeling et al., 1989). Most stations are located on remote islands, in coastal areas and the South Pole; only one station (Niwot Ridge) is in the interior of the North American continent (Table 1). In the initial phase of the ALE measurements (1978-1984) four air measurements were made per day with in situ gas chromatographs (GC), while the partly overlapping GAGE measurements (1981 to present) have consisted of 12 measurements per day. The precision of the N_2O analysis was 0.35% ($\pm$ 1 ppb) for GAGE and 0.13% (0.4 ppb) for ALE observations. Flask samples are taken once per week at the CMDL stations and Niwot Ridge and analyzed with a manually operated GC. In addition, continuous GC systems measuring the atmospheric N_2O concentration every hour with a precision of 0.15% or < 0.5 ppb (Hall et al., 1990) were installed at CMDL stations between 1987 and 1989. For the CMDL stations and Niwot Ridge only the data from the flask samples have been published in tabular form (Montzka et al., 1992).

Table 1. Sites and coordinates of monitoring stations for atmospheric N_2O

Site	Country	Coordinates	Type†
ALE/GAGE			
Adrigole/Mace head	Ireland	52/53°N, 10°W	GC
Cape Meares	Oregon	45°N, 124°W	GC
Ragged Point	Barbados	18°N, 65°W	GC
Point Matatula	American Samoa	14°S, 171°W‡	GC
Cape Grim	Tasmania	41°S, 145°E§	GC
CMDL			
Alert	Canada	82°N, 62°W	F
Point Barrow	Alaska	71°N, 156°W	F+GC
Niwot Ridge	Colorado	40°N, 106°W	F+GC
Mauna Lowa	Hawaii	20°N, 156°W	F+GC
South Pole		90°S	F+GC

† F = flask sampling; GC = in situ gas chromatographic determination

‡ Also a CMDL collection site, with flask sampling and in situ GC

§ Also a CMDL flask sampling site

In this study we will test hypotheses concerning temporal and spatial patterns of N_2O fluxes. We will use a priori estimates of N_2O sources proposed by Bouwman et al. (in press) in an adapted version of the three-dimensional lagrangian tropospheric tracer transport model for N_2O described by Taylor (1991). We will describe a five-year forward model run and compare the simulated latitudinal gradients and seasonality of N_2O in the troposphere with measurements of N_2O concentrations recorded at a number of monitoring stations.

2. A PRIORI ESTIMATES OF EMISSIONS

Bouwman et al. (in press) compiled grid-based inventories including all known important N_2O sources; the a priori estimates used here represent the inventory with the smallest overall deviation from estimates inferred from inverse modelling presented by Prinn et al. (1990) for 4 latitudinal zones. Some inventories were originally produced on a monthly basis. For some other sources the annual inventories had to be converted to monthly distributions for this study (Table 2):

- *Soils under natural vegetation and grasslands.* Monthly N_2O emissions are from Kreileman and Bouwman (1994).
- *Fertilized arable lands.* The emission from arable lands includes 'background' emissions and the fertilizer-induced emission. The background emission is the N_2O release from unfertilized arable fields calculated on a monthly basis with the Kreileman and Bouwman (1994) model. The fertilizer-induced emission of 1.25% of the synthetic N-fertilizer application is an annual estimate. Monthly distributions for the fertilizer-induced emission were based on the start and length of the growing season in each agricultural grid cell according to Leemans and Van den Born (1994). The growing season is the period during which temperature and moisture conditions are favorable for crop growth. For growing periods shorter than 180 days, application of fertilizers is assumed to take place at the start of the season, with most N_2O losses (60%) occurring in the first month after fertilizer application, another 20% in the month thereafter and the remaining 20% distributed in equal parts over the remaining 10 months of the year. For growing seasons of 180 - 300 days (allowing for more than one crop and with assumed split fertilizer applications), 80% of the fertilizer-induced emission is assumed to occur as a constant flux during the growing period and the remaining 20% is distributed in equal portions over the rest of the year. For growing seasons with favorable cropping conditions lasting 300-365 consecutive days, the monthly fertilizer-induced emission is assumed to be equal for the whole year.
- *Animal excreta.* The annual N_2O emission induced by animal excreta is given a monthly distribution identical to the emissions from soils under natural vegetation. We assumed that animal excreta have a long-lasting effect (Van Faassen, 1994, personal communication) and continue releasing N_2O during dry and cold periods. Therefore, months with zero "natural" soil emission are assigned a portion of the N_2O emission from excreta equal to the month with the lowest non-zero natural soil flux for that grid cell.
- *Biomass burning.* The temporal distribution for burning during deforestation and savanna burning is taken from Hao et al. (1990), who presented periods of most probable burning activity.
- *Agricultural waste burning.* Agricultural waste burning is assumed to occur at the end of the growing seasons of less than 300 days, with 25% occurring during the last month of the growing period, 50% in the consecutive 30 days and 25% in the four weeks thereafter. For growing seasons longer than 300 days the waste burning is assumed to be constant throughout the year.
- *Post-clearing effects on N_2O emission.* Enhanced N_2O fluxes from recently cleared forest land are assumed to have a monthly distribution identical to the "natural" soil fluxes from Kreileman and Bouwman (1994).
- *Fossil-fuel and biofuel burning, and industrial sources.* Annual emissions from combustion of fossil fuels and biofuels and nitric acid production, are distributed spatially on the basis of the

Table 2. A priori global estimates for N_2O sources

Source	Annual N_2O emission (Tg N yr^{-1})†	Time scale‡	Major criterion for producing monthly N_2O fluxes
Soils under natural vegetation & grasslands	5.7	M	__ §
Arable lands	1.0	M	__ §
N-fertilizer use	1.0	A	Growing seasons
Animal excreta	1.0	A	Natural soils
Biomass burning	0.1	M	__ §
Agricultural waste burning	0.1	A	Growing seasons
Post-clearing enhanced soil flux	0.4	A	Natural soils
Fossil fuel combustion & traffic	0.3	A	Constant flux
Biofuel combustion	0.1	A	Constant flux
Industry	0.5	A	Constant flux
Oceans	3.6	M	__ §
Total sources	13.6	M	
Stratospheric loss	10.5	M	
Atmospheric increase	3.1	M	

† Tg = teragram; 1 Tg = 10^{12} g

‡ M = monthly emission inventories; A = annual emission inventory

§ Monthly estimates are included in the inventory.

database of population densities developed by Lerner et al. (1988). Adipic acid (AA) production was distributed on the basis of the coordinates for the major plants worldwide, except those in China where production was assumed to take place in Beijing. The inventory does not include production in the Commonwealth of Independent States (CIS) and Eastern Europe because of the uncertainty regarding both the volume and location of production. Emissions from combustion of fossil fuels and biofuels, and those from industrial sources, are assumed to be constant throughout the year.

- *Oceans*. The temporal distribution of N_2O emission from the oceans is the result of the stability dependent model presented by Nevison (1994), who produced monthly emissions on the NCAR-CCM2 grid of about 2.9° x 2.9° longitude – latitude. These grids were converted to 1° x 1° grid cells, resulting in a slightly lower global annual emission (3.6 Tg N_2O-N yr^{-1}) than the original 3.8 Tg N yr^{-1} due to scale differences.

3. ATMOSPHERIC TRACER TRANSPORT MODEL

The model used in this study is based on the Lagrangian tracer modelling approach described in Taylor et al. (1991) and Taylor (1989; 1992). This model uses a stochastic Lagrangian advection scheme, which conserves the mass of tracer by definition to move 1,000,000 air parcels representing a known mass of a tracer gas in air according to wind fields on a 2.5° x 2.5° longitude – latitude grid with seven vertical levels at 1000, 850, 700, 500, 300 and 100 hPa. Each wind field has a mean and a time-variable component. Wind fields are based on the European Centre for Medium Range Weather Forecasting (ECMWF) observational analysis fields reported on a 2.5° x 2.5° grid. These fields include mean vertical motion, while mixing processes are modeled by including a time-variable component in the computation of each air parcel velocity. The surface flux of a trace gas is added or removed from air parcels in the lowest vertical level grids. Concentration fields are presented by translating the air-parcel coordinates to the model grid coordinates. The time step is 24 hours. Loss of N_2O to the stratosphere and its return to the troposphere is represented by a simple linear model relating losses to latitude with no net flux occurring at the poles and a maximum flux to the stratosphere at the equator. The initial conditions were based on the observed concentration and latitudinal gradient in the late 1980s, chosen to arrive at a present-day mean concentration of about 310 ppb and at an mean annual increase of 0.8 ppb.

4. RESULTS

The global inventory of annual N_2O emissions (Figure 1) indicates that most of the N_2O stems from the northern hemisphere, with major source regions in the tropics. At southern temperate latitudes the Antarctic Ocean forms an important source region. The temporal distribution of N_2O emissions in this inventory is spatially variable. The monthly average latitudinal N_2O emission rates for all sources (Figure 2a) are highest in the northern temperate regions in the period April-August, while in the tropics the seasonality is much lower. In southern temperate latitudes the maximum fluxes are in July-November. Comparison of Figures 2a and 2b shows that the maximum N_2O emission from the Antarctic Ocean is in September. The maxima around May in the northern temperate regions are also caused by oceanic emissions. The seasonal variation in N_2O emission in the northern hemisphere is greater than in the southern hemisphere. A number of point sources in the inventory represent adipic acid factories, such as in the southern U.S.A., and large cities with assumed industrial activities and high emissions from traffic and combustion. Net annual uptake of N_2O in the oceans occurs in a number of regions as shown in Figure 1; e.g. October-November at 70-80°N (Figure 2a). Uptake of N_2O by ocean water in other regions can be inferred from comparing Figures 2a and 2b (e.g. March-May at 35-40°S). We refer the reader to Nevison (1994) for the location and seasonality of oceanic uptake.

We will present the results of a five-year model run for the lowest surface level at 1000-925 hPa, and discuss latitudinal gradients and the seasonality in concentrations for the monitoring sites listed in Table 1. The mean modeled northern hemisphere mixing ratios were approximately 1 ppb higher than those for the southern hemisphere (Figure 3a). The latitudinal variation changed with time and showed smaller differences and highest concentrations in the tropics, for instance, in January (Figure 3b); and maximum differences and highest concentrations were predicted in northern temperate latitudes in June (Figure 3c). Simulated continental concentrations over strong source regions were generally in the order of 310-315 ppb (Figures 4a and 4b for January, and 3c and 3d for July). Local N_2O concentrations reached 316 ppb over humid tropical forested areas with high soil fluxes throughout the year (Figure 2a). The simulated N_2O concentrations reached a maximum over the European continent, North America and East Asia in July and August (Figures 4c and 4d). All modelled results for stations in the temperate zones of the northern hemisphere showed a similar pattern, with somewhat higher summer than winter concentrations. The simulated N_2O concentration in Alert (North Pole) showed a minimum in March and a maximum in August-September (Figure 5a). The seasonality in prescribed emissions for the northern hemisphere with maxima in July resulted in predicted concentrations for Point Barrow (Alaska) and Cape Meares (Oregon) as in Figures 5b and 5d. These concentrations are similar to the seasonal trend in Alert with somewhat larger oscillations. In Ireland the predicted values indicate a minimum early in the year and a maximum beginning somewhat earlier in the year than in the other temperate northern hemisphere stations.

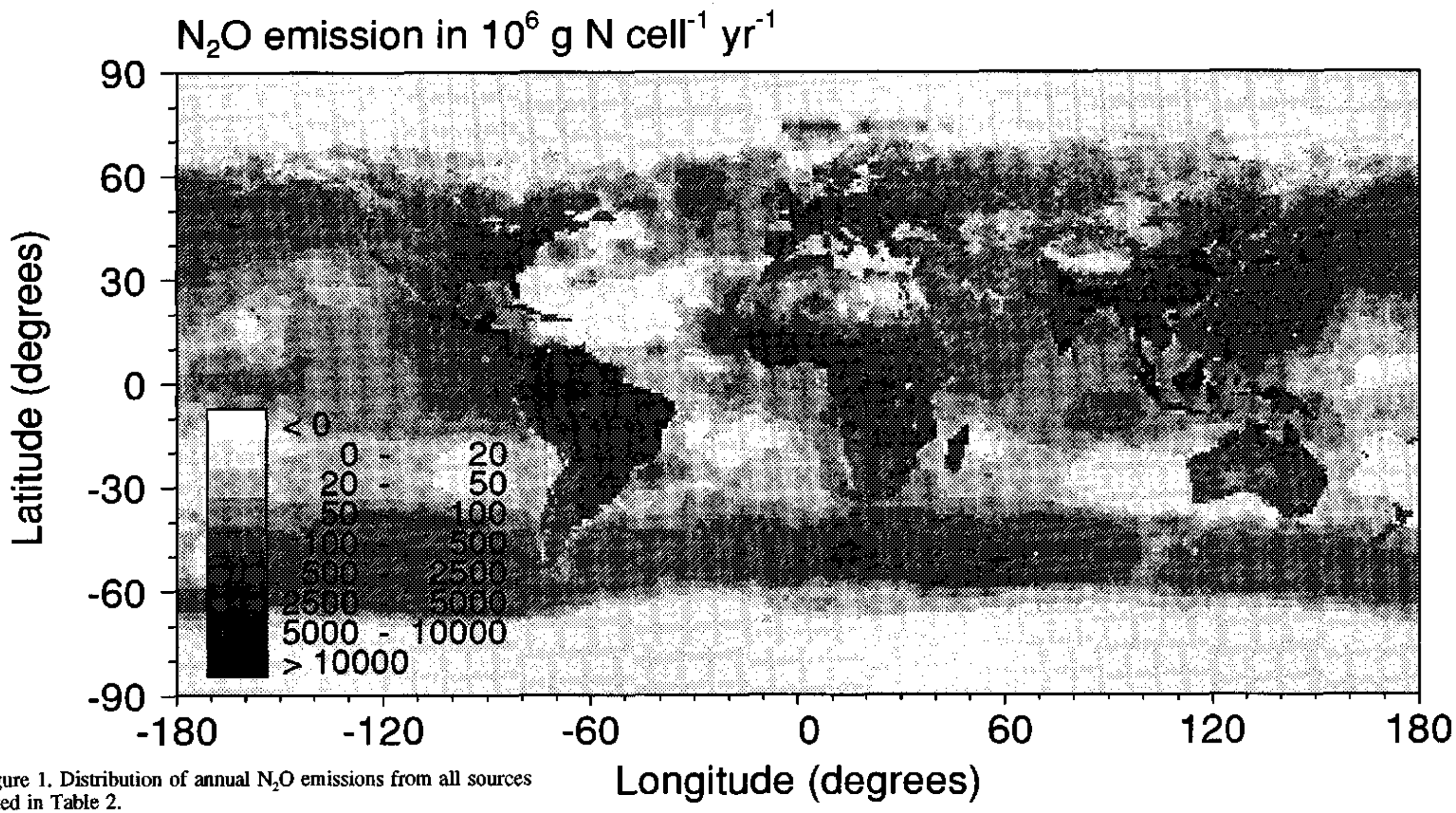
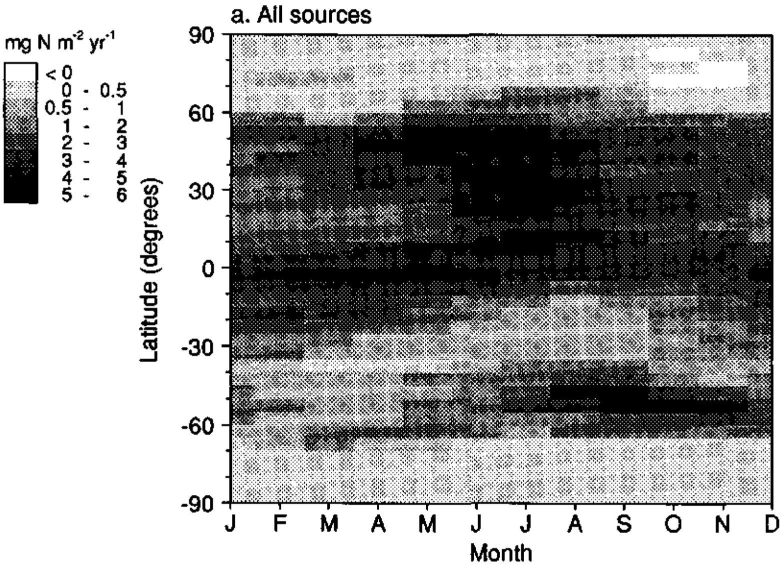
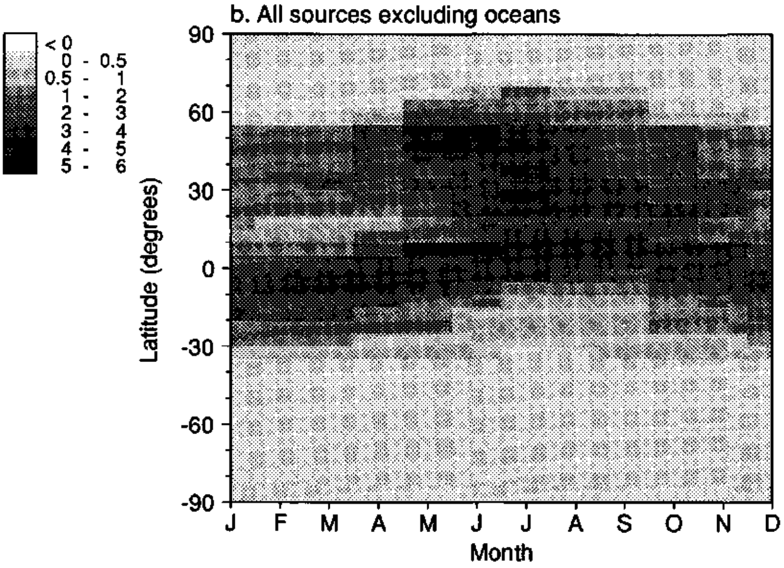


Figure 1. Distribution of annual N_2O emissions from all sources listed in Table 2.

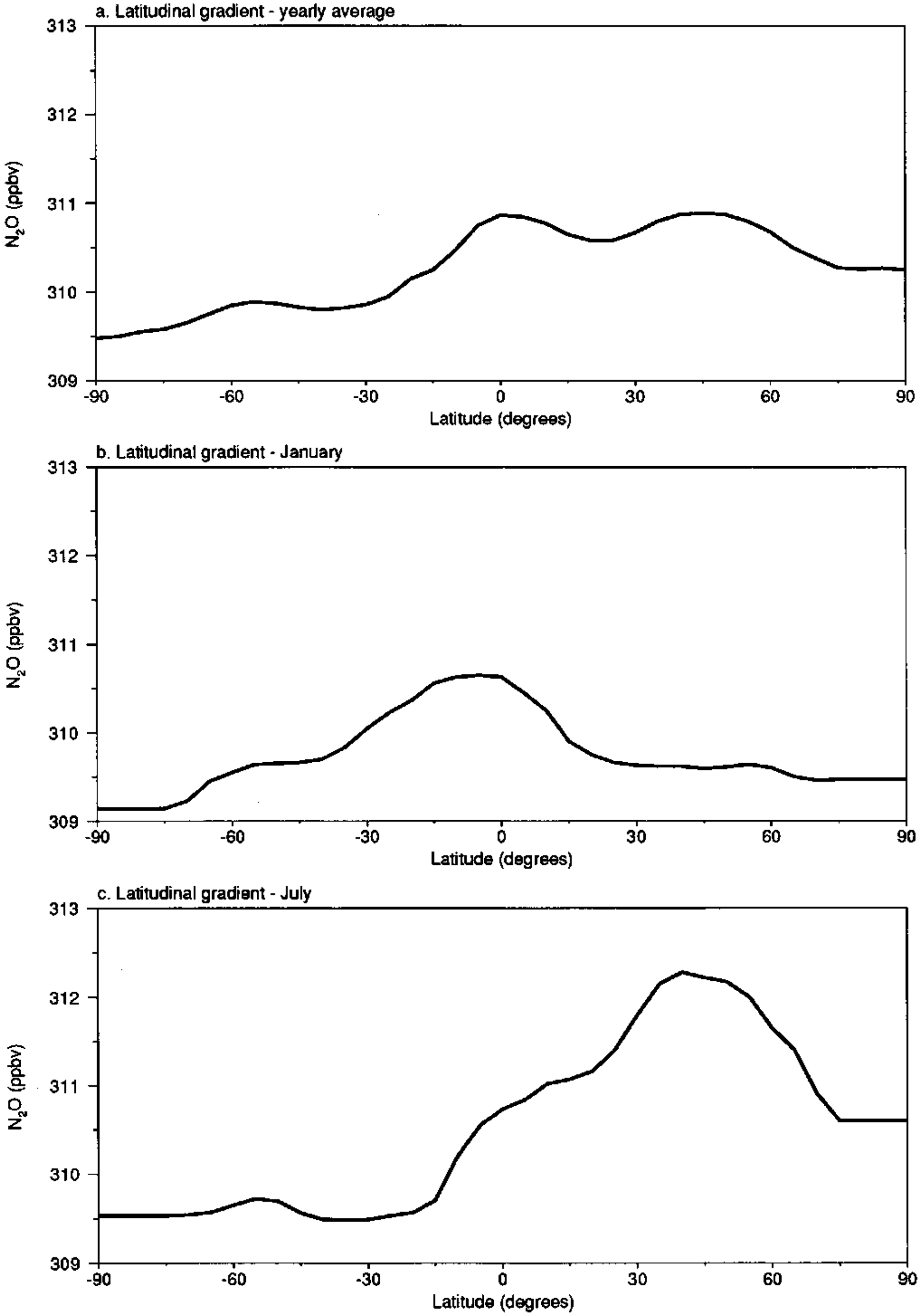


a. All sources



b. All sources excluding oceans

Figure 2. Seasonal and latitudinal distribution of emissions of N_2O for all sources (a) and all sources excluding the oceanic emissions (b).

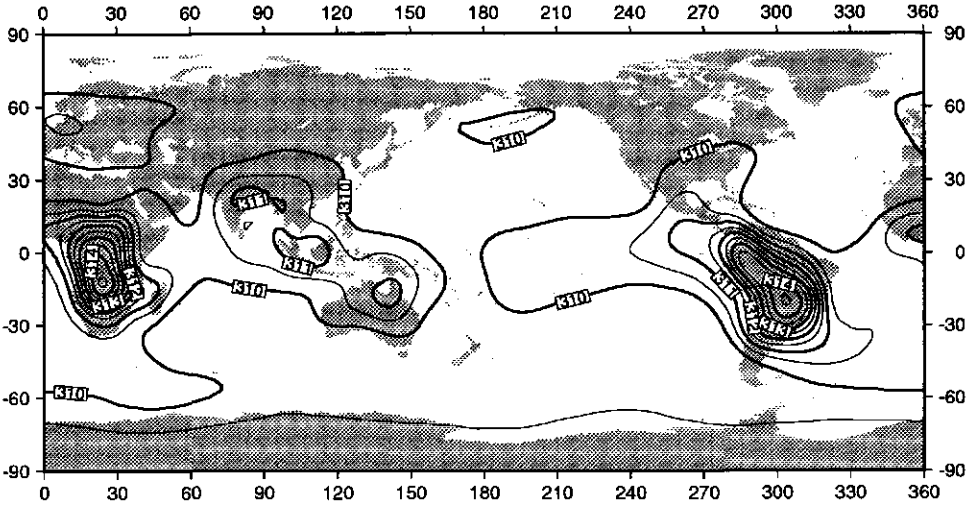

a. Latitudinal gradient - yearly average

b. Latitudinal gradient - January

c. Latitudinal gradient - July

Figure 3. Simulated latitudinal gradient of N_2O concentrations; yearly average (a); gradient in January (b); gradient in July (c).

a. January



b. July

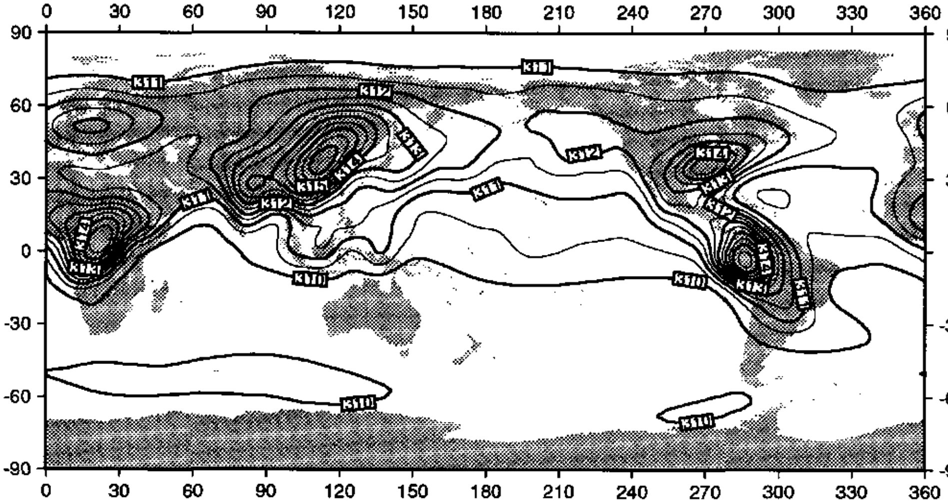


Figure 4. Simulated distribution of N_2O concentration for January (a) and July (b).

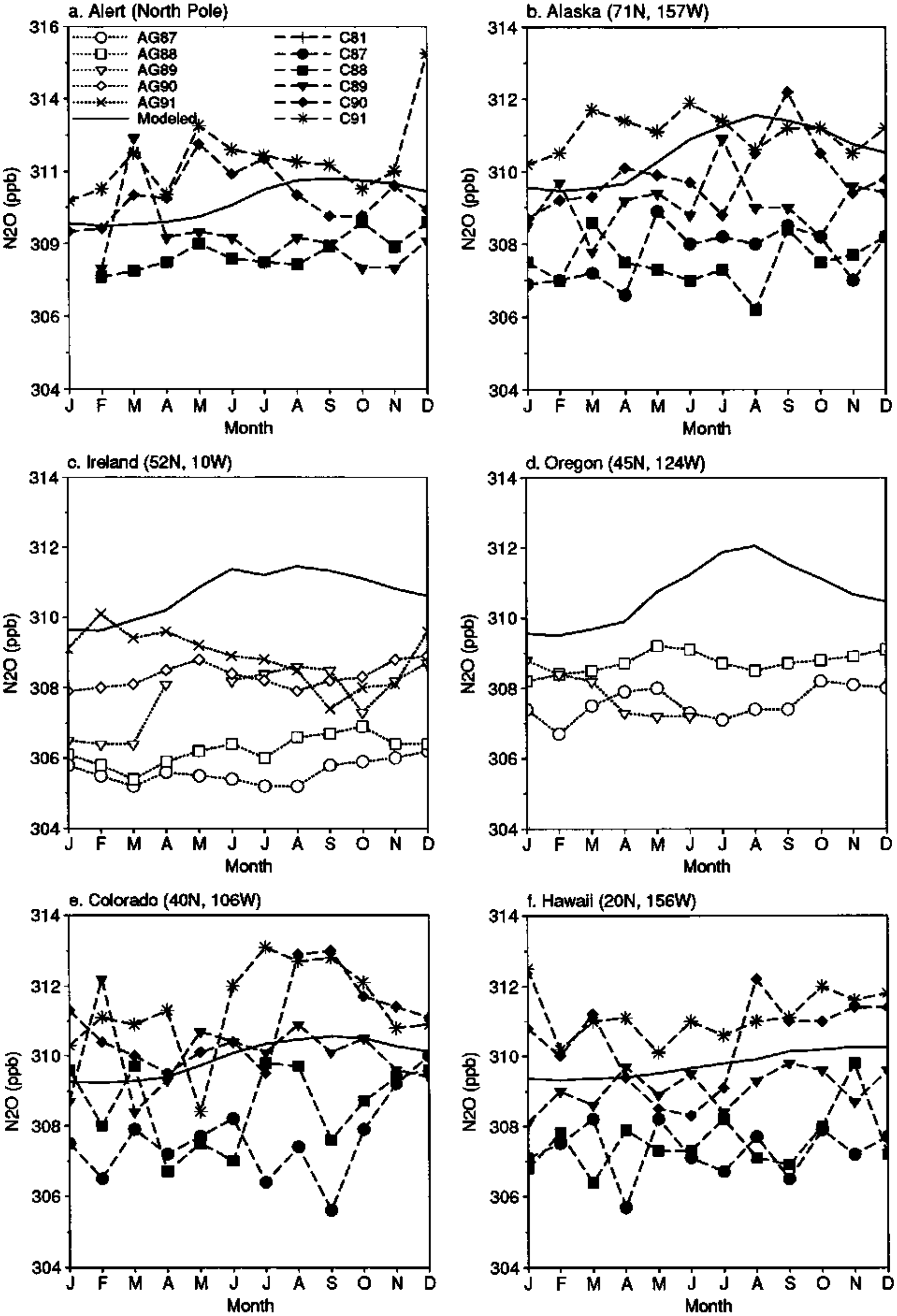


Figure 5. Monthly mean N_2O concentration for 10 monitoring stations and different years. AG = ALE/GAGE high frequency (4 - 12 measurements per day) real-time in situ gas chromatographic measurements (not corrected for pollution events) (Prinn et al., 1994). C = CMDL measurements in weekly flask samples (Montzka et al., 1992). Simulated monthly concentrations at 10 different monitoring stations presented in Table 1 are also shown.

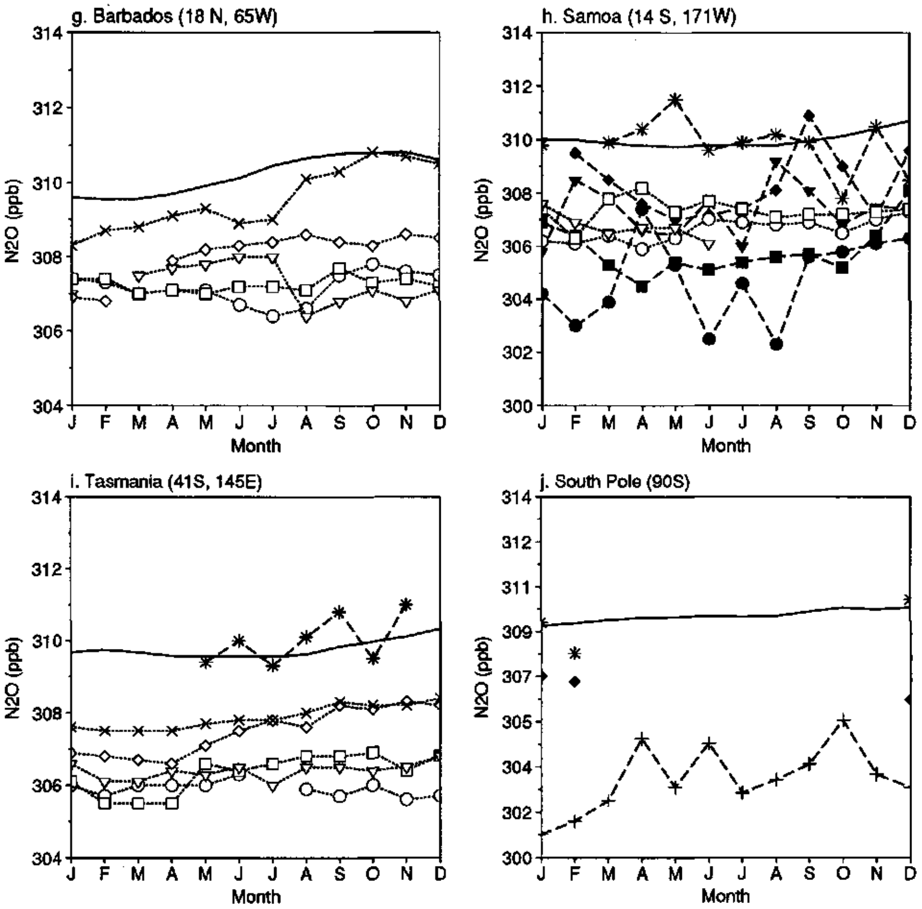


Figure 5 (Continued)

The modeled N_2O concentration for Niwot ridge, Colorado (Figure 5e), located in the interior of the North American continent, also peaked in July, but the oscillation was about half that in Point Barrow and Cape Meares. Mauna Lowa (Hawaii) showed no strong oscillations in the N_2O concentration (Figure 5f). The average oscillation for the northern hemisphere ocean was 1.2 ppb, corrected for the trend. The predicted seasonal variation for the southern hemisphere stations was much smaller than for the northern hemisphere stations. The southern hemisphere has prescribed emissions that are high in December-June in the tropics (Figure 2a). In July-September the maximum emissions in the 30-90°S zone are caused by the oceanic emission (Figure 2a-b). The simulated concentrations for Barbados, Samoa and Tasmania were slightly higher in September-January in accordance with the temporal distribution of sources (Figures 5g, 5h and 5i). At the South Pole (Figure 5j) the modelled results showed no seasonal trend. The average oscillation for the southern hemisphere ocean corrected for the trend was 0.1 ppb.

5. DISCUSSION

The simulated semi-hemispheric difference of about 1 ppb is consistent observations by Prinn et al. (1990), Weiss (1981) and Butler et al. (1989). This suggests that the overall latitudinal distribution of sources is correct. The concentrations obtained over continental regions can only be compared with the station in Colorado. In 1991 the N_2O concentration reached 313 ppb in July-September (Figure 5e), confirming the simulated high continental concentrations. The overall concentrations for the monitoring stations in Figure 5 are consistent with the most recent CMDL flask sample observations from 1990-1991. The simulated seasonality of concentrations (Figure 3b and 3c) will be compared with the data for the 10 monitoring stations listed in Table 1.

- The predicted seasonal concentration for Alert with a maximum in September contradicts the observations (Figure 5a). The measurement data for Alert from Montzka et al. (1992) indicate strongly fluctuating patterns, with a peak in March in 1989, 1990 and 1991, a high level in May in 1990 and 1991 and a low level in October. In December 1991 a very high concentration (315 ppb) was observed.
- Similar to the simulated peak in June-August for Alaska, the CMDL observations for 1991 showed a peak in June (Figure 5b). The CMDL flask measurements for 1990 and the 1987-1991 CMDL average shows a maximum in September in 1990. The CMDL measurements for 1987-1989 have a strongly oscillating pattern with no clear seasonality. Surprisingly, the in situ measurements for Alaska (Figure 5.4 in Swanson et al., 1993) show late summer minima.
- The simulated concentrations for Ireland can be compared with published ALE/GAGE observations for 2 years only. Contrary to the predicted May-August maximum the observations for 1987 in Adrigole, Ireland, show weak, if any, seasonality (Figure 5c).
- Modeled concentrations have a strong peak in July-August at the Oregon station, while the ALE/GAGE observations do not have a discernible seasonal pattern (Figure 5d).
- Similar to our results, the CMDL concentrations in flask samples in 1990-91 for the Colorado station show maxima in July-September (Figure 5e), while the average for 1987-1991 peaks in August. In contrast, the CMDL observations in 1987 peak in December, and those for 1988 have maxima in March and July-August. The results from the in situ GC measurements show summer minima (Figure 5.4 in Swanson et al., 1993) in 1990 and 1991, and no seasonal trend in 1992.
- The flask sample concentration measurements for Hawaii show a very complicated pattern with no clear seasonality. The simulated results show a minimum early in the year and an increase that begins in August-September (Figure 5f). The in situ GC measurements for Hawaii show no seasonal trend (Figure 5.4 in Swanson et al., 1993).
- Published observations for the Barbados station are available from ALE/GAGE. These data

have no seasonality, and do not confirm the modeled September maximum, although in some years there seems to be an increase in August-September. However, in 1985 two peaks were observed, and in 1986 a strong minimum in May.

- Likewise, a disagreement between simulated results and measured concentrations was found for Samoa (Figure 5h), where modelled results have no observable seasonality, whereas the flask sample measurements show strong oscillations lacking seasonal trends. The in situ GC measurements of both ALE/GAGE (Figure 5h) and those of CMDL (Figure 5.4 in Swanson et al., 1993) show much less variability and lack seasonality.
- At the station in Tasmania (Figure 5g) the ALE/GAGE measurements lack a clear seasonality with weaker oscillations than the other stations. The simulations indicate a rise that seems to start in September, and a minimum in March-July, prescribed by the oceanic emission from mainly the Antarctic Ocean. The CMDL results for 1991 for Tasmania show stronger oscillations than the ALE/GAGE data.
- The model predicts a similar pattern for the South Pole station with a maximum in September and a minimum on June-July, but with small oscillations (Figure 5j). In contrast, seasonal trends are absent in the CMDL flask sample observations, although the oscillations are high. Oscillations of the in situ CMDL GC measurements are much lower with no seasonal trend (Figure 5.4 in Swanson et al., 1993).

There may be a number of reasons for inconsistencies in seasonal trends between model results and atmospheric observations:

(i) *Incorrect assumptions in the emission inventories.* The uncertainty of emission estimates has been discussed by Bouwman et al. (in press); in the a priori emission inventories there are some evident omissions of the N_2O cycle that may cause exaggeration of the seasonal variability:

- *Soil N_2O consumption.* Net annual consumption of N_2O in soils has been reported by several authors. Ryden (1981; 1983) observed net annual uptake of N_2O by grassland soils when conditions were conducive to microbial reduction of N_2O. Uptake occurred when NO_3^--N was essentially exhausted and the sites had high moisture contents and high temperatures. Net annual uptake was also observed by Castro et al. (1993) in soils under spruce-fir forest in New Hampshire and Vermont. The uptake was attributed to consumption of N_2O by denitrification under conditions of low nitrification leading to low NO_3^- concentrations. In many other studies episodical soil N_2O uptake was observed (e.g. Keller et al., 1983; Bowden et al., 1990). In general tropical forests are less limited by nitrogen than temperate forests (Bouwman et al., 1993). In cold temperate and boreal forests preferential immobilization of N occurs in the forest floor and its associated decomposers (Vogt et al., 1986). Immobilization and slow mineralization of N and other soil conditions unfavorable to nitrification (such as low pH) in temperate forests may be conducive to N_2O consumption in wet and warm periods. Consumption of N_2O may also occur in wetlands with periodic or continuous anaerobic conditions (Bouwman et al., 1993). Uptake of N_2O may have important repercussions for the distribution and seasonality of N_2O in the atmosphere. In our model N_2O consumption in spring-autumn would lead to a lower amplitude of the seasonal trend in the northern temperate zones.
- *Episodic soil N_2O emissions.* Goodroad and Keeney (1984), Schmidt et al. (1988), Bowden et al. (1990), Brumme and Beese (1992) and Castro et al. (1993) reported similar general seasonal trends in temperate forests, with high emission rates in spring and summer. The monthly emission inventory used in this study was based on relations between temperature and N_2O fluxes and soil moisture and N_2O fluxes, leading to highest fluxes in spring and summer in temperate forests. However, elevated autumn N_2O fluxes in temperate forests have also been reported (Duxbury et al., 1982; Schmidt et al., 1988). Dörsch et al. (1993) observed that N_2O

emissions from arable fields during thaw periods contribute significantly to the annual flux. Sommerfeld et al. (1993) showed that N_2O formation occurs at low temperatures below 0°C, and observed that N_2O formed may be emitted from the snow cover. These episodical emissions are probably lower than the spring and summer emissions, but on the global scale the vast areas of warm and cold temperate and boreal ecosystems may account for a significant contribution. These phenomena were not included in the simple soil model used to produce the emission inventories for soils under natural vegetation and agricultural soils. The N_2O release during winter, early spring and autumn may cause a decrease in amplitude of the seasonal emission pattern. This will lead to reduced modeled seasonal variability in the northern hemisphere atmosphere.

- *Consumption of N_2O in aquatic systems.* The model of Nevison (1994) that was used to estimate oceanic emissions predicts net annual uptake of N_2O in some parts of the ocean, but there is considerable uncertainty in the inventory. Kim and Craig (1993) suggested the possibility of a large tropospheric flux of N_2O to and from the ocean, resulting in a residence time of N_2O comparable to that of CO_2 (~7 years). It is not clear how such intense fluxes might influence the simulated seasonality.
- *Underestimation of combustion-related and industrial sources.* There is still considerable uncertainty in the estimates for the anthropogenic abiogenic sources. Industrial emissions may not have a significant seasonality, and combustion may be elevated in winters. Hence, higher estimates for these sources of N_2O leads to a decrease in the seasonal oscillations for the northern hemisphere.
- *Other sources.* No quantitative estimate of N_2O emissions from coastal marine and freshwater systems, aquifers and soils with inputs from N deposition could be given by Bouwman et al. (in press). Since these sources may be related to human, industrial and agricultural activity, most associated emissions may come from the northern hemisphere (Bouwman et al., in press). Other N_2O source candidates, such as industrial and chemical processes, medical and industrial use of N_2O, production and use of explosives and corona power losses from electric transmission lines are located in the 30-90°N zone. Assuming that these sources are significant and do not show seasonal patterns, this may further reduce the simulated seasonal amplitude of the N_2O concentration in the northern hemisphere.

(ii) *Biases in the transport model.* No atmospheric transport model can give a fully realistic description of atmospheric mixing and transport. However, in previous studies the atmospheric model correctly reproduced the latitudinal gradient and seasonality of CFC-11 and other tracers (Taylor, 1991). We therefore assume that the model disagreement with atmospheric observations is not caused by biases in the model and adequately reflects the seasonality in atmospheric concentrations induced by a seasonally varying source.

(iii) *Reliability of the atmospheric N_2O measurements.* Both the CMDL and ALE/GAGE data have a precision of close to 0.15% or 0.5 ppb in the N_2O concentration measurements, but the standard deviation in the daily and monthly averages in much higher (Montzka et al., 1992; Prinn et al., 1994). The seasonal variations predicted by the model in most northern hemisphere locations is in the order of 1 ppb (corrected for the 0.8 ppb trend), while in locations in the southern hemisphere the simulated seasonality is even less. Hence, it is difficult to validate our model results with the available observational data. The oscillations observed by the CMDL flask sampling program are in general larger than those of ALE/GAGE, while the ALE/GAGE and CMDL data show completely different patterns for the same monitoring station even when data for the same year are compared (Samoa, Figure 5h). This may be caused by differences in the sampling frequency between CMDL and ALE/GAGE measurements. Prinn et al. (1990) and Elkins (personal communication) concluded that CO_2 does not interfere with N_2O measurements. However, inconsistencies could be the result of other interferences in the N_2O concentration measurements.

The modelled results are in general agreement with simulations performed by Taylor (1992). However, the latitudinal gradients and the seasonal oscillations simulated in this analysis are smaller than those of Taylor (1992). This is caused by the weak oceanic source (2 Tg yr^{-1}) assumed by Taylor, and the dependence of soil and oceanic fluxes upon net primary production causing strong differences between winter and summer fluxes. Results also agree with simulations of Nevison (1994), although in that study much lower continental maxima were produced. The latitudinal and seasonal source estimates in this study are similar to those of Nevison (1994). A reason for the higher maxima in this study may be the grid size, which is ~6 times smaller than Nevison's grids. The higher resolution model we used better resolves the variability of the 1° x 1° resolution N_2O flux fields because it includes less numerical diffusion and is able to resolve concentration gradients more precisely. This results in a more detailed description of advection and mixing.

6. CONCLUSIONS

The simulated N_2O concentrations are in general agreement with observations. The calculated semi-hemispheric difference is consistent with the observations, indicating that the overall distribution of the a priori emission inventories is correct. Modelled results suggest N_2O concentrations of up to 5 ppb higher than those predicted over the oceans over tropical humid forest regions throughout the year, and over temperate continental interiors in summer. At present there are no atmospheric observations available to verify this result because most monitoring stations are located in coastal areas, on remote islands and at the South Pole.

Simulated seasonal distributions are not consistent with observations at a number of monitoring sites. In particular, at northern mid-latitudes the simulations resulted in seasonal trends, while the observations show no seasonality at all. For the southern hemisphere the simulated seasonality is much smaller than in the northern hemisphere and more consistent with observations.

There is still a great uncertainty in the various individual source estimates that could not be resolved in this analysis and in a previous comparison with source estimates from inverse modeling. We have discussed possible errors and omissions in our source estimates. Emissions from soils, the major global source, are highly uncertain. The emission patterns in the inventories of soil emissions are consistent with observations. However, soil consumption of N_2O and the process of N_2O emission from soils at low temperatures was neglected in the a priori estimates. Including the emissions in winter, early spring and autumn, and soil N_2O consumption, would lead to a decrease in seasonality in temperate zones. Consumption of N_2O in aquatic systems is represented in the a priori estimate for oceans, but the magnitude, seasonality and spatial patterns are extremely uncertain. If N_2O consumption in soils and aquatic systems is significant, the current estimates of the atmospheric lifetime and source estimates of N_2O will have to be revised.

Combustion-related N_2O emissions may be underestimated. If these emissions prevail in temperate winters, this may also lead to lower amplitudes. Finally, the a priori estimates do not include estimates for coastal waters and fresh water systems. Neither do they include estimates of N_2O from aquifers, effects of N deposition on soil N_2O fluxes, medical and industrial use of N_2O, production and use of explosives, corona power loss from electric transmission systems, and other potential industrial and chemical processes. Most of these sources may not have a seasonal pattern, and it is not clear how they would influence the simulated concentrations.

Modeling of atmospheric N_2O is hampered by the small number of long-term observation sites.

Most existing stations are located in remote places where signals from continental sources are not observed. In some cases the observations from different reports are contradictory, possibly caused by differences in sampling frequency. There may also be unknown interferences in the N_2O concentration measurements. The comparison of data from different groups also indicated that the precision of the available atmospheric N_2O measurements may not be sufficient to resolve relatively small seasonal variations. Resolving an annual averaged latitudinal gradient may be testing the limits of current measurement technology. Future forward and inverse modeling approaches will be more useful for N_2O when high-precision atmospheric data are available, including measurements in continental interiors.

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Chapter 7

CONCLUSIONS

The major global sources of nitrous oxide (N_2O) are soils under natural vegetation, followed by oceans. Next come agricultural soils and grasslands including the effects of synthetic N fertilizer use and animal excreta. Most other sources, including stationary and mobile fossil fuel combustion, industrial processes, biomass burning, atmospheric N_2O formation and coastal and fresh water systems are of minor importance at the global scale, but their local contribution to emissions may be high.

7.1. EMISSION INVENTORIES

- *Soils under natural vegetation.* The spatial and seasonal distributions of N_2O production in soils under natural vegetation are very similar to climate patterns. Comparison with measurement data showed that levels of N_2O production predicted by the model discussed in chapter 2 are in general agreement with measured fluxes. The model confirms conclusions from earlier studies that the major source regions of N_2O are in the tropics. Because of the simplicity of the model and the sparsity of data, the estimate of global N_2O emission from natural ecosystems based on the regression analysis is extremely uncertain.
- *Agriculture.* Literature data on N_2O fluxes from fertilized agricultural lands are not suitable to estimate the local N_2O losses as a function of fertilizer type and crop, but they may be used to estimate the order of magnitude of the global emission. Analysis of the literature data showed that the fertilizer-induced N_2O emission resulting from measurements covering longer periods is higher than for measurements representing short periods. A simple method was developed to estimate the total annual direct N_2O emission from fertilized fields, based on measurements covering periods of one year: N_2O-N emission = 1 kg N $ha^{-1}yr^{-1}$ plus 1.25 ± 1% of the fertilizer N applied (kg N $ha^{-1}yr^{-1}$). This method applies to all fertilizer types.

The inventory based on estimates of N excretion by various categories of domestic animals show that the global flux of N in animal excreta (~ 100 x 10^{12} g N yr^{-1}) about equals that applied as synthetic fertilizers (80 x 10^{12} g N yr^{-1}) and that their associated N_2O emissions may be of the same order of magnitude.

The global N_2O emission from synthetic N-fertilizers and animal excreta constitutes about 15% of the total global N_2O source, and about 2/3 of the atmospheric N_2O increase.

- *Forest clearing.* Soil organic matter losses are generally high in the first few years after forest clearing. The N mineralized from the decomposed soil organic matter causes elevated rates of nitrification, denitrification and associated N_2O emissions in the initial period following the clearing; in old clearings (>15 yr) easily decomposable soil organic matter is depleted, causing lower rates than in the original forest. Soil N budgets for a deforestation sequence in the humid tropics of Costa Rica indicate that part of the mineralized N is lost via leaching, while most of the N loss occurs through denitrification. The N losses from cleared forest soils may be an important source of pollution of the atmosphere and aquatic systems. The estimate of soil N_2O emission from denitrification following deforestation, accounting for declining N_2O fluxes with time, indicates that this process contributes perhaps 10% to the atmospheric increase. This is lower than previous estimates, which disregarded the effect of ageing of the clearings.
- *Energy-related and industrial sources.* The global N_2O emission from fossil fuel combustion, traffic and industrial sources contributes about 5% of the total N_2O source, and 15% of the atmospheric N_2O increase. In addition, deposition of NO_x emitted during combustion may cause N_2O formation in soils. Traffic is probably a global source of N_2O that is growing at a fast rate, caused by the fast penetration of automobiles equipped with 3-way catalysts. The N_2O emission increases with the age of the catalysts, but the emission factors are yet uncertain. The inventories

of N_2O emissions from nitric acid and adipic acid production are uncertain because of scarcity of data on the volumes of production and emission rates. Although our knowledge of the global anthropogenic N_2O sources has improved in recent years, other as yet unidentified industrial processes may also generate N_2O.

7.2. LATITUDINAL DISTRIBUTIONS

The emission inventories presented in this study, complemented with inventories of N_2O emissions from oceans and biomass burning from the literature, are in general agreement with source estimates inferred from inverse atmospheric modeling. Major uncertainties are in the tropics. All sources include uncertainties. The general agreement between the emission inventory and source estimates from inverse modelling may be fortuitous if misrepresentations of the largest global sources (soils and oceans) happen to balance out. The uncertainty in the global estimates of N_2O emission from soils and oceans is caused primarily by scarcity of measurement data in large parts of the world. The global oceanic N_2O emission may be higher than assumed by Watson et al. (1992), and upwelling regions in the 0°-30°N and 30°S-90°S latitudinal zones may be major source regions with N_2O fluxes which are much higher than the world average oceanic flux.

The major part of the N_2O emission from N fertilizers, animal excreta, combustion, and industrial processes stem from the northern hemisphere. Atmospheric N_2O formation is prevalent in the tropics. No quantitative estimate of N_2O emissions from cultivation of leguminous crops, coastal marine and fresh water systems, aquifers, and soils with inputs from N deposition could be given. However, since these sources may be closely related to human population, industrial and agricultural activity, most associated emissions may come from the northern hemisphere. Other N_2O source candidates, such as industrial and chemical processes, medical and industrial use of N_2O and production and use of explosives, are located primarily in the 30°N-90°N zone.

Predicted N_2O concentrations, based on monthly estimates of N_2O emission used to prescribe a three-dimensional atmospheric transport model, are ~1 ppb higher in the northern than in the southern hemisphere. This is consistent with the atmospheric observations, indicating that the overall distribution of the a priori emission inventories is correct. Model results suggest high N_2O concentrations over tropical humid forest regions throughout the year, and over temperate continental interiors in summer, of up to 5 ppb higher than concentrations predicted over the oceans. At present there are no atmospheric observations available to verify this result, because most monitoring stations are located in coastal areas, on remote islands and at the South Pole.

7.3. SEASONAL DISTRIBUTIONS

Simulated seasonal distributions based on monthly estimates of N_2O emission prescribing a three-dimensional atmospheric transport model, are not consistent with observations at a number of monitoring sites. The simulations resulted in seasonal trends at northern mid-latitudes, where the observations show no seasonality. For the southern hemisphere the simulated seasonality is much smaller than in the northern hemisphere and more consistent with concentration measurements.

For soils and oceans, the major global sources of N_2O, both the global emissions and their seasonal distribution are highly uncertain. Soil consumption of N_2O and the process of N_2O emission from soils at low temperatures were omitted in the a priori estimates. These potential errors may exaggerate the seasonality in temperate zones. Consumption of N_2O by aquatic systems is represented in the a priori estimate for oceans, but the magnitude, seasonality and spatial patterns are extremely uncertain. If N_2O consumption in soils and aquatic systems is significant, the current estimates of the atmospheric lifetime and source estimates of N_2O need to be revised.

Industrial N_2O sources may not show a seasonal pattern, but combustion-related N_2O emissions may prevail in temperate winters. Underestimation of the N_2O emission from combustion during the winter period may, therefore, lead to exaggerated amplitudes of the N_2O concentration in the northern hemisphere.

The N_2O emission from coastal waters and fresh water systems, aquifers, elevated N_2O emission from soils and aquatic systems caused by atmospheric N deposition, medical and industrial use of N_2O, production and use of explosives, formation of N_2O during corona power loss from electric transmission systems, and other potential industrial and chemical processes generating N_2O, were not included in the a priori estimate. The magnitude and seasonal pattern of N_2O emissions from these sources is highly uncertain.

Modeling of atmospheric N_2O is hampered by the small number of long term observation sites. Most existing stations are located in remote places where signals from continental sources are not observed. The precision of the available atmospheric N_2O measurements is inadequate to resolve relatively small seasonal variations. Resolving an annual averaged latitudinal gradient may be testing the limits of current measurement technology.

7.4. FUTURE RESEARCH

Improvement of models of N_2O production in soils of the type described in chapter 2 requires more long-term measurements in those regions with high expected N_2O fluxes, and in ecosystems which may be important contributors to the global production of N_2O due to their large area. There are no flux measurement data available for Asia, Africa or Australia to validate models. The effects of N deposition on N_2O emissions from temperate forest soils, and the role of biological N fixation in the N_2O production from soils under natural vegetation, are uncertain. Measurements are also required in ecosystems prone to N_2O uptake, e.g. N-limited temperate forests. Description of the processes leading to N_2O pulses resulting from rainfall events, and episodic emissions at low temperatures in autumn, during thaw periods in early spring, and from snow covered soils in winter, would constitute a major improvement in models such as the one described in chapter 2.

Further improvement of global-scale models describing trace gas fluxes requires development of soil data bases that include a number of soil characteristics vital to evaluating soil physical and chemical characteristics. More spatial and temporal detail in data bases of vegetation (NPP) and litter composition allow a better description of the temporal and geographic variability of controls on N_2O fluxes and, possibly, episodic pulses of N_2O.

Estimates of N excretion for the different animal categories discussed in chapter 4 can be improved by assessing the relation between quality and consumption of forage and feed, the fraction of energy required for maintenance, the efficiency of energy conversion to body weight or milk, and N excretion. Such studies would require information on the herd composition, the period of grazing and the storage conditions of manure in the stable, which determine N_2O evolution. Measurements of N_2O and development of soil models describing the regulating factors of N_2O emission and their variability may improve the estimates for fields receiving inputs from fertilizers and animal excreta.

Global biological N fixation in agricultural systems may be an important global N_2O source. Statistics on harvested areas and production of leguminous crops are available, and the N fixation could be estimated from the N content of the various leguminous crops. Measurements in fields with leguminous crops are required to relate N_2O fluxes to N fixation. In addition, data on the occurrence of legumes in pastures and legumes grown as green manures, and their N fixation, are required.

Future research to improve estimates of N_2O emission from the oceans, coastal marine and fresh water systems include measurements coupled with modeling approaches, as discussed in detail by Nevison (1994). A number of future research needs regarding fossil fuel combustion and industrial sources as summarized by Kroeze (1994) include technical solutions, such as the development of a new catalyst for nitric acid production and automobiles.

Forward and inverse atmospheric modelling approaches will be more useful for N_2O studies when high precision atmospheric data are available, including measurements at stations in continental interiors.

7.5. POLICY IMPLICATIONS

The atmospheric N_2O concentration started to increase at an accelerated rate in the middle of this century. Most of this increase is related to increasing agricultural and industrial production and fossil fuel combustion, including technologies that reduce NO_x emissions. The major part of the N_2O increase stems from sources related to food production, including synthetic fertilizer use, animal excreta, biomass burning, biological N fixation, land clearing and biomass burning. There are technical options to reduce soil N_2O emission from activities related to food production. CAST (1992), Kroeze (1994) and Mosier (pers. comm.) suggested N management strategies to mitigate N_2O emission without decreasing production (Table 1). The proposed measures would probably reduce rather than increase costs. Emission reductions achieved by some of these management strategies may be considerable. For example, use of slow release fertilizers may reduce N_2O emission by up to 30-65% (Kroeze, 1994). Mosier (pers. comm.) estimated that a reduction of the global annual N_2O emission of about 0.2 x 10^{12} g N_2O-N can be achieved by using controlled release fertilizers and nitrification inhibitors. A reduction of 0.4 x 10^{12} g N_2O-N can be achieved by increased fertilizer efficiency and by closing the N flow cycles. The total 0.6 Tg N_2O-N reduction is about 30% of the total current annual N_2O release from synthetic fertilizers and animal excreta.

A growing world population will inevitably lead to increased demand for food and increased emission of N_2O. Synthetic fertilizer use prevails in the developed countries, although the use is growing at a fast rate in developing countries (FAO, 1991). In addition, an increasing portion of the use of synthetic fertilizers is driven by the demand for feed for animal production. At present close to 40% of the global cereal production and 25% of the production of root and tuber crops are fed to animals. For the developing countries 16% of cereal consumption and 20% of root and tuber crops are used as animal feed, while in the developed countries this is 62% and 33%, respectively (FAO, 1991). With the increasing input of N in the form of fertilizers, it will be difficult to mitigate emissions associated with food production. However, important reductions could be achieved by a shift-away from animal production, which reduces both synthetic fertilizer use and production of animal excreta.

Stationary combustion is a minor direct source at the global scale. Indirect effects of combustion caused by NO_x emission and atmospheric deposition of N leading to enhanced soil N_2O fluxes, may be more important. Energy saving will, therefore, lead to reduction of both direct and indirect N_2O emission. Options to avoid or reduce the direct N_2O emission from stationary fossil fuel combustion include technical solutions and switch to other fuel types. Mobile combustion is a growing source of N_2O. The N_2O emissions from vehicles can be reduced by developing new catalysts as well as by reduction of automobility, a switch to non-fossil sources of energy and increased energy efficiency. The N_2O emission can also be reduced by low NO_x engines and energy saving. However, if neither technological solutions or reduction in energy consumption can be achieved, policy makers will be faced with a choice between reduction of one pollutant (NO_x) at the expense of the other (N_2O). As NO_x contributes to urban smog and acidification, actions to reduce N_2O may not be given priority.

Industrial sources also contribute significantly to the atmospheric N_2O increase, and unidentified processes may exist that generate N_2O. The N_2O losses from nitric acid production can be reduced

Table 1. Strategies to reduce N_2O emissions associated with food production

1. Increase N use efficiency by matching N supply with crop demand
   - Determination of the fertilizer N requirement by soil testing adjusting the N rate to a reasonable yield goal for specific fields, accounting for N from soil mineralization, residues, manures, organic wastes and mineral N added by irrigation water or atmospheric deposition. This will also reduce leaching of nitrate from soils
   - Timing of N application to when it is needed by the crop
2. Close N flow cycles
   - Integration of animal and crop production systems in terms of manure reuse and plant production
   - Avoidance of N from plant residues from the production site
3. Advanced fertilization techniques
   - Slow release fertilizers.
   - Placement of synthetic fertilizers below the surface
   - Nitrification inhibitors to keep N in reduced form and avoid denitrification. However, some of these chemicals may have adverse environmental side-effects.
   - Foliar application of fertilizers
4. Additional strategies
   - Strategies to avoid N_2O losses from animal manure storage
   - Options to avoid NH_3 and NO_x emission to reduce N deposition
   - Shift away from animal production

by catalysts (Kroeze, 1994). Finally, there is one source that is currently decreasing. In 1991 the major nylon producers announced their intention to phase out processes leading to N_2O production within 5 years (Du Pont, 1991).

Global warming may cause increased fluxes of N_2O from soils and aquatic systems. Model results relating N_2O fluxes to temperatures indicated that the observed temperature decrease associated with the eruption of Mount Pinatubo may have caused a significant decrease in N_2O emission from soils, and the oceanic N_2O emission may be related to temperature in a similar way. This decrease in emissions may explain the observed significant decrease in the growth rate of N_2O in the atmosphere during the same period. If the decrease of the growth rate of the atmospheric N_2O concentration is in actual fact caused by the temperature feedback on natural emissions, future emissions may increase along with the expected global warming.

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In deze studie zijn de mondiale emissies van lachgas (distikstofoxide, N_2O) naar de atmosfeer in kaart gebracht. Hiertoe werden inventarisaties met een resolutie van 1° x 1° (lengtegraad x breedtegraad) gemaakt van de bekende bronnen van lachgas, waaronder emissies door bodems onder natuurlijke vegetatie, landbouwgronden en het effect van bemesting met stikstof (N), graslanden en dierlijke N-excretie, emissies tijdens het verbranden van biomassa, verhoogde emissies na ontbossing, mobiele en stationaire verbranding, en industriële bronnen.

De mondiale N_2O-emissie door bodems onder natuurlijke vegetatie is in kaart gebracht met behulp van een simpel model gebaseerd op de vijf belangrijkste factoren die een rol spelen bij de vorming van N_2O in bodems: (i) de input van organisch materiaal; (ii) bodemvruchtbaarheid; (iii) bodemvochttoestand; (iv) temperatuur; en (v) de beschikbaarheid van zuurstof in de bodem. Indices voor deze vijf factoren werden beschreven op basis van mondiale gegevensbestanden van bodemtype en textuur van de bovengrond, de "normalized difference vegetation index" (NDVI) en klimaat, met een geografische resolutie van 1° x 1°. Het ontwikkelde model verklaart ongeveer 60% van de variabiliteit in de waargenomen emissies op ongeveer 30 verschillende lokaties in de wereld waar in natuurlijke ecosystemen N_2O-emissies zijn gemeten. De resultaten bevestigen conclusies van eerdere studies dat de belangrijkste brongebieden in de tropen liggen.

Uit analyse van literatuurgegevens over de uitstoot van N_2O uit bemeste landbouwgronden bleek dat naarmate de emissie gedurende langere perioden wordt gemeten, de gemeten fractie van de toegediende N-meststof die als N_2O ontwijkt, groter is. Op grond van deze conclusie werden alleen metingen, die een periode van 1 jaar besloegen, gebruikt om de jaarlijkse N_2O-emissie als volgt te schatten: achtergrondemissie van 1 kg N_2O-N plus 1.25 ± 1% van de toegediende N-meststof.

Voor het in kaart brengen van de N_2O-emissies uit dierlijke mest werden de N-excreties door verschillende landbouwhuisdieren geschat. De berekende mondiale dierlijke N-excretie is van dezelfde orde van grootte als de mondiale hoeveelheid N-kunstmestsof gebruikt in de landbouw. De N_2O-emissie uit dierlijke mest is daarom mogelijk ook van de zelfde orde van grootte als die veroorzaakt door het gebruik van kunstmest.

Voor een aantal lokaties in de Atlantische Zone in Costa Rica waar na ontbossing zeer hoge N_2O-emissies zijn gemeten, is een stikstofbalans opgesteld voor de situatie voor en na ontbossing. Een belangrijke hoeveelheid bodem-N wordt gemineraliseerd na het kappen van het bos. Een groot deel van de gemineraliseerde N gaat verloren na nitrificatie tot nitraat. Een gedeelte spoelt uit, en het grootste deel van de nitraat gaat verloren door denitrificatie. Na 3 tot 5 jaren na de ontbossing is het merendeel van de gemakkelijk afbreekbare bodem organische stof afgebroken en nemen denitrificatie en N_2O-emissie sterk af. Uit de mondiale schatting van de toename van de N_2O-emissie door denitrificatie die gedurende enige jaren optreedt na ontbossing blijkt dat deze bron aanzienlijk bijdraagt aan de toename van de N_2O-concentratie in de atmosfeer.

Mondiale inventarisaties met een resolutie van 1° werden ook gemaakt voor de N_2O-emissie veroorzaakt door het gebruik van fossiele brandstoffen, het verbranden van biomassa en industriële bronnen. Inventarisaties van N_2O-emissies door oceanen en het verbranden van biomassa werden verkregen uit de literatuur. De complete mondiale inventarisatie van alle bronnen van N_2O werd op jaarbasis vergeleken met resultaten van een "inverse" modelstudie, d.w.z. het schatten van emissies met behulp van atmosferische modellen door uit te gaan van gemeten atmosferische concentraties en het terugrekenen naar emissies. De emissie inventarisatie stemt overeen met de schattingen gemaakt met inverse modellering, en de belangrijkste conclusies van de vergelijking zijn:

- De uitstoot van N_2O door oceanen is waarschijnlijk hoger dan eerdere schattingen uit de literatuur. In de oceanen zijn de belangrijkste brongebieden waarschijnlijk in de tropen op het Noordelijk halfrond (0°-30°NB) en nabij Antarctica (30°ZB-90°ZB).
- Het grootste deel van de N_2O-emissie door akkerlanden en beweide graslanden vindt plaats op het Noordelijk halfrond. Andere belangrijke bronnen zijn N_2O-emissies veroorzaakt door N-

depositie en door het verbouwen van N-bindende gewassen (vlinderbloemigen, zoals erwten, bonen en soya).

- De N_2O-emissies door het gebruik van fossiele brandstoffen en industriële bronnen komen hoofdzakelijk uit de 30°NB-90°NB zone, terwijl emissies gerelateerd aan het gebruik van brandhout het belangrijkst zijn in de zone tussen 0° en 30°NB.
- Het grootste deel van de N_2O-emissie uit kustwateren en zoetwatersystemen komt van het Noordelijk halfrond.

Maandelijkse schattingen van mondiale N_2O-emissies met 1°x1° resolutie werden gebruikt als invoer voor een drie-dimensionaal atmosferisch transportmodel. De gesimuleerde concentratie voor het Noordelijk halfrond was ongeveer 1 ppb hoger dan op het Zuidelijk halfrond, in overeenstemming met de gemeten waarden. De gesimuleerde concentraties boven belangrijke brongebieden, zoals tropische regenwouden, waren tot 5 ppb hoger dan boven de oceanen. De voorspelde concentraties boven de continenten op het Noordelijk halfrond waren hoger in de zomer dan in de winter als gevolg van de seizoensverschillen in de emissies door bodems. In de atmosferische metingen is echter geen seizoensvariatie zichtbaar. Op het Zuidelijk halfrond vertoonden de voorspelde concentraties vrijwel geen seizoensverschillen, hetgeen overeenstemt met de atmosferische metingen. De inconsequentie tussen voorspelde en gemeten concentraties op het Noordelijke halfrond is mogelijk veroorzaakt door overschatting van de seizoensvariatie in de emissies. De mondiale inventarisatie houdt geen rekening met mogelijke consumptie van N_2O in oecosystemen die N-gelimiteerd zijn. Bovendien is in de inventarisatie geen rekening gehouden met emissies gedurende de winter, lente en herfst in gematigde streken. Deze omissies en mogelijke onderschatting van de N_2O-emissie door verbranding van fossiele brandstoffen in de winter op het Noordelijk halfrond zijn wellicht de oorzaak van overschatting van de seizoensverschillen in emissies.

De meeste meetstations waar N_2O-concentraties worden gemeten liggen in kuststreken en andere plaatsen ver verwijderd van de belangrijkste continentale bronnen. Dit is mogelijk een oorzaak van het gebrek aan seizoensvariatie in de metingen. Het is ook mogelijk dat kleine verschillen niet kunnen worden waargenomen met de bestaande meettechnieken voor N_2O-concentraties.

Ongeveer tweederde van de toename van N_2O in de atmosfeer wordt veroorzaakt door emissies die gerelateerd zijn aan de produktie van voedsel. De toename van de wereldbevolking zal de vraag naar voedsel en de uitstoot gerelateerd aan voedselproduktie verder doen stijgen, terwijl het aandeel van de kunstmeststoffen die gebruikt worden ten behoeve van de produktie van veevoer toeneemt. Ongeveer 40% van de huidige mondiale graanproduktie en 25% van de wortel- en knolgewassen wordt aan dieren gevoerd. Een aanzienlijke vermindering van N_2O-emissies kan worden bereikt door het beperken van de vleesconsumptie. Reducties kunnen ook worden gerealiseerd door het verhogen van de efficiëntie van het gebruik van N in de landbouw en door het aanpassen van bemestingstechnieken.

Bij mobiele- en stationaire verbranding kunnen technische oplossingen (bijvoorbeeld het ontwikkelen van verbeterde katalysatoren) en energiebesparing leiden tot reductie van de N_2O-emissie. Katalysatoren kunnen ook gebruikt worden om de N_2O-emissie te beperken bij industriële processen, zoals produktie van salpeterzuur. De belangrijkste producenten van adipinezuur (gebruikt voor de produktie van nylon) hebben afgesproken om de N_2O-emissie terug te brengen.

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CURRICULUM VITAE

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