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Closing the global N_2O budget: A retrospective analysis 1500 - 1994

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Abstract. We present new estimates of global nitrous oxide (N_2O) emissions for the period 1500 - 1994 based on revised Intergovernmental Panel on Climate Change guidelines [*Intergovernmental Panel on Climate Change (IPCC)*, 1997; *Mosier et al.*, 1998]. Use of these estimates as input to a simple atmospheric box model resulted in a closed N_2O budget over time, showing that increases in atmospheric N_2O can be primarily attributed to changes in food production systems. We hypothesize that before the ninetheenth century conversion of natural land to agriculture had no net effect on N_2O. During the twentieth century a fast expansion of agricultural land coupled with intensification of land use may have caused a net increase in N_2O. In our base scenario the total N_2O emissions increased from 11 Tg N yr^{-1} in 1850 to 15 Tg N yr^{-1} in 1970 and to 18 Tg N yr^{-1} in 1994.

1. Introduction

Global budget studies show that it is difficult to explain the observed increase in atmospheric nitrous oxide (N_2O). The major problem is to quantify natural and anthropogenic sources of N_2O and their changes over time. N_2O is one of the natural components of Earth's atmosphere and contributes to the natural greenhouse effect. Its atmospheric concentration has been increasing at an accelerated rate for several decades [*Prather et al.*, 1995]. The N_2O budget has been the least well constrained of the global trace gas budgets.

Global inventories of anthropogenic N_2O emissions have changed considerably during the past 15 years. During the 1980s, coal combustion was considered to be the most important anthropogenic source of N_2O, but the measurement data were shown to be in error [*Muzio and Kramlich*, 1988]. At present it is widely accepted that both natural and anthropogenic emissions stem to a large extent from biogenic sources [e.g., *Bouwman et al.*, 1995]. Nevertheless, in both the 1990 and 1992 Intergovernmental Panel on Climate Change (IPCC) Scientific Assessments it was concluded that the estimated ranges for known anthropogenic sources of N_2O could not explain the atmospheric increase [*Watson et al.*, 1990, 1992].

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More recently, the 1994 IPCC Scientific Assessment presented a closed global N_2O budget, based on a preliminary revision of the IPCC Guidelines for National Greenhouse Gas Inventories [*Prather et al.*, 1995; *Cole et al.*, 1996]. The IPCC Guidelines have been developed for the purpose of assisting countries in estimating their national emissions of greenhouse gases. The revised method for estimating N_2O emissions from agriculture includes the full impact of agriculture on the global nitrogen cycle and provides default factors for N_2O emissions from agricultural soils, animal waste management systems, and indirect emissions caused by nitrogen losses from agriculture through leaching and volatilization [*Intergovernmental Panel on Climate Change (IPCC)*, 1997; *Mosier et al.*, 1998]. In this paper we present estimates of global nitrous oxide for the period 1500 - 1994 based on the revised IPCC Guidelines [*Intergovernmental Panel on Climate Change (IPCC)*, 1997; *Mosier et al.*, 1998]. We calculate trends in atmospheric N_2O from these emission estimates using a simple atmospheric model and compare the results to observed trends in atmospheric N_2O.

2. Estimating Global Emissions of N_2O

We estimated global N_2O emissions from natural and anthropogenic sources for the years 1500, 1600, 1700, 1800, 1900, 1930, 1950, 1960, 1970, 1980, 1990, and 1994. Our estimates for natural emissions are presented below. Anthropogenic emissions are from agriculture, energy, industry, and biomass burning. Of these, agriculture is

generally considered the most important on a global scale [*Bouwman et al.*, 1995; *Prather et al.*, 1995].

Emissions from energy, industry, and biomass burning were mainly taken from *Prather et al.* [1995] and *Kroeze* [1994]. Emissions from energy include both stationary sources (power plants) and mobile combustion (traffic). Industrial emissions include N_2O formation during production of adipic acid and nitric acid. We ignored the use of anesthesia, the contribution of which to global N_2O is likely to be small [*Bouwman et al.*, 1995]. We assumed that before 1900 emissions from energy and industry did not occur and that from 1990 trends continued. Emissions during biomass burning include N_2O formation during the burning and subsequent enhanced soil emissions. We tentatively set emissions from biomass burning before 1900 at 0.1 Tg N yr^{-1}.

We applied the revised IPCC Guidelines [*Intergovernmental Panel on Climate Change (IPCC)*, 1997; *Mosier et al.*, 1998] to estimate agricultural emissions. This methodology aims at assessing the full nitrogen cycle, taking into account all direct and indirect effects of human activities on N_2O emissions. The IPCC Guidelines distinguish between (1) direct soil emissions of N_2O as induced by N inputs (fertilizers, animal waste, crop residues, and biological N_2 fixation), or organic soil cultivation, (2) N_2O production in animal waste management systems, and (3) indirect N_2O emissions caused by nitrogen leaching to ground or surface waters or volatilization and consecutive deposition of nitrogen oxides and ammonia. The IPCC Guidelines are by definition applicable to any country. The economic input data needed to estimate emissions can be obtained from readily available databases (e.g., Food and Agricultural Organization (FAO) databases) of fertilizer use, livestock numbers, crop production, cultivation of organic soils, and human population.

We estimated agricultural emissions on the basis of input data from FAO databases [*Food and Agricultural Organization (FAO)*, 1995] or estimated as summarized in Table 1. Crop production, animal waste, and fertilizer use were extrapolated back in time based on human population estimates. These estimates were then used to calculate N input to agricultural systems using the *IPCC* [1997] methodology for each time period. The year 1970 was used as a base time because FAO data were available to estimate N input from each source. After 1970 the FAO data were used directly to make N input estimates.

3. Atmospheric Box Model

The emission estimates were used to simulate atmospheric concentrations using a one-box atmospheric model [*Khalil and Rasmussen*, 1988, 1992; *Kroeze*, 1994]:

$$dC/dt = S/F - C/T$$

where

C concentration (ppbv);
S emissions (Tg N);
T atmospheric lifetime (years);
t time (years);
F conversion factor (Tg N $ppbv^{-1}$).

Table 1. Development for the Period 1500 - 1970 of the Volumes of Crop Production, Animal Waste Production, and Growth of Human Population and Fertilizer Use Used for the Calculation of Global N_2O Emissions From Agriculture

Year	Crop Production (Fraction of 1970)	Animal Waste (Fraction of 1970)	Human Population, billion	Fertilizer Use, Tg N yr^{-1}
1500	0.13	0.13	0.5	0
1600	0.15	0.15	0.6	0
1700	0.17	0.17	0.6	0
1800	0.27	0.27	1	0
1900	0.43	0.43	1.6	0
1930	0.57	0.57	2.1	1.7
1950	0.67	0.67	2.5	3.4
1960	0.82	0.82	3.0	9.2
1970	1	1	3.7	28.7

Data for the years from 1970 were taken from FAO databases [*FAO*, 1995]. Crop production and animal waste for 1500 - 1970 were estimated on the basis of a linear relationship between population and production. The source for animal wast production data is *Kroeze* [1994]; the sources for the growth of human population data are *Grigg* [1974, 1987], *Kroeze* [1994], *FAO* [1995], and the United Nations (World population from year 0 to stabilization accessed at gopher://gopher.undp.org: 70/00/ungophers/popin/wdtrends/histor, 1996). The sources for fertilizer use data are *Kroeze* [1994] and *FAO* [1995]. Data from *Mitchell* [1992, 1993, 1995], *Klein Goldewijk and Battjes* [1997], and United Nations [1996] were used to analyze the relationship between global population and crop and livestock production for the period 1890 - 1967. Crop production was found to be strongly correlated to population with R^2 of 0.96 for a linear function according to production = -0.61+0.67 x population (in millions). The data show irregularities in times of war (1914 - 1918 and 1940 - 1945), when crop production grew slower than population, and in the last two decades of the period, when crop production grew slightly faster than population. We assumed that animal waste production is proportional to livestock production. For livestock production the historical data from *Mitchell* [1992, 1993, 1995] are not yet available in digital form. Only animal population data are available, which show a similar relationship to population as crop production. Therefore we used the same linear relationshop for livestock and animal waste production as for crop production. We also analyzed data for Canada, United States of America, Brazil, and India for 1890 - 1967, all showing relationships between population and crop and livestock production similar to the global one.

Atmospheric concentrations were calculated and compared to observed values [*Montzka et al.*, 1992; *Boden et al.*, 1993] and values deduced from air in the snow pack (firn) or ice cores on the South Pole [*Machida et al.*, 1995; *Battle et al.*, 1996]. Observed concentrations are from the Atmospheric Lifetime Experiment/Global Atmospheric Gases Experiment (ALE/GAGE) and the Climate Monitoring and Diagnostic Laboratory (CMDL). Annual available data for different ALE [*Boden et al.*, 1993], GAGE [*Boden et al.*, 1993], and CMDL [*Montzka et al.*, 1992] monitoring stations were averaged; following this procedure, we ignored the interhemispheric gradient which may have slightly affected the global averaged values.

For the base scenario we used the values published by IPCC for the atmospheric lifetime (120 years), the conversion factor (4.8 Tg N = 1 ppbv), and the preindustrial (i.e., pre-1800) atmospheric concentration (275 ppbv) [*Prather et al.*, 1995]. To test the sensitivity of the model for these assumptions, we varied the atmospheric lifetime (100 or 170 years) based on studies by *Prinn et al.* [1990], *Mischwaner et al.* [1993], and *Kroeze* [1994] and preindustrial concentration (270 or 280 ppbv) based on *Machida et al.* [1995] and *Battle et al.* [1996] in four scenarios (low 1 and 2 and high 1 and 2) (see Table 2). In addition we tested the sensitivity of the calculated concentrations to variations in the emission estimates (see Table 2).

4. Trends in N_2O Before the Nineteenth Century

The increase in atmospheric N_2O may not have started before the middle of the nineteenth century [*Khalil and Rasmussen*, 1988; *Machida et al.*, 1995; *Battle et al.*, 1996], indicating that the human impact on N_2O emissions was relatively small before that time. We therefore assumed that atmospheric N_2O concentrations were constant or changing at a rate that is undetectable from ice core analyses (dC/dt = 0) prior to 1850 (base scenario), 1800 (scenario high 1 and 2), or 1900 (scenario low 1 and 2). Our estimate of pre-1800 emissions is 11 (8 - 13) Tg N yr^{-1} (Table 3). This is consistent with estimated emissions from global preagricultural N_2O emissions from soils (6 - 7 Tg N yr^{-1}) [*Bouwman et al.*, 1993]; the present deep oceans (3 - 4 Tg N yr^{-1}) [*Nevison et al.*, 1995; *Prather et al.*, 1995], and the natural part of atmospheric N_2O formation and other aquatic sources (< 1 Tg N yr^{-1}, inferred from *Dentener and Crutzen* [1994] and *Seitzinger and Kroeze* [1998].

The N_2O emissions from denitrification of animal waste, biological N_2 fixation, crop residues and from biomass burning increased from 0.6 Tg N yr^{-1} in 1500 to 1.4 Tg N yr^{-1} in 1850 (Figure 1 and Table 3). Since the measurement data suggest that prior to the nineteenth century the atmospheric N_2O concentrations were constant, the base scenario emissions from undisturbed soils decrease by 0.8 Tg N yr^{-1} between 1500 and the middle of the nineteenth century (Figure 2 and Table 3), reflecting the decreasing emissions caused by the conversion of forests to agricultural land and reduced biogenic emissions caused by lower ambient temperatures during the little ice age [*Khalil and Rasmussen*, 1989].

In our analysis we thus assume that before the nineteenth century the decrease in N_2O emissions caused by conversion of forests to agricultural land was about balanced by increasing intensities in agricultural production. Prior to the nineteenth century, yield increases in agriculture were slow. Between 1300 and 1800 the yields of wheat and rice possibly doubled in England and China and increased much slower elsewhere [*Grigg*, 1987]. Until about 1600 there was probably very little difference in agricultural productivity between Europe, India, and China [*Grigg*, 1982]. Although eliminated in much of the Low Countries (at present the Netherlands and Belgium) and parts of England by the seventeenth century, fallow persisted in much of Europe until well into the nineteenth century. In the mid-nineteenth century it was possible to find farmers whose methods and implements had hardly changed since medieval times. Gradually, in the nineteenth century, legumes, potatoes, and sugar beet replaced the fallow. The root crops allowed for the removal of weeds during growth and provided fodder for livestock, which, in turn, provided farmyard manure for

Table 2. Overview of Scenario Assumptions and Impact on Variation in Anthropogenic Emissions Estimates on Calculated Concentration of N_2O in the Atmosphere

Scenario	Anthropogenic Emissions	Preindustrial Emissions	Preindustrial Concentration, ppbv	Atmospheric Lifetime, years	Year Increased Emissions Began	Calculated Atmospheric 1995 Concentration, ppbv
Base case	see Table 3	11.0	275	120	1850	316
Low 1	as base case	13.0	270	100	1900	306
Low 2	as base case	as base case	as base case	as base case	1900	312
High 1	as base case	7.9	280	170	1800	330
High 2	as base case	as base case	as base case	as base case	1800	322
em x 0.75	base case x 0.75	as base case	as base case	as base case	as base case	298
em x 1.25	base case x 1.25	as base case	as base case	as base case	as base case	334

Estimates for preindustrial emissions change with assumed atmospheric lifetime and preindustrial concentration. Abbreviation em indicates total anthropogenic emission in base scenario.

Table 3. Global N_2O emissions From Nonagricultural Soils and Oceans (Natural), Fossil Fuel Combustion (Energy), Adipic Acid and Nitric Acid Production (Industry), Biomass Burning, and Agriculture for 1500 - 1994 as Used in the Base Scenario and the Net Annual Addition of N_2O to the Atmosphere

Year	Natural	Energy	Industry	Biomass Burning	Agriculture	Total Anthropogenic	Total Global	Net additions to Atmosphere
1500	10.4	0.0	0.0	0.1	0.5	0.6	11.0	0.0
1600	10.4	0.0	0.0	0.1	0.6	0.7	11.0	0.0
1700	10.3	0.0	0.0	0.1	0.6	0.7	11.0	0.0
1800	9.9	0.0	0.0	0.1	1.0	1.1	11.0	0.0
1850	9.6	0.0	0.0	0.1	1.3	1.4	11.0	0.0
1900	9.6	0.0	0.0	0.1	1.6	1.7	11.3	0.3
1930	9.6	0.0	0.0	0.2	2.2	2.4	12.0	1.0
1950	9.6	0.1	0.0	0.2	2.9	3.1	12.8	1.8
1960	9.6	0.2	0.1	0.2	3.4	3.9	13.5	2.5
1970	9.6	0.3	0.3	0.3	4.2	5.1	14.7	3.7
1975	9.6	0.4	0.3	0.3	4.8	5.8	15.4	4.4
1980	9.6	0.5	0.4	0.3	5.3	6.5	16.1	5.1
1985	9.6	0.6	0.4	0.4	5.8	7.2	16.9	5.9
1990	9.6	0.7	0.5	0.5	6.2	8.0	17.6	6.6
1994	9.6	0.9	0.3	0.6	6.2	8.0	17.7	6.7

Emissions are in Tg ($=10^{12}$ g) N_2O-N yr^{-1}. Natural emissions include N_2O from soils under natural vegetation, oceans, aquatic systems and formation in the atmosphere (see text). Emissions from energy, industry, and biomass burning for the period 1900 - 1990 are based on the Intergovernmental Panel on Climate Change [*Prather et al.*, 1995] and *Kroeze* [1994], and assumed to be 0 before 1900 except 0.1 Tg N yr^{-1} from biomass burning. Emissions from agriculture are calculated using the revised IPCC Guidelines [*IPCC*, 1997; *Mosier et al.*, 1998] using FAO input data or assumptions summarized in Table 1 (see Figure 1). Net additions to atmosphere are total emissions minus 11 Tg N (1500 - 1800 emissions).

maintaining crop yields, while clover, beans, and peas all added nitrogen to the soil [*Chorley*, 1981; *Slicher van Bath*, 1968].

5. Trends in N_2O During the Ninteenth and Twentieth Centuries

5.1. Trends in Agriculture

Apart from overall fast population growth (Table 1) it was industrialization in the later 19th century that caused the major changes in agriculture. Several mutually related developments occurred simultaneously, including urbanization, economic growth inducing increasing demand for food and a shift toward more meat and milk, and mechanization and intensification in agriculture [*Grigg*, 1982].

Industrialization led to fast urbanization in Europe and northern America. Factory employment increased so rapidly that by the 1880s migration to towns was causing a decline in the agricultural population, compelling farmers to adopt labor-saving technologies. Examples include the introduction of labor-saving machinery, the use of steam and later electricity on the farms and the petrol-driven tractor, and the

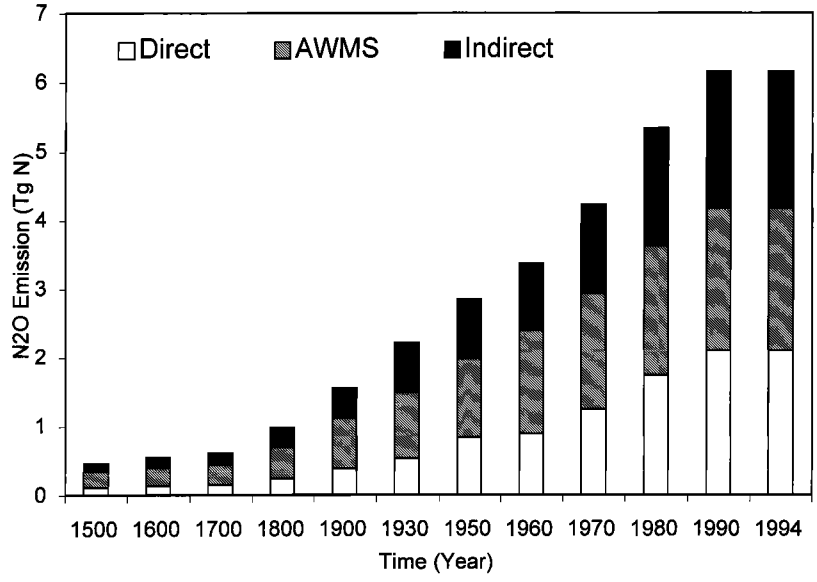

Figure 1. Global emissions of N_2O from agriculture since 1500 as calculated following the revised Intergovernmental Panel on Climate Change Guidelines for National Greenhouse Gas Inventories [*IPCC*, 1997; *Mosier et al.*, 1998]. AWMS is animal waste managament systems.

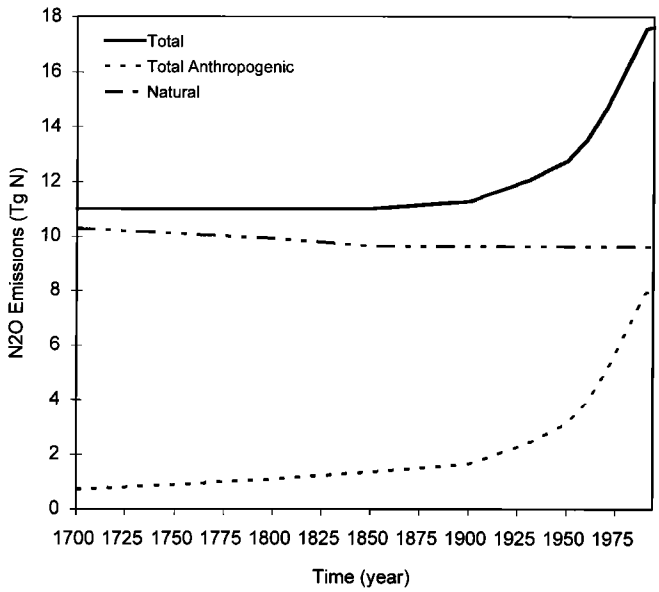

Figure 2. Global emissions of N_2O (1700 - 1994) used in the base scenario calculations. Annual emissions were linearly interpolated between the years listed in Table 3.

application of scientific knowledge to plant cultivation and animal breeding [*Grigg*, 1987]. Human ingenuity made incremental gains in technique by trial-and-error experimentation in Europe and North America, which had important implications for land use [*Singer et al.*, 1958]. Mechanization allowed for cultivation of lands that previously had been considered as unsuitable for cropping [*Buringh and Dudal*, 1987].

Another reason for the need to increase the agricultural output per unit of labor and land was the shift in demand from grain to livestock products, vegetables, and specialized tropical products, driven by fast economic growth and rising incomes of workers in industry. In addition, factory industries generated a demand for cotton, wool, jute, rubber, and vegetable oils. The railway and steamship together reduced freight costs so that grain could be shipped long distances to markets, allowing for specialization on both a local and international scale.

It is interesting to analyze how the increases in productivity could be achieved without synthetic fertilizers, which were introduced only at the end of the nineteenth century and which became widely used in agriculture after 1930 (Table 1). As mentioned above, one of the changes in production systems was the introduction of legumes (nitrogen fixing crops) in crop rotations. A second input of nitrogen became from animal manure. In some parts of Europe, animal manure (e.g., pig manure) was not used as a fertilizer. The value of animal manure as a fertilizer was increasingly appreciated [*Grigg*, 1982]. With the fast growing animal populations the recycling of animal manure formed a growing source of nutrients allowing for increasing crop yields. A third major source of nutrients was human excreta and household wastes [*Olson*, 1987], which accumulated in the growing towns in industrializing regions. The recycling of human waste has been practiced for centuries in China, enabling the maintenance of high crop productivity in rice cultivation. In Europe the need to increase productivity also induced trade in all kinds of fertilizers including human wastes (various references given by *Reijnders* [1997]). Other sources of nutrients were various industrial and town wastes and ashes.

The agricultural revolution did not occur in Asia, Africa and much of Latin America in the nineteenth century [*Grigg*, 1982]. On these continents, there was a slow agricultural evolution, resorting to the traditional methods of increasing output, including reduction of fallow land and introduction of multiple cropping, increasing the amount of labor in preparing the seedbed, weeding and harvesting, and expansion of the cultivated area. The change was slow, because the population growth was fast, and there was no parallel increase in urban employment to relieve the rural congestion. This continued increase in rural population growth gave rise to subdivision of holdings, increasing fragmentation, underemployment, and malnutrition [*Grigg*, 1982]. Farmers in Asia, Africa, and Latin America had no access to the new inputs and technologies which transformed western agriculture in the 19th century.

5.2. Natural Emissions

Our estimates for "natural" emissions (Table 3) include N_2O from soils under natural vegetation, oceans, aquatic systems, and formation in the atmosphere. As described in section 4, the decline between 1500 and 1850 balances the increase in agricultural emissions and reflects the combined effect of conversion of forests to agricultural land and reduced biogenic emissions during the little ice age [*Khalil and Rasmussen*, 1989]. From 1850 onward the total natural emission is assumed constant at 9.6 Tg N yr^{-1}, as a net effect of several trends. On the one hand, historical land use changes [*Mitchell*, 1975] and estimates of emissions from soils under natural vegetation [*Bouwman et al.*, 1993] result in a decline of this source of about 1 Tg N yr^{-1} since 1850. On the other hand we assume that this decrease of natural soil emissions is compensated by increases in N_2O formation that have not been accounted for explicitly in this analysis, including (1) enhanced N_2O formation in the atmosphere, (2) enhanced biogenic formation of N_2O induced by global warming, and (3) enhanced biogenic N_2O formation due to increasing N input from deposition of industrial NOx and NH_3 injected into the atmosphere and increasing industrial inputs of N to aquatic systems. The emissions from these sources are rather uncertain. The global amount of N_2O that results from oxidation of atmospheric ammonia (NH_3) is currently estimated at 0.3 - 1.2 Tg N yr^{-1} [*Dentener and Crutzen*, 1994] about half of which may be anthropogenic; emissions induced by global warming may amount to <0.6 Tg N yr^{-1} [*Khalil and Rasmussen*, 1989; *Kroeze*, 1994]; emissions from inland and coastal waters are estimated at 1.9 (0.9 - 9.0) Tg N yr^{-1} [*Seitzinger and Kroeze*, 1998] of which less than one fifth may be induced by industrial N inputs; emissions induced by industrial N deposition may be 0.3 Tg N yr^{-1} [*Kroeze*, 1994]. The global population has increased by about a factor 3 between 1850 and the present. Since we assume that these emissions are correlated to population, the increase of these sources may have been of the order of 1 Tg, which equals the estimated decrease of

natural soil emissions since 1850. We ignored the possibility that under certain conditions, soils and lakes may act as a sink for N_2O; although uncertain, the contribution of these sinks to global N_2O is likely to be small.

5.3. Emissions from Agriculture

It is clear that the transformations in agriculture in Europe and North America induced a fast intensification of the use of nitrogen, which has probably caused increasing losses of N_2O from crop and livestock production systems. We estimate that the N_2O emission from global agricultural systems increased from 0.6 to 1.6 Tg N_2O-N yr^{-1} between 1700 and 1900, with major increases in the nineteenth century (Table 3). This increase is the result of both expansion of the area used for agricultural production by perhaps a factor of 2 [*Buringh and Dudal*, 1987] and the intensification of the nitrogen cycling in agriculture. This intensification occurred mainly in Europe and North America. The accelerated increase in the period 1900 - 1950 is the result of agricultural development of the other continents and the rapid adoption of the use of chemical fertilizers worldwide.

It is obvious that global N input into agricultural systems from synthetic fertilizer production has increased dramatically from less than 2 Tg N yr^{-1} in 1930 to 77 Tg yr^{-1} in 1990 (about 40 times). This increase in synthetic N production has been given major responsibility for human alteration of the global N cycle [e.g., *Vitousek et al.*, 1997] and the increase in atmospheric N_2O content [*McElroy et al.*, 1977]. Total N input into agricultural soils, however, comes from synthetic fertilizer, animal excreta used as fertilizer, incorporation of crop residue from N-fixing and non-N-fixing crops and has increased only 4.1 times, from about 44 to 180 Tg N yr^{-1} during the past 60 years. Although this N is not all "newly" fixed, N_2O is emitted as organic N is mineralized. As a result, the recycling of N through crop residue and animal waste has been contributing to N_2O production throughout time. One must also be aware that crop production induces more rapid mineralization of complex organic molecules that contain N in soil organic matter. The decline of soil fertility through uptake by crops and erosion, when appropriate soil management practices are not followed, has been documented throughout history and continues today. About half (typically 30 - 70%) of the N in a crop is derived from soil organic matter mineralization. Increasing crop production therefore will increase the release of N that would have otherwise been mineralized over decades or centuries. As a result, N_2O emissions from food production continue to increase at a rate related to food production demand due to population increase. World human population increased from about 1.6 billion in 1930 to about 5.3 billion in 1990, a 3.3-fold increase. During this time, N_2O emissions from food production increased from about 2.2 to 6.2 Tg N_2O-N yr^{-1}, a 2.8-fold increase.

5.4. Total anthropogenic emissions and calculated atmospheric concentrations

In our base scenario the total N_2O emissions increased from 11 Tg N yr^{-1} in 1850 to 15 Tg N yr^{-1} in 1970 and 18 Tg N yr^{-1} in 1994. Total anthropogenic emissions (8 Tg N yr^{-1}) exceed the net increase in the atmosphere (7 Tg N yr^{-1}) because of decreasing emissions from natural systems (Figure 2). Agriculture is by far the most important anthropogenic source of N_2O (Table 3).

The calculated atmospheric concentrations are 275 ppbv in 1700, 276 ppbv in 1900, and close to atmospheric observations from 1975 in the base scenario (Figure 3). Thus, using the estimated emissions as presented in Table 3 and the model described above, the increase in atmospheric N_2O could be explained to a reasonable extent. In the base scenario the total increase in atmospheric N_2O between 1500 and 1995 amounts to 41 ppbv.

We analyzed the sensitivity of the model output to uncertainties in the model parameters (see Table 2 for scenario assumptions). Figure 3 summarizes the results of changes in the assumed preindustrial concentration, atmospheric lifetime, and the year at which emissions start to increase on the calculated N_2O concentrations. In the scenarios low 1 and 2 and high 1 and 2 the calculated 1995 concentrations differ up to 14 ppbv from the base case (Figure 3 and Table 2).

We also tested the sensitivity of the model output to uncertainties in anthropogenic emissions. As mentioned above, agriculture is the most important anthropogenic source of N_2O. This source is also relatively uncertain. *Mosier et al.* [1998] estimate that the uncertainty range for global N_2O emissions from agriculture is 1.2 - 17.9 Tg N yr^{-1}, based on uncertainties in emission factors. The lower

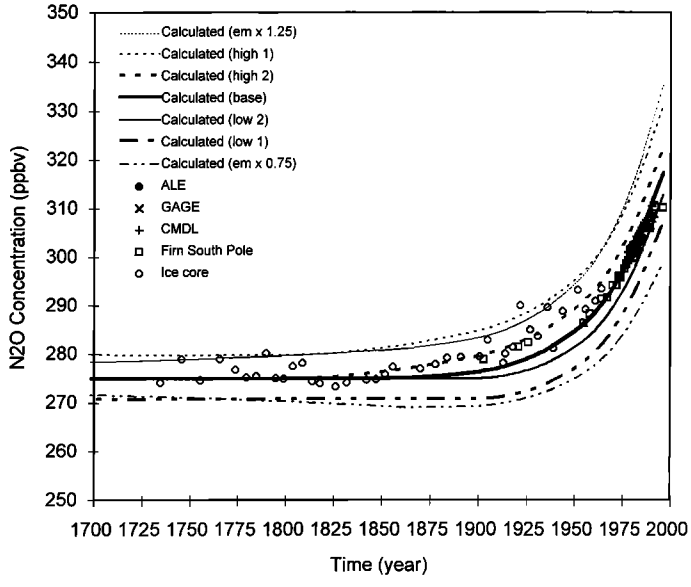

Figure 3. Simulated atmospheric concentrations of N_2O for the period 1700 - 1994 and observed concentrations in the Atmospheric Lifetime Experiment (ALE), the Global Atmospheric Gases Experiment (GAGE), the Climate Monitoring and Diagnostics Laboratory (CMDL), and concentrations deduced from firn at the South Pole [*Battle et al.*, 1996] and ice cores [*Machida et al.*, 1995]. Atmospheric concentrations are calculated for a base scenario (preindustrial concentration is 275 ppbv, atmospheric lifetime is 120 years and global emissions exceed the preindustrial level from 1850 onward) and six alternative scenarios (low 1 and 2, high 1 and 2, em x 0.75 and em x 1.25); see Table 2.

and higher end of this range were estimated by taking the lowest or highest possible values for all emission factors considered. This range should therefore be considered to reflect the theoretical range of uncertainty. Quantitative uncertainty analysis taking into account the frequency distribution of parameter values could indicate a smaller uncertainty range than the range presented by *Mosier et al.* [1998]. *Van Aardenne et al.* [1998], for instance, used Monte Carlo sampling techniques (Latin hypercube sampling) to analyze the uncertainty in calculated emissions of N_2O emissions from Dutch agricultural soils as influenced by the uncertainties in 16 parameters. Their preliminary results indicate a standard deviation equaling about 20% of the mean value. The standard deviation can be used as an indicator for the absolute uncertainty in the estimate. Here we used estimated anthropogenic emissions that differ by +/- 25% from the base scenario (Table 2 and Figure 3). This difference could also include alternatives to the assumption that the pre-1900 loss of forests were balanced by increased agricultural activities. For these two scenarios the calculated 1995 N_2O concentrations differ by up to 18 ppbv from the base scenario (Figure 3 and Table 2).

5.5. Global Budget

We estimated for the base scenario that total N_2O emissions increased from 11 Tg N yr^{-1} prior to 1850 to 11.3 Tg N yr^{-1} in 1900 (Figure 2 and Table 3). The small net additions of N_2O to the atmosphere (<0.3 Tg N yr^{-1}) throughout the nineteenth century increased to 1 Tg N yr^{-1} in 1930 (Table 3). Agricultural emissions were calculated to be the most important anthropogenic source, increasing from 1.6 Tg N yr^{-1} in 1900 to 6.2 Tg N yr^{-1} in 1994 (Figure 1). Emissions from energy use, biomass burning, and industry increased from 0.1 Tg N in 1900 to 1.8 Tg N in 1994 (Table 3).

During the 1970s and 1980s the net additions of N_2O to the atmosphere were estimated to amount to 3.7 - 6.6 Tg N yr^{-1} in the base scenario (Table 3). This is consistent with *Khalil and Rasmussen* [1992], who calculated 4 - 5 Tg N yr^{-1} for the period 1977 - 1987, based on inverse modeling and the assumption that the natural source had not changed since preindustrial times.

Our study indicates that despite still existing uncertainties, present knowledge about sources and sinks of N_2O is sufficient to explain the observed trend in atmospheric N_2O. Our budget takes a new approach to estimate N_2O emissions, accounting for the whole nitrogen cycle and relating N_2O formation to the amount of nitrogen in circulation.

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