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Nitrous Oxide Emissions from Fertilized and Unfertilized Soils in a Subtropical Region (Andalusia, Spain)

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Abstract. Field measurements of N_2O emission rates were carried out from August until October 1982 in a subtropical region in Europe, i.e. in Andalusia, Spain. The measurements were performed by using an automatic sampling and analysis technique allowing the semi-continuous determination of N_2O emission rates. The N_2O emission rates were positively correlated to the soil surface temperature and exhibited a diurnal rhythm with maximum rates in the afternoon and minimum rates in the early morning with average values of 1 μg $N_2O-N/m^2/h$ for the grass lawn and 15 μg $N_2O-N/m^2/h$ for cultivated land. Application of urea and ammonium nitrate resulted in elevated N_2O emission rates when compared to the unfertilized control. The loss of fertilizer–nitrogen as N_2O was 0.18% for urea and 0.04% for NH_4NO_3 which compares very well with data obtained in a temperate climate (Germany). The total source strength of fertilizer-derived N_2O is estimated to be 0.01–2.2 Tg N_2O-N per year. The N_2O flux from unfertilized natural soils may be as high as 4.5 Tg N_2O-N, indicating that the N_2O emission from soils contributes significantly to the global N_2O budget.

Key words: N_2O emission, nitrogen fertilizer, soil, subtropics, atmospheric budget.

1. Introduction

Nitrous oxide is an intermediate product of the denitrification and nitrification in soil (Bremner and Blackmer, 1978; Bremner and Blackmer, 1981; Schmidt, 1982; Firestone, 1982). The N_2O in soil atmosphere was found to be produced and consumed simultaneously in the uppermost soil layer, resulting in a net flux of N_2O from soil into the atmosphere (Seiler and Conrad, 1981). The flux rates are dependent on the chemical composition of the soil, soil moisture, oxygen content in soil air, and soil temperature and thus show high spatial and temporal variations (Breitenbeck *et al.*, 1980; Blackmer *et al.*, 1982; Bremner *et al.*, 1980; Matthias *et al.*, 1980; McKenney *et al.*, 1978; Mosier *et al.*, 1981; Seiler and Conrad, 1981).

Application of mineral nitrogen fertilizer results in an enhancement of the N_2O production rates in soil and, thus, an increase of the total N_2O flux from soil into the atmosphere. The loss of fertilizer–nitrogen as N_2O ranges between 0.01 and 2.0% of the applied mineral nitrogen fertilizer (Breitenbeck *et al.*, 1980; McKenney *et al.*, 1978; Mosier and Hutchinson, 1981; Conrad and Seiler, 1980) and are dependent on the type of applied mineral fertilizer, application form, etc. (Conrad *et al.*, 1983).

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Most of the measurements of N_2O emission rates from fertilized and unfertilized soils were carried out in countries with temperate climates. Data on N_2O flux rates from soils into the atmosphere in tropical and subtropical climates are lacking.

Since the nitrogen cycles in tropical and subtropical ecosystems are qualitatively and quantitatively different from those in temperate ecosystems (Clark and Rosswall, 1981), the N_2O emission rates may be different from those observed in temperate climates. Furthermore, because of the increasing application rates of mineral nitrogen fertilizers in developing countries (FAO, 1982), information on the N_2O emission in tropical and subtropical regions is of increasing importance for estimating the effect of fertilization on the global N_2O budget and, thus, on the chemistry and radiative budget of the atmosphere (Crutzen, 1970; Lacis et al., 1981). This paper reports on results from field measurements carried out on cultivated land and a grass lawn in a subtropical region of Europe, i.e. in Andalusia, Spain.

2. Experimental

Measurements on cultivated land were carried out from August until October 1982 on a cultivated field of the farm station of BASF Española near Utrera (37°N, 5.6°W) approximately 30 km south of Sevilla, Spain. The soil of the field was reddish-brown and consisted of a loamy sand with a pH value of 7.4 and a size fraction of 7% for particles with $\emptyset < 2\,\mu m$, 4% for particles with $\emptyset$ between 2–20 μm and 89% for particles with $\emptyset$ between 20–2000 μm. Soybeans were cultivated on the field during spring and summer 1981 and were ploughed under in September of the same year. Since then, until the measurements in August 1982, the field remained unplanted and did not receive any mineral nitrogen fertilizer.

The measurements on the grass-covered area were performed on the lawn of the station consisting almost exclusively of Bermuda grass (Cynodon dactylon). This grass lawn was fertilized in spring with ammonium nitrate fertilizer (75 kg N/ha) and irrigated using a lawn-sprinkler once every two or three days at a rate of a few millimeters per day. The regular irrigation was interrupted during the period of our experiments. During this time, the grass lawn received irregular watering on 18, 20, 23 and 25 August (see Figure 2).

Nitrous oxide measurements were performed on three plots on the cultivated land and on two plots on the grass lawn. Each plot had a surface area of approximately 800 cm². One plot in each experimental area remained unfertilized and was used as a control. The remaining two plots on the cultivated land were fertilized with urea and ammonium nitrate, respectively, both analytical grade (Merck, Darmstadt). The plot on the grass lawn received a commercial ammonium nitrate fertilizer (Hakaphos Naranja, BASF) including 15% P_2O_5 and 30% K_2O. The application rate was 100 kg N/ha for all cases. The fertilizers were applied as aqueous solutions. The amount of water added to the plots by this procedure was equivalent to a precipitation of 7 mm. To enable comparison of the N_2O emission rates obtained from the individual experimental plots, the unfertilized plots received the same amount of water without fertilizer. Water equivalent to a precipi-

tation of 7 mm was applied for the artificial irrigations of the individual experimental plots.

The N_2O emission rates were determined by using the closed-box technique applying the automatic sampling and analysis technique. The applied boxes as well as the sampling technique have already been described by Conrad *et al.* (1983). This technique allowed the simultaneous determination of N_2O emission rates on several soil plots and provided six individual data points per plot and day on the cultivated field with a total of three plots, and eight individual data points per plot and day on the grass lawn with a total of two plots. The lower detection limit of the N_2O emission rates was 0.2 μg $N_2O-N/m^2/h$. The precision was 0.5% at 10 μg $N_2O-N/m^2/h$.

The total loss L of applied mineral fertilizer–nitrogen as N_2O was calculated by

$$L = \frac{\int_0^t (E_i - E_0)\, dt}{M}$$

where E_i = Emission rates of N_2O-N measured on the fertilized plots.
E_0 = Emission rates of N_2O-N measured on the control plot.
M = Amount of applied mineral fertilizer–nitrogen.
t = Length of observation period.

Using this system, loss rates of $\geqslant 0.02\%$ can be determined.

The soil surface temperature was recorded at 3 to 5 cm depths using thermocouples (Iron/Constantan). The soil moisture content (g water/100 g moist soil) was determined gravimetrically in the soil samples taken from the top 5 cm soil layer of a nearby soil plot treated in the same way as the experimental plots.

3. Results

The temporal variations of the N_2O emission rates from cultivated land are illustrated in Figures 1a and 1b together with the temporal variation of the soil surface temperature and soil moisture. The soil surface temperature showed pronounced diurnal variations with maximum temperatures of 35–48°C in the afternoon and minimum temperatures of 18–24°C in the early morning. The soil moisture was extremely low during the whole observation period and generally ranged below the 2% indicative for the semi-arid climate of Andalusia during this particular season. Higher soil moisture was only observed during those periods with artificial irrigation (e.g. on 9, 16, 20, 24 September and 1 October) and rain showers on 25 September. During these days, the soil moisture of the upper 5 cm of the soil layer reached values of about 15% which, however, dropped very rapidly within 1 to 2 days to the former value of < 2%.

The N_2O emission rates measured on the unfertilized field plot showed long-term variations with a time-scale of several days superimposed by diurnal variations with maximum values in the afternoon. The N_2O emission rates varied between 4 and 35 μg $N_2O-N/m^2/h$ with an average value of 15 μg $N_2O-N/m^2/h$. This figure is considerably higher than the
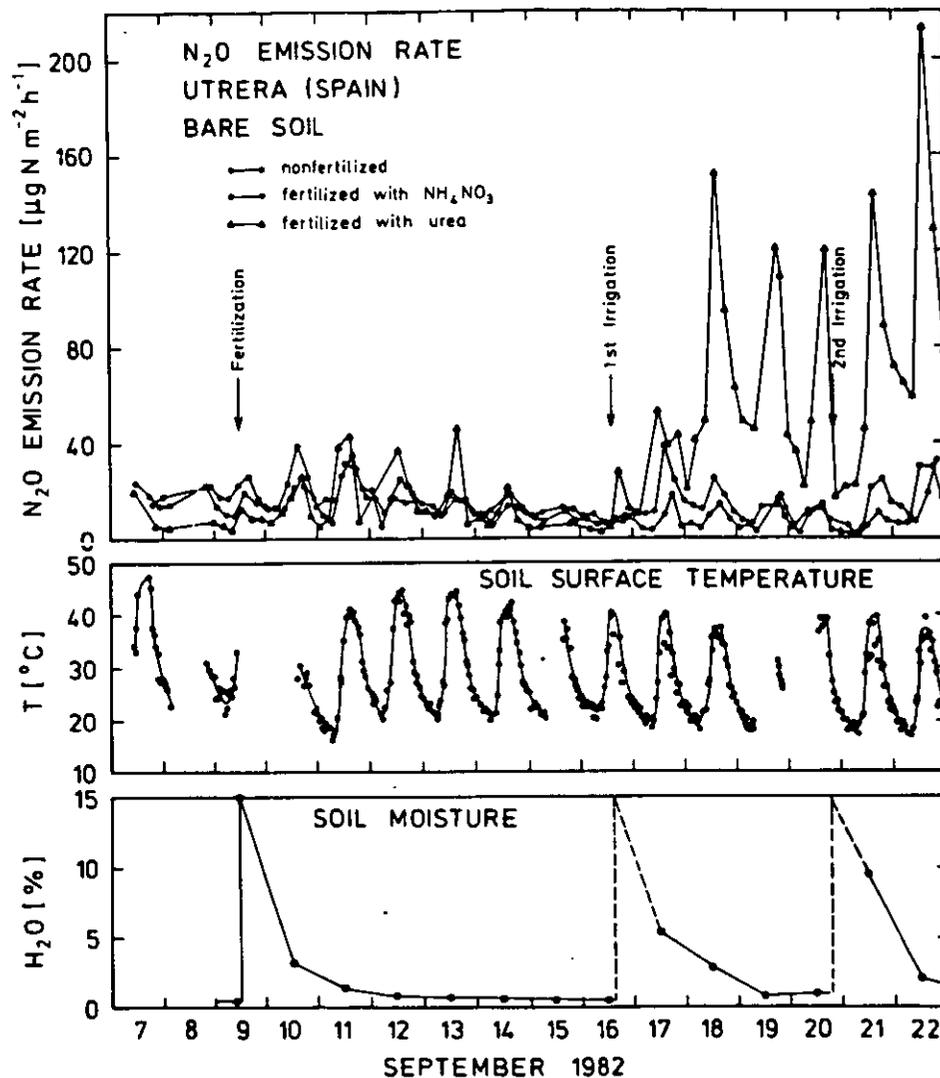
Fig. 1a. N$_2$O emission rates after fertilization with urea or ammonium nitrate. Nitrous oxide emission rates were measured between 7 September and 8 October on an unplanted field. Measurements were carried out on unfertilized soils and soils fertilized with urea or ammonium nitrate. The lower part shows the temporal variation of soil temperature at 3 to 5 mm depth and the soil moisture of the upper 5 cm.

N$_2$O emission rates observed on unfertilized field plots during our experiments in Germany where the individual figures varied between 1 and 15 μg N$_2$O–N/m^2/h with an overall average of about 4 μg N$_2$O–N/m^2/h. The latter values were calculated from data summarized in Figure 5–7 of the recently published paper by Conrad *et al.* (1983). The difference of the N$_2$O emission rates observed in Andalusia and Germany may be due to the influence of the soil temperature on the N$_2$O emission rate with higher soil tempera-
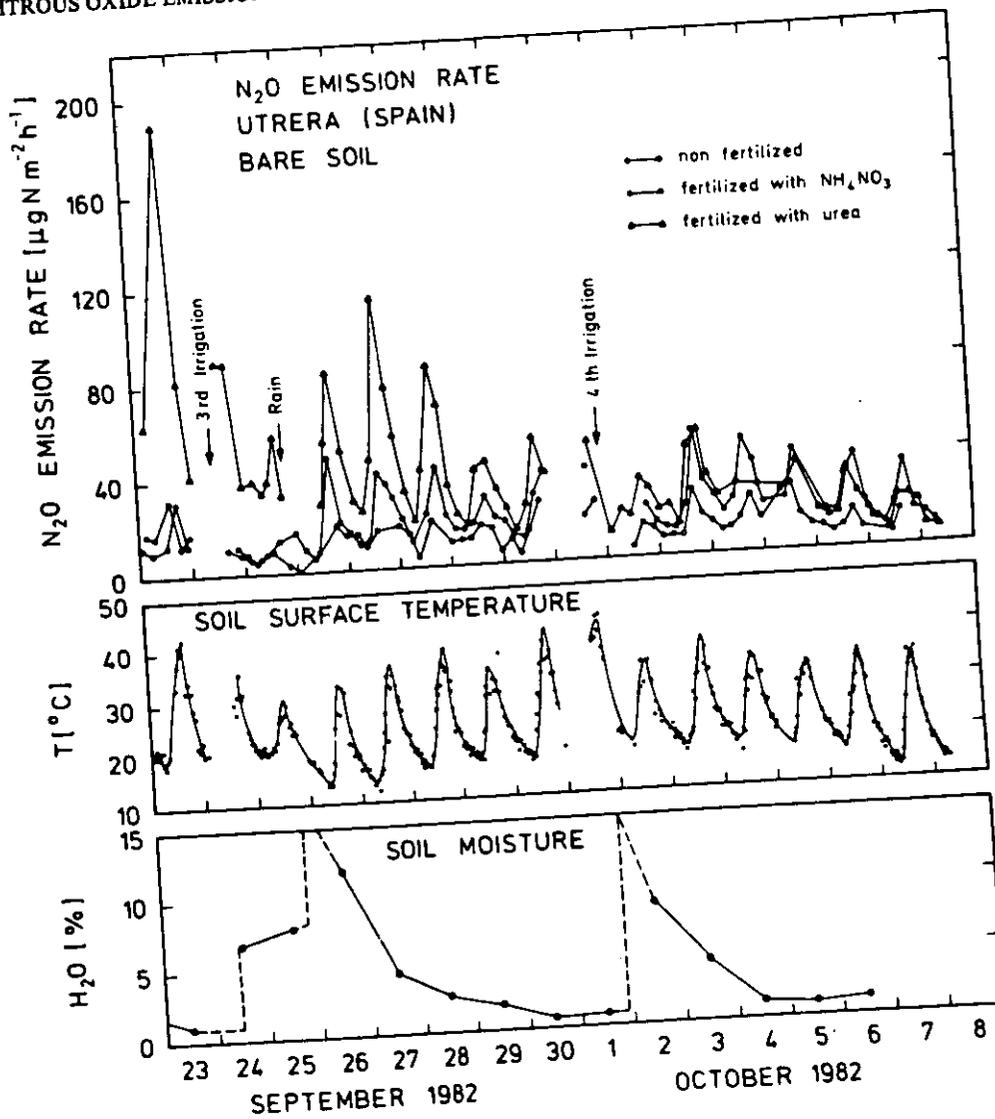
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Fig. 1b. Continuation of Figure 1a.

tures and, consequently, higher N_2O emission rates in Andalusia. It is also possible that the higher N_2O emission rates found in Andalusia are due to the plant residues of soybeans which had been ploughed under one year before the measurements and which may still provide substrate for nitrification and/or denitrification.

It seems that the N_2O emission rates measured on the unfertilized plots are influenced by irrigation and, thus, by the soil-moisture content. This effect is very pronounced for

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the period between 9 and 15 September when the N_2O emission rates increased from 20 μg $N_2O-N/m^2/h$ shortly before the irrigation to maximum values of 35 μg $N_2O-N/m^2/h$ on 11 September. The influence of irrigation on the N_2O emission decreased with an increasing number of irrigations. A similar relationship has been observed by Conrad *et al.* (1983) on unfertilized noncultivated land where the N_2O emission rates increased from about 5 μg $N_2O-N/m^2/h$ before heavy rain showers to 20 μg $N_2O-N/m^2/h$ a few days later.

Nitrous oxide emission rates measured on fertilized plots also showed strong diurnal variations with maximum values in the afternoon and minimum values in the early morning. Very often, the maximum values exceeded the morning values by more than a factor of 3–5. The daily amplitudes as well as the daily average of the N_2O emission rates were found to be dependent on the magnitude and the variation of the soil surface temperature. Significant positive correlations between N_2O emission and soil-surface temperature were found on 48–57% of the total observation period (see Table I). The N_2O emission rates were also positively correlated with the soil moisture, with relatively low values at soil moistures lower than 5%.

Most surprisingly, the N_2O emission rates measured on the fertilized plots did not respond to the application of mineral fertilizers within the first week after fertilization. During this period, the N_2O emission rates were comparable to those obtained on the unfertilized plot. A substantial increase of the N_2O emission rates was observed on the experimental plot fertilized with urea after the first irrigation, which was seven days after fertilization. Maximum N_2O emission rates occurred on 22 September after the second irrigation. After the fourth irrigation, the N_2O emission rates approached the values observed at the unfertilized plot.

The temporal pattern of the fertilizer-induced N_2O emission is in contrast to the findings by different groups in temperate climates and higher soil moistures (for summary see Conrad *et al.*, 1983) which always show a very fast response of the N_2O emission rates on the application of nitrogen fertilization. Generally, the fertilizer-induced N_2O emission rates reached their maximum values one to five days later and approached the background values 10 to 12 days after fertilization.

The total loss of mineral fertilizer–nitrogen as N_2O is calculated to be 0.18% for urea and 0.04% for ammonium nitrate.

Table I. Activation energy of N_2O emission from unfertilized and fertilized soil of a bare field in Utrera, Spain (9 September–7 October, 1982)

Soil conditions (100 kg N ha^{-1})	Days with significant correlation ($P < 0.05$)	Activation Energy (kJ mole^{-1})	
		Range	Mean value
without fertilizer	6 (28%)	21–50	36.3 ± 10.7
NH_4NO_3	12 (57%)	26–64	47.2 ± 11.3
Urea	10 (48%)	40–75	55.0 ± 12.6

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The N₂O emission rates obtained from measurements on the grass lawn are summarized in Figure 2 together with the soil surface temperature. The data obtained on the un-fertilized plots show a large scatter of the individual emission rates and also very low N₂O flux rates with values of < 2.0 μg N₂O–N/m²/h. Despite the high scatter of the data, it seems that the daytime N₂O emission rates were higher than the nocturnal ones. The most
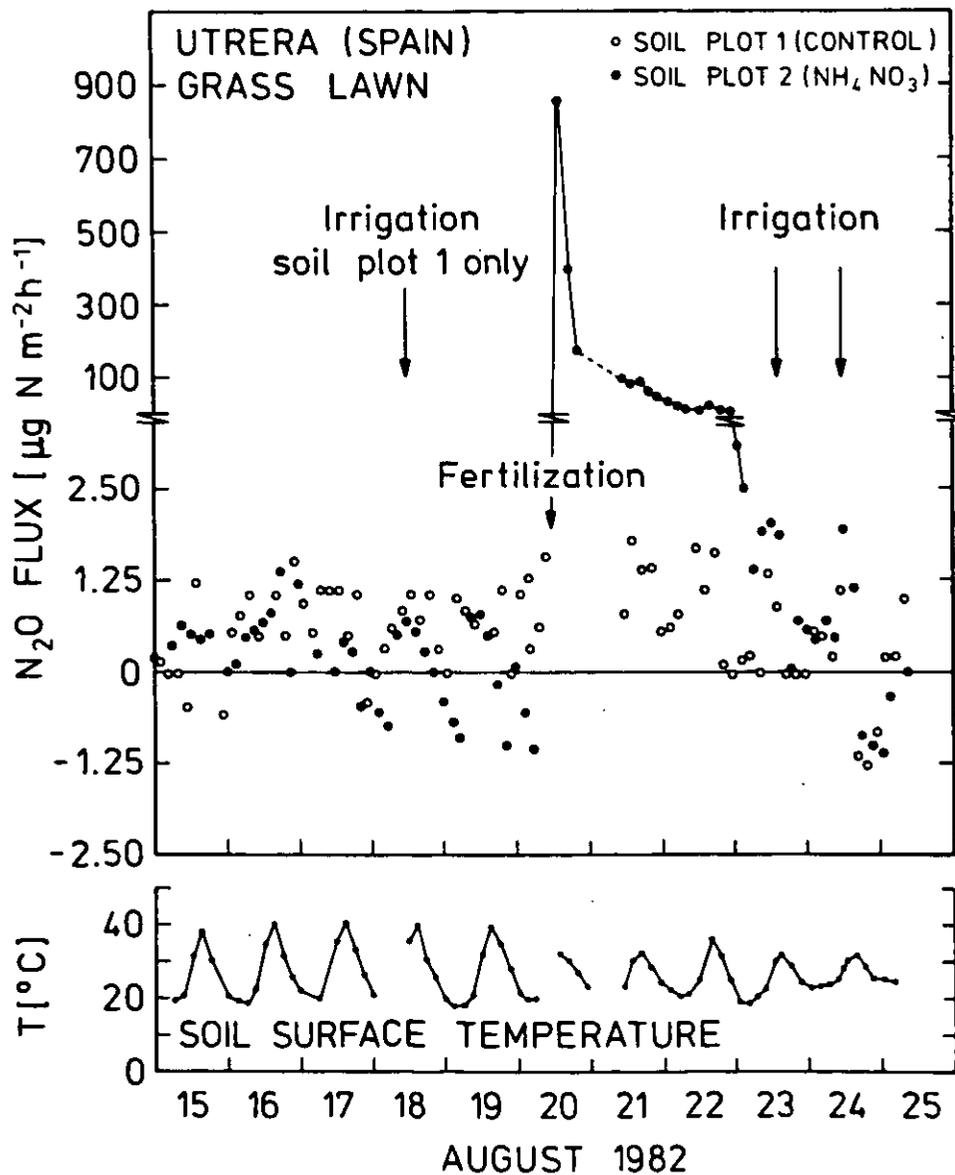

Fig. 2. N₂O emission rates measured on a Bermuda grass lawn. Plot 1 remained unfertilized; plot 2 was fertilized with ammonium nitrate (75 kg N/ha).

interesting result is the observation of negative fluxes at the soil-air interface indicating that the soil of the grass lawn acted both as a source and sink of atmospheric N_2O.

In contrast to the observations on the cultivated field plots, the fertilization of the grass lawn with NH_4NO_3 resulted in a rapid increase of the N_2O emission rates. Maximum rates of 850 μg $N_2O-N/m^2/h$ were reached about 6 h after fertilization. The background values of 1 μg $N_2O-N/m^2/h$ were approached 2 days after fertilization. Repeated irrigation of the grass lawn after fertilization with amounts of H_2O equivalent to 7 mm rainfall did not result in any significant enhancement of the N_2O emission rates. Despite these differences, the total loss of mineral fertilizer—nitrogen as N_2O (0.075%) agrees reasonably well with the corresponding figure (0.040%) obtained from the plot on the cultivated land.

4. Discussion

N_2O emission rates measured on fertilized and unfertilized soils are positively correlated with the soil surface temperatures indicating that the N_2O production processes must be located within the upper centimeters of the soil layers. Nitrous oxide production in deeper layers would cause a considerable time delay between the N_2O emission rates at the soil-air interface and the soil surface which, however, have never been observed. Similar findings were already reported for soils in Germany (Conrad et al., 1983) and in Australia (Denmead et al., 1979). The only contradictory data were published by Blackmer et al. (1982) who found a phase lag of 2 to 12 h between the maximum N_2O emission rates and the maximum soil temperature measured in a depth of 2 cm. They, therefore, concluded that the N_2O production must have occurred in layers considerably lower than 2 cm.

Using the data obtained on individual days with positive correlations between the N_2O emission and soil temperature and applying the Arrhenius equation, the activation energies are calculated to be 36–55 kJ/mole which are significantly lower than the values of 60–76 kJ/mole obtained from measurements in Germany (Conrad et al., 1983). This difference may be explained by the possibility that the N_2O production in hot soils of arid climates may occur at slightly lower depths than in soils of midlatitudes.

In general, the soils both fertilized and unfertilized acted as a source of atmospheric N_2O. Exceptions were measurements on a grass lawn where small N_2O uptake rates were observed during nighttime. N_2O uptake was also observed by Ryden (1981) on soils covered with perennial ryegrass (Lolium perenne) which gives a similar lawn structure as the Bermuda grass (Cynodon dactylon) used for our experiments. Small N_2O sink activities have also been observed by Bremner et al. (1980) in spring on soils in Iowa (U.S.A.) which, however, are of minor importance for the global N_2O cycle.

Average N_2O emission rates on unfertilized soils varied between 1 μg $N_2O-N/m^2/h$ on the grass lawn and 15 μg $N_2O-N/m^2/h$ on the cultivated land. The latter figure is higher than the values of about 4 μg $N_2O-N/m^2/h$ observed on unfertilized soils at midlatitudes in Germany (Conrad et al., 1983). This difference may be explained either by the higher soil temperatures in arid climates and/or by the incorporation of plant residues

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of soybeans into the soil carried out one year before our measurements in Andalusia. Assuming the figures of 15 μg $N_2O-N/m^2/h$ to be representative for subtropical and tropical regions with a total surface area of 25×10^6 km^2 (Lieth, 1975), the total N₂O emission from unfertilized soils of this climatic region may account for 3 Tg N_2O-N/yr. Correspondingly, the N₂O emission from soils of temperate climates (area 45×10^6 km^2) applying an average N₂O emission rate of 4 μg $N_2O-N/m^2/h$ is calculated to be 1.5 Tg/yr so that the total N₂O flux from unfertilized soils may be as high as 4.5 Tg N_2O-N/yr.

Fertilization resulted in an increase of the N₂O emission rates reaching maximum values of 850 μg $N_2O-N/m^2/h$ on the grass lawn fertilized with NH_4NO_3 and values up to 200 μg $N_2O-N/m^2/h$ on the cultivated land fertilized with urea. In the case of regularly sprinkled humid soil of the grass lawn, the maximum N₂O emission rates were observed 6 h after fertilization. Two days later, the N₂O emission rates reached background values indicating that nitrification and/or denitrification of mineral fertilizer nitrogen had approached completion. This might be due to either the nitrogen uptake by the grass vegetation or by microorganisms associated with rhizosphere or by the leaching of nitrogen out of the root zone. The influence of vegetation and rhizosphere on denitrification and, thus, on the N₂O production, has recently been discussed by Firestone (1982) and Knowles (1982). Tiedje (1982) has pointed out that denitrification rates in the rhizosphere are only high as long as high nitrate concentrations are present. As soon as nitrate concentrations are low, the denitrification rates are much lower on vegetated than on unplanted soils. This finding is consistent with our observations of lower N₂O emission rates at the grass lawn relative to those at the cultivated land under unfertilized conditions.

In the case of cultivated land, the maximum N₂O emission rated did not occur until 13 days after fertilization with urea. This time lag may be caused by the low soil moistures which were generally lower than 5% and thus low denitrification and nitrification activities. Another explanation may be the gradual degradation of the applied urea by urea to NH_3 (Mulvaney and Bremner, 1981) that is necessary to initiate nitrification. A similar delay of fertilizer induced N₂O emission after treatment with urea was observed by Mosier et al. (1981) and Duxbury et al. (1982).

The loss of mineral fertilizer–nitrogen as N₂O obtained from the measurements in Andalusia varied between 0.04 and 0.075% for application of ammonium nitrate and 0.18% for application of urea. The higher loss rates by application of urea may be due to the hydrolysis of urea into NH_3 that increases the alcalinity and cation binding capacity of the soil (Boomsma and Pritchett, 1979) and thus stimulates nitrification and N₂O production (Bremner and Blackmer, 1981). Although the climatic conditions in Andalusia and Germany are quite different, the loss of mineral fertilizer-nitrogen as N₂O measured in both regions agree reasonably well with values of 0.04–0.18% for Andalusia and 0.01–0.38% for Germany (Conrad et al., 1983). Based on these observations, it might be possible that the total loss of mineral fertilizer-nitrogen, as N₂O does not strongly depend on the climatic conditions. This would mean that the values of 0.01–2% as proposed by Conrad et al. (1983) for different types of mineral fertilizers, different application rates, and different forms of application may be representative for global conditions. In this

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case, the total source strength of fertilizer-derived N_2O would be in the range of 0.015–2.2 Tg N_2O–N/yr for present conditions.

Summing up, we receive a total N_2O emission of 4.5–7.7 Tg N_2O–N/yr from unfertilized and fertilized soils. Because of the limited data basis, the given figure is rather preliminary, but indicates that the soils may provide 50% of the N_2O source rate presently estimated to be 8–15 Tg N_2O–N/yr (Schmeltekopf et al., 1977; Johnston et al., 1979).

Acknowledgements

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