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# Field Measurements of Emission of Nitric Oxide from Fertilized and Unfertilized Forest Soils in Sweden

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**Abstract.** Application of nitrate fertilizers on two types of forest soils led to a marked increase in the NO emission rate indicating a large potential for NO production in these soils. The largest fluxes on the fertilized plots were up to 60 ng NO–N $m^{-2}$ $s^{-1}$. About 0.35% of the applied nitrogen was lost as NO within about 14 days after fertilization. The fluxes from the unfertilized forest soils were in the range 0.1 to 0.8 ng NO–N $m^{-2}$ $s^{-1}$ with a median value of 0.3 ng NO–N $m^{-2}$ $s^{-1}$. If this value, obtained during June and August to September, is representative for the growing season (150 days), it corresponds to an annual emission of 0.04 kg NO–N $ha^{-1}$. This is about 30% of the value obtained for an unfertilized agricultural soil. Because of the large areas occupied by forests in Sweden the flux of NO from forest soils represents a significant contribution to the total flux of NO from soils in Sweden.

Earlier observations of equilibrium concentrations for NO have been verified. These were found to range from 0.2 to 2 ppbv for an unfertilized forest soil and up to 170 ppbv for a fertilized soil. At the rural site in Sweden where these measurements were performed the ambient concentrations where found to be less than this equilibrium concentration, and consequently there was generally a net emission of NO.

There are still large uncertainties about the global flux of NO from soils. Using direct measurements on three different types of ecosystems and estimates based on a qualitative discussion for the remaining land areas, a global natural source for NO of the order of 1 Tg N $a^{-1}$ was obtained. If 0.35% of the total annual production of fertilizer nitrogen is lost as NO, fertilization of soils may contribute with 20% to the natural flux from soils.

**Key words.** Nitric oxide, emission, forest soils, fertilized, unfertilized.

## 1. Introduction

Production of nitric oxide (NO) in soils represents an important source of $NO_x$ in the lower atmosphere (Galbally and Roy, 1978; Galbally *et al.*, 1980: Logan *et al.*, 1981). In order to achieve an estimate of the global flux of NO from the ground, field measurements on different types of soils are needed. Earlier measurements have been performed in different ecosystems: forests (Kim, 1973), pastures (Galbally and Roy, 1978) and arable land (Johansson and Granat, 1984). In these measurements NO emissions ranging from less than 0.1 ng NO–N $m^{-2}$ $s^{-1}$ up to 70 ng NO–N $m^{-2}$ $s^{-1}$ have been observed.

Some progress has been made in the investigation of how environmental factors such as soil temperature, soil moisture and nitrate content in the soil affect the NO emission

*in situ* (Galbally and Roy, 1978; Johansson and Granat, 1984). It has been shown that NO emission is higher on grazed or fertilized soils than on ungrazed or unfertilized soils. Our earlier measurements on arable land (Johansson and Granat, 1984) have shown that for the vegetation period the NO emission was three times higher on the fertilized area (200 kg N $ha^{-1}$) than on the unfertilized area. A rough estimate showed that approximately 0.2% of the applied nitrate nitrogen ($Ca(NO_3)_2$) was lost as NO.

Recent laboratory studies using gas flow through soil columns, where the NO loss is maximized, have shown that as much as 30 to 40% of the $NO_3^-$–N may be lost as NO within less than 20 hours under anaerobic conditions (McKenney *et al.*, 1982; Johansson and Galbally, 1984). Earlier laboratory studies report nitrogen losses in the range 1 to 5.3% of the applied nitrogen after the addition of ammonium or urea fertilizers (Steen and Stojanovic, 1971; Bundy and Bremner, 1974; Keeney *et al.*, 1970; Marshall and Debell, 1980).

This paper presents field measurements of fertilizer-induced NO emission. The flux of NO from both unfertilized and from freshly fertilized forest soils was measured at two different sites.

## 2. Experimental Sites and Method

The measurements were made in two different forests, at Sörentorp, 10 km north of Stockholm, and at Jädraås, 220 km northeast of Stockholm. The characteristics of the sites and the concentrations of ammonium ($NH_4^+$), nitrate ($NO_3^-$) and nitrite ($NO_2^-$) in the surface (upper 2 cm) of the soil are given in Table I.

The site at Jädraås is a 20- to 25-year-old stand of Scots pine (Pinus sylvestris) at the Swedish Coniferous Forest Project site, Invantjärnsheden. The ground vegetation is of a dry to very dry dwarf shrub type. The soil is an iron podzol on sandy sediments. Further characteristics regarding vegetation cover, nutrient status of the soil and climatic data are given in Axelsson and Bråkenhielm (1980). The site at Sörentorp is a rather open park-

Table I. Characteristics of the two sites

| Site | Vegetation | Soil type and texture (10 cm depth) | pH | $NH_4^+$ | $NO_3^-$ | $NO_2^-$ |
|---|---|---|---|---|---|---|
| | | | | (mg N/100 g dry soil) | | |
| Sörentorp | Scots pine mixed with Norway spruce, silver birch and aspen. Undergrowth of grasses and herbs | Grey-brown podzolic soil. 15% clay, 76% silt, 9% sand | 4.5 | 3.0 | <0.01 | 0.015 |
| Jädraås | Scots pine heath on sand. Undergrowth of dwarf shrubs and lichens | Iron podzol. 2% clay, 43% silt, 46% sand | 4.0 | 1.6 | <0.01 | 0.094 |

like forest dominated by Scots pine mixed with Norway spruce, silver birch and aspen. The stand is approximately 75–80 years old. Undergrowth consists mainly of grasses, some herbs and small shrubs. The ground surface is covered with a carpet of mosses. The soil is an intermediate type between podzol and brown earth and could be characterized as grey-brown podzolic soil.

The flux measurements were made using the chamber technique described in Johansson and Granat (1984). With this method a chamber is placed over the surface and the increase in the concentration of NO during the first few minutes is monitored. The emission rate is calculated from the increase in NO concentration right after the chamber is installed over the soil. The chamber was an aluminium cylinder with the inside walls lined with a teflon film. The air in the chamber was stirred with a stainless steel stirrer blade. The volume of the chamber was 110 l and the covered area 0.5 $m^2$. An area of approximately 25 $m^2$ was selected at each site. Within this area six aluminium frames were inserted approximately 5 cm into the soil. The air inside the box was sealed with a rubber collar attached to the frames and the chamber. Two plots were fertilized, two treated with just water and two plots were untreated. Each plot was 1 $m^2$ and contained one aluminium frame (each 0.5 $m^2$). At Jädraås the frames were placed on plots covered with Cladonia lichens, and at Sörentorp they covered plots with grasses and mosses. Soil surface temperature was measured a few meters beside the flux measurements at about 2 cm depth. In the first and second fertilization calcium nitrate ($Ca(NO_3)_2$) was applied (4.64 g $NO_3^-$–N $m^{-2}$), and in the third sodium nitrate ($NaNO_3$) was applied (1.12 g $NO_3^-$–N $m^{-2}$). The $Ca(NO_3)_2$ fertilizer contained 14.5% $NO_3^-$–N and 1% $NH_4^+$–N; thus the $Ca(NO_3)_2$ fertilized plots also received 0.32 g $NH_4^+$–N $m^{-2}$. The fertilizers were dissolved in water, and 10 l $m^{-2}$ was added.

Nitric oxide was analyzed with a modified chemiluminescent NO detector. Calibration was made using cylinders containing 1.02±0.05 ppmv ($10^{-6}$ v/v) and 50.5±1.0 ppmv NO. The sensitivity was ±0.10 ppbv ($10^{-9}$ v/v). The least detectable flux when the chamber was allowed to cover the soil for 10 min was ~0.07 ng NO–N $m^{-2}$ $s^{-1}$. The mechanical and chemical analyses of the soil were made at the National Laboratory for Agricultural Chemistry, Uppsala, Sweden. Levels of $NO_3^-$ and $NO_2^-$ were determined by an automated colorimetric method (Technicon, 1973). In this procedure $NO_3^-$ was first reduced to $NO_2^-$ with a copper-cadmium reductor column. Ammonium was analyzed using the indophenol blue method.

## 3. Results and Discussion

When the chamber was placed on the same plot several times in succession and removed for short periods (1–2 min) in between, the estimated rate varied slightly with a standard deviation of less than 6% of the mean value (Table II).

Table III gives the results of the emission measurements on the unfertilized plots at Sörentorp and Jädraås. The fluxes at the two different sites were similar in magnitude. All fluxes were in the range 0.10 to 0.76 ng NO–N $m^{-2}$ $s^{-1}$, and the median value of all data (82 measurements on 12 different plots) was 0.29 ng NO–N $m^{-2}$ $s^{-1}$. The mag-

Table II. Successive measurements of NO emission on the same plot

| Local time | Soil surface temperature (°C) | NO emission (ng NO–N $m^{-2}$ $s^{-1}$) | Average | Standard dev. |
|---|---|---|---|---|
| 0943 | 15.8 | 3.17 | 3.42 | 0.19 |
| 0947 | 15.6 | 3.48 | | |
| 0951 | 16.0 | 3.63 | | |
| 0945 | 15.5 | 4.99 | 5.14 | 0.12 |
| 0949 | 15.9 | 5.14 | | |
| 0954 | 16.1 | 5.29 | | |
| 1544 | 11.4 | 6.29 | 6.52 | 0.21 |
| 1549 | 11.4 | 6.48 | | |
| 1554 | 11.4 | 6.99 | | |
| 1558 | 11.4 | 6.48 | | |
| 1603 | 11.4 | 6.29 | | |
| 1608 | 11.4 | 6.76 | | |
| 1614 | 11.5 | 6.38 | | |
| 1619 | 11.5 | 6.57 | | |
| 1625 | 11.5 | 6.38 | | |
| 1630 | 11.5 | 6.57 | | |

nitude of these emissions is smaller than earlier reported measurements on forest soils (Kim, 1973) and pastures (Galbally and Roy, 1978). It is about 30% of the value obtained previously for an unfertilized agricultural soil (Johansson and Granat, 1984).

On the freshly fertilized plots the emission increased dramatically, indicating a large potential for NO production in these soils. Figures 1, 2 and 3 show the temporal change in NO emission together with soil surface temperature on the three fertilized areas, the first at Sörentorp and the second and third at Jädraås. The duplicate plots showed very similar development of maximum fluxes and subsequent temporal decrease of emission. The emissions from the controls (watered plots) were generally in the range of the

Table III. Summary of the measurements performed on the unfertilized plots at Sörentorp (June) and Jädraås (August and September)

| Site | No. of measurements | Soil surface temperature (range, °C) | NO emission (ng NO–N $m^{-2}$ $s^{-1}$) Range | Median | Standard dev. |
|---|---|---|---|---|---|
| Sörentorp | 35 | 11.3–22.5 | 0.10–0.76 | 0.35 | 0.15 [a] |
| Jädraås | 47 | 9.0–17.3 | 0.10–0.56 | 0.23 | 0.04 [b] |
| All | 82 | 9.0–22.5 | 0.10–0.76 | 0.29 | 0.09 |

[a] Average value on 5 plots.
[b] Average value on 4 plots.

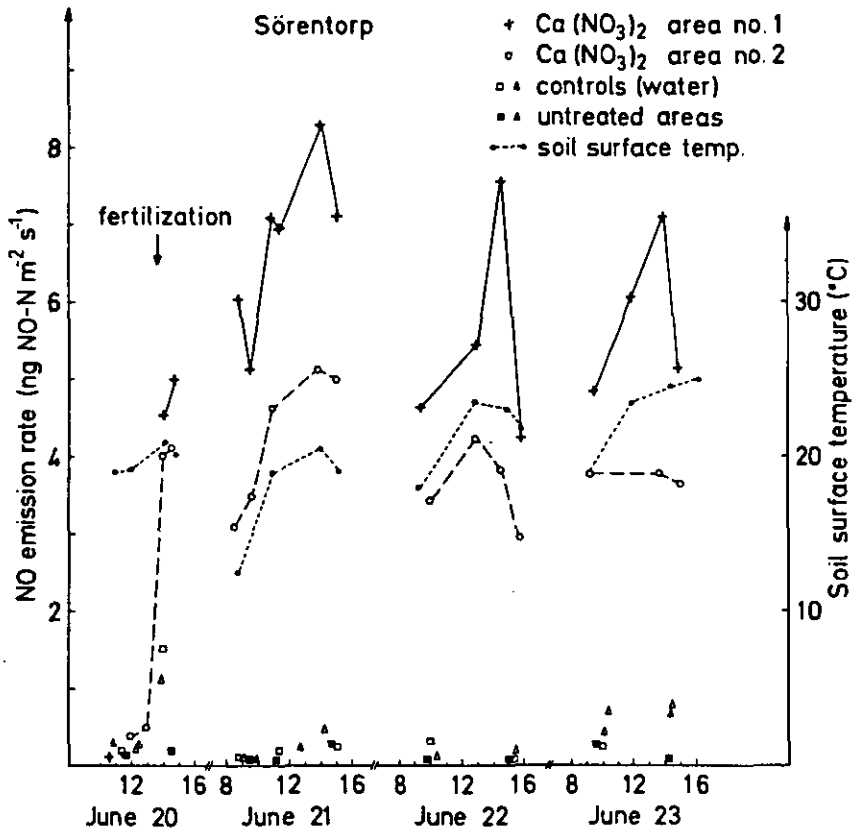


Fig. 1. Temporal change of the NO emission rate after calcium nitrate fertilization at Sörentorp (4.64 g $NO_3^-$–N $m^{-2}$ was applied).

emissions from the untreated plots. Only immediately after the soil was watered in the first experiment did the emission increase slightly; it returned to the background values in a short period of time (cf. Figure 1). The NO emission was observed to increase within only 10 min after fertilization. The maximum fluxes were observed after about 20 h. As is shown in Figures 1 and 3 the NO fluxes were still above the control and untreated plots when the measurements were interrupted, indicating that the fertilizer-induced NO production continued.

Figures 1 and 2 show a strong dependecy of the NO emission rate on the temperature at the soil surface. This is in agreement with what has been found earlier for arable land (Johansson and Granat, 1984). On the unfertilized plots, where the emission was much lower, the covariation with temperature was not so evident. The soil surface temperature ranges were 11.3–22.5 °C and 9–17.3 °C at Sörentorp and Jädraås, respectively.

The magnitude of fertilizer-induced emission was quite different at the two sites. Comparing fluxes at similar temperaures, the maximum values at Jädraås amounted to

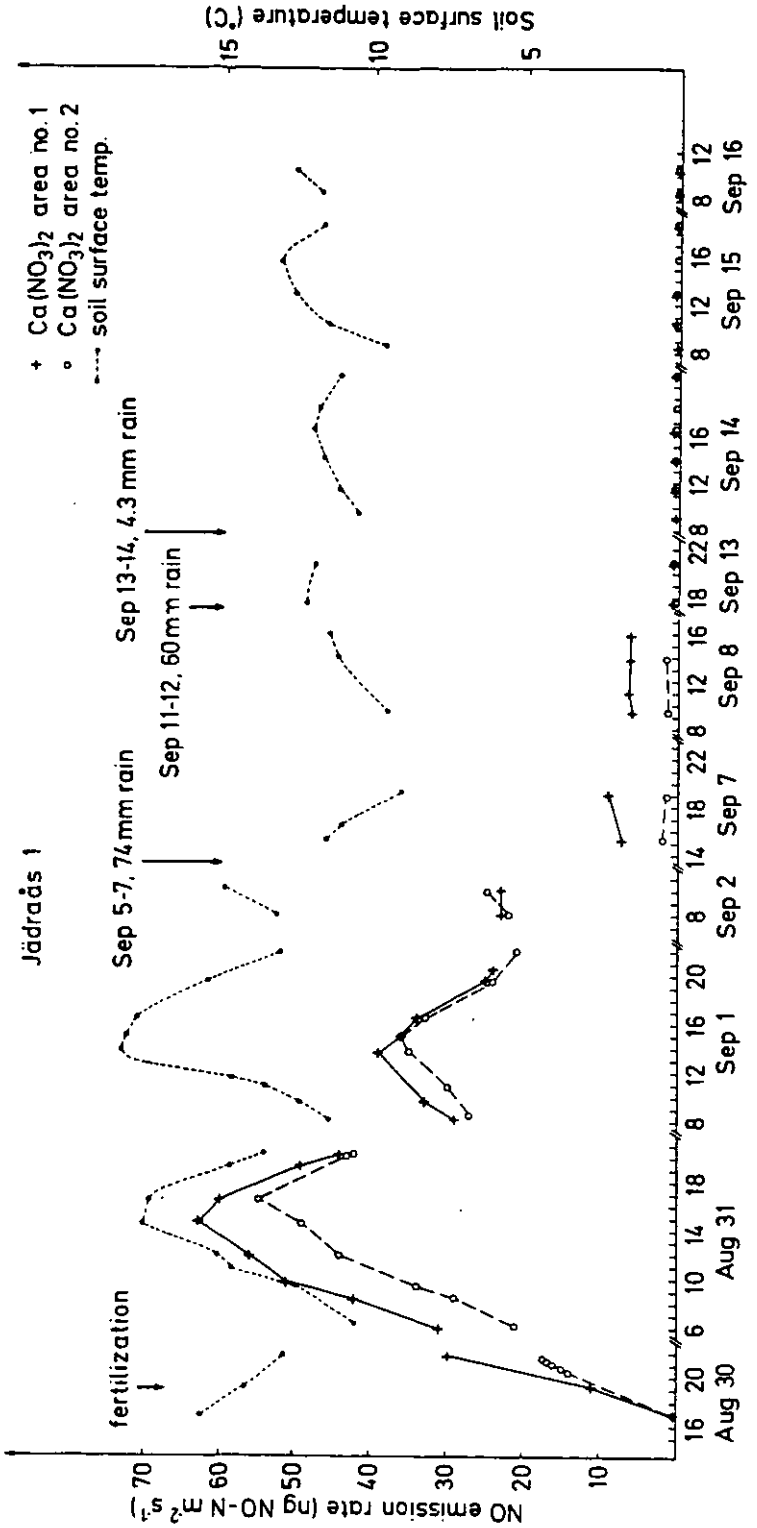


Fig. 2. Temporal change of theNO emission rate after calcium nitrate fertilization at Jädraäs (4.64 g $NO_3^-$–N $m^{-2}$ was applied).

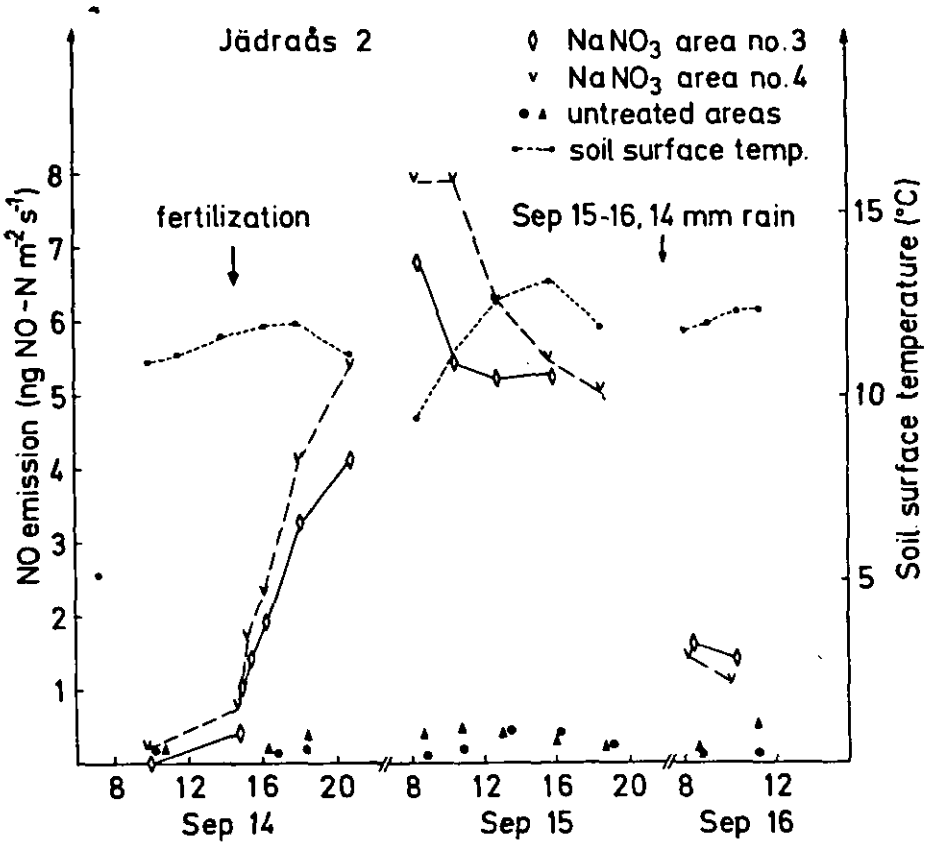


Fig. 3. Temporal change of the NO emission rate after sodium nitrate fertilization at Jädraås (1.12 g $NO_3^-$–N $m^{-2}$ was applied).

about 40 ng NO–N $m^{-2}$ $s^{-1}$ (Figure 2), whereas the maximum fluxes at Sörentorp were only about 6 ng NO–N $m^{-2}$ $s^{-1}$ (Figure 1). This difference may be attributed to both different soils and different types of vegetation covering the soils.

Emission rates and loss of nitrogen as NO were both lower on the $NaNO_3$ fertilized plots than on the $Ca(NO_3)_2$ fertilized plots at the same site (Figures 2 and 3, Table VI). As the $NaNO_3$ fertilized plots received 1.12 g $NO_3^-$–N and the $Ca(NO_3)_2$ fertilized plots 4.64 g $NO_3^-$–N, this indicates that the NO production was highly dependent on the amount of $NO_3^-$ applied. Comparing the emission rates at equal temperatures (Figures 2 and 3), the emission rate from the calcium $Ca(NO_3)_2$ fertilized plots was approximately 7 times higher than the emission rate from the $NaNO_3$ fertilized plots.

As has been pointed out, the $Ca(NO_3)_2$ fertilized plots received 0.32 g $NH_4^+$–N $m^{-2}$. Earlier laboratory studies have indicated that the NO production in soils under aerobic conditions is independent of the $NH_4^+$ concentration (Johansson and Galbally, 1984). In addition, field measurements on different plots on an agricultural soil fertilized with equal amounts of $NH_4^+$ –N or $NO_3^-$ –N showed higher NO emissions from the $NO_3^-$

Fig. 2. Temporal change of the NO emission rate after calcium nitrate fertilization at J[illegible] as ap[illegible] 4.64 g [illegible]$_3$–N

Table IV. Ammonium, nitrate and nitrite concentrations in the surface soil at Sörentorp. Fertilization with $Ca(NO_3)_2$ was performed June 20

| Date | Treatment | Plot No. | $NH_4^+$ | $NO_3^-$ | $NO_2^-$ |
|---|---|---|---|---|---|
| | | | (mg N/100 g dry soil) | | |
| June 16 | Untreated | 1 | 2.5 | <0.01 | 0.017 |
| | | 2 | 1.9 | <0.01 | 0.015 |
| | | 3 | 4.0 | <0.01 | 0.019 |
| | | 4 | 4.7 | <0.01 | 0.015 |
| | | 5 | 1.8 | <0.01 | 0.011 |
| June 21 | Fertilized | 1 | 5.5 | 6.1 | 0.027 |
| | | 2 | 4.5 | 2.2 | 0.052 |
| | Watered | 5 | 2.8 | <0.01 | 0.019 |

fertilized plots. Thus, it seems improbable that the small amount of $NH_4^+$ present in the $Ca(NO_3)_2$ fertilizer could have generated such a large difference between the $Ca(NO_3)_2$ and $NaNO_3$ fertilized plots. Although the soil has a low water-retaining capacity, it might have been caused by different soil moistures, since the $NaNO_3$ fertilizer was added after a period with two heavy rainfalls (cf. Figure 2).

The $NO_2^-$ contents of the soils at Jädraås and Sörentorp were very low. After the addition of $Ca(NO_3)_2$ fertilizer, the $NO_2^-$ concentration increased at both sites, although the increase was much larger at Jädraås (Tables IV and V). The highest $NO_2^-$ concentrations were observed at Jädraås 14 days after $Ca(NO_3)_2$ fertilization. At that time the observed NO fluxes had returned to background values. This seems to indicate that the NO production in this soil was not the result of $NO_2^-$ decomposition.

In Figures 4, 5 and 6 the cumulative fluxes are plotted versus time. The losses of $NO_3^-$–N as NO are given in Table VI. The differences in the cumulative fluxes from the

Table V. Ammonium, nitrate and nitrite concentrations in the surface soil at Jädraås. Fertilization with $Ca(NO_3)_2$ was performed August 30

| Date | Treatment | Plot No. | $NH_4^+$ | $NO_3^-$ | $NO_2^-$ |
|---|---|---|---|---|---|
| | | | (mg N/100 g dry soil) | | |
| Aug. 31 | Untreated | 5 | 0.17 | <0.01 | 0.036 |
| | Watered | 3 | 2.0 | <0.01 | 0.087 |
| | | 4 | 0.49 | <0.01 | 0.061 |
| | Fertilized | 1 | 5.9 | 26 | 0.32 |
| | | 2 | 1.5 | 8.4 | 0.15 |
| Sept. 14 | Untreated | 5 | 3.0 | <0.01 | 0.15 |
| | Fertilized | 1 | 6.5 | 0.22 | 0.57 |
| | | 2 | 1.3 | <0.10 | 0.24 |

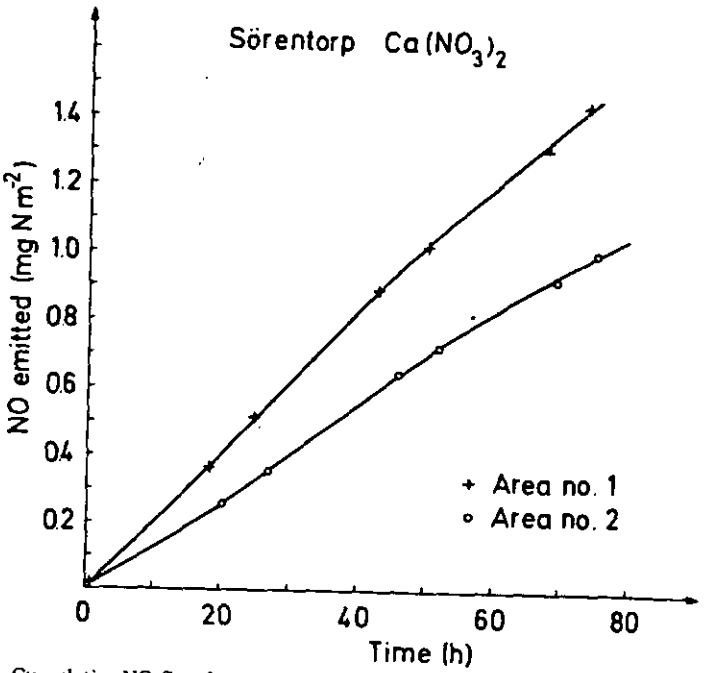


Fig. 4. Cumulative NO flux from the calcium nitrate fertilized plots at Sörentorp.

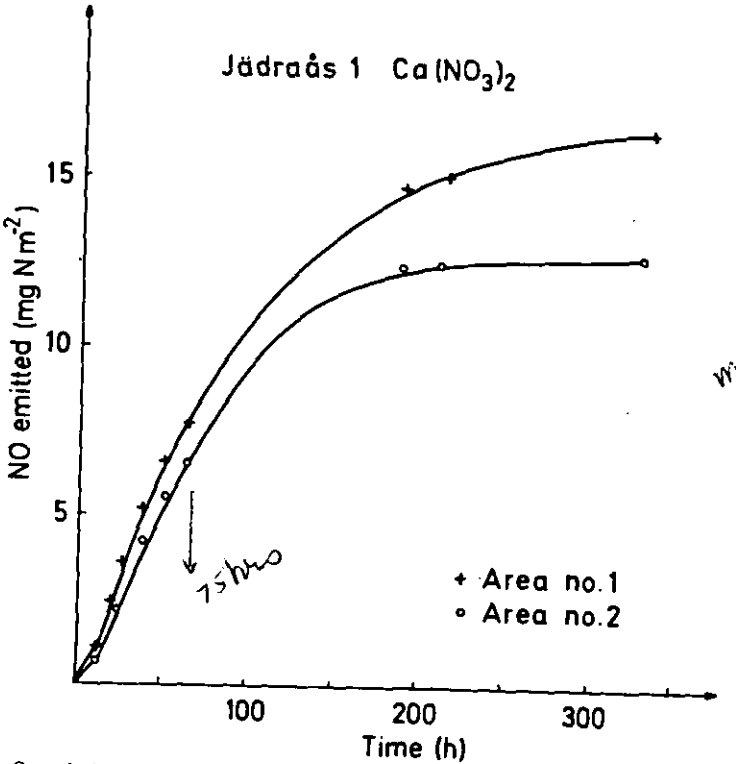


Fig. 5. Cumulative NO flux from the calcium nitrate fertilized plots at Jädraås.

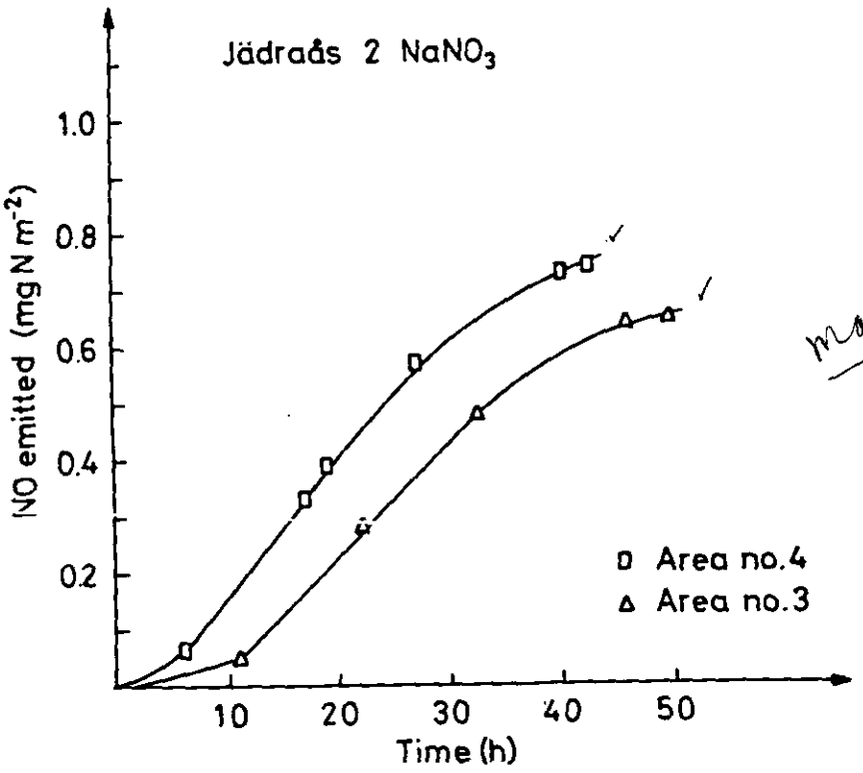


Fig. 6. Cumulative NO flux from the sodium nitrate fertilized plots at Jädraås.

duplicate plots were within 37% of the average value. The resultant losses of nitrogen as NO from the $Ca(NO_3)_2$ fertilized plots at Sörentorp were smaller by a factor of 6 than the losses from the $Ca(NO_3)_2$ fertilized plots at Jädraås (Table VI).

In the second fertilization the NO flux from the fertilized plots was down in the same range as the flux from the unfertilized plots after about 14 days (Figure 2). The subsequent soil analyses showed very low $NO_3^-$ concentrations, indicating that the decrease in NO flux was the result of exhaustion of the available nitrate (Table V). In this experiment 0.35% of the applied $NO_3^-$ was lost as NO (Table VI). This can be compared with the estimate of the corresponding value for an agricultural soil, which was 0.2% (Johansson and Granat, 1984), and with the observations on $N_2O$ losses, which are between 0.01 and 2% (Conrad *et al.*, 1983), suggesting that the fluxes of NO and $N_2O$ are about equally important for losses of nitrogen from soils.

Equilibrium concentrations, i.e., the concentration at which the uptake of NO balances production, were in the range 0.2 to 2 ppbv on the unfertilized plots. When the concentration in the chamber was increased above the equilibrium concentration, NO was found to be absorbed on the soil and vegetation. Only on four occasions at Sörentorp did the ambient NO level exceed the equilibrium concentration so that a net uptake of NO was observed. The average ambient concentrations observed during the measurements at

Table VI. Cumulative NO fluxes from the fertilized plots. On the $Ca(NO_3)_2$ fertilized plots 4.64 g $NO_3^-$–N was applied; on the $NaNO_3$ fertilized plots 1.12 g $NO_3^-$–N was applied

| Site | Fertilizer | Plot No. | NO emitted (mg N $m^{-2}$)[a] | | |
|---|---|---|---|---|---|
| | | | 45 hours | 75 hours | 340 hours |
| Sörentorp | $Ca(NO_3)_2$ | 1 | 0.92 (0.020) | 1.4 (0.031) | – |
| | $Ca(NO_3)_2$ | 2 | 0.63 (0.014) | 1.0 (0.022) | – |
| Jädraås 1 | $Ca(NO_3)_2$ | 1 | 6.9 (0.15) | 8.3 (0.18) | 16.4 (0.35) |
| | $Ca(NO_3)_2$ | 2 | 4.9 (0.11) | 7.1 (0.15) | 12.7 (0.27) |
| Jädraås 2 | $NaNO_3$ | 3 | 0.74 (0.066) | – | – |
| | $NaNO_3$ | 4 | 0.65 (0.058) | – | – |

[a] The figures in parentheses indicate the fertilizer-induced emission of NO in percent of $NO_3^-$–N applied.

Sörentorp and Jädraås were 0.48 ± 0.38 ppbv (87 measurements) and 0.10 ± 0.10 ppbv (82 measurements), respectively. The average concentrations at Sörentorp are influenced by the city of Stockholm, whereas those observed at Jädraås are more representative of NO concentrations at a rural site in Sweden. At Jädraås there was always a net emission of NO. The equilibrium concentration was observed to be dependent on the emission rate. On the fertilized plots, where the emission was much higher, equilibrium concentrations up to 170 ppbv was observed. These results are similar to those observed earlier for arable land (*Johansson and Granat*, 1984).

## 4. Speculation on Global NO Fluxes

Söderlund and Svensson (1976) estimated the average global flux of $NO_x$ from soils from a mass balance calculation. The value obtained as a residual source was 21–89 Tg N $a^{-1}$. (a = annum). Galbally and Roy (1978) estimated the global flux of NO based on their measurements on pastures in Austria to be of the order of 10Tg N $a^{-1}$. Lipschulz *et al.* (1981) measured production of NO and $N_2O$ by pure cultures of nitrifying bacteria. A ratio of about 1:5 between NO and $N_2O$ production was observed. Based on this value and an estimate of the global source of $N_2O$,they arrived at a global flux for NO of 15 N $a^{-1}$. These three estimates have been used in papers dealing with the global nitrogen budget (Söderlund, 1980; Logan *et al.*, 1981; Ehhalt and Drummond, 1982; Sanhueza, 1982; Söderlund and Rosswall, 1982; *Crutzen*, 1983).

With more direct field measurements available, it is of interest to attempt a new estimate of the global flux of NO. Table VII summarizes measurements made on three different ecosystems together with estimated values for the remaining land area.

For cultivated land the emissions reported in Johansson and Granat (1984) on unfertilized agricultural land were used. The measurements on pastures in Australia by Galbally and Roy (1978) were taken as representative for temperate grasslands. For

Table VII. Ecosystem type and respective estimated fluxes of NO. The respective areas are taken from Bolin *et al.* (1979)

| Ecosystem type | Based on | Area ($10^{-12}$ m$^2$) | Length of season (days) | Yearly flux range median (mg N m$^{-2}$) | | Global flux range (Tg N a$^{-1}$) | median |
|---|---|---|---|---|---|---|---|
| Cultivated land | Johansson and Granat, 1984 | 16 | 200 | 2–86 | 20 | 0.03–1.4 | 0.3 |
| Temperate grass-lands | Galbally and Roy, 1978 | 2.4 | 200 | 28–60 | 43 | 0.07–0.14 | 0.10 |
| Temperate forests, mixed forests, taigas | This paper | 26 | 200 | 2–14 | 5 | 0.05–0.36 | 0.13 |
| Tropical seasonal and rain forests | Estimated | 16 | 300 | 1–50 | 15 | 0.01–0.80 | 0.24 |
| Savannas and tundras | Estimated | 26 | 200 | 1–20 | 5 | 0.03–0.52 | 0.13 |
| Total | | 86 | | | | 0.2–3.2 | 0.9 |

temperate forests, mixed forests and taiga the emissions on the unfertilized forest soils obtained in this paper were used. To the knowledge of the author, there are no measurements of NO production in tropical soils, deserts, tundras or savannas. Data on nitrification and denitrification are also very scanty. In tropical soils, where nitrification occurs at high temperatures and where pH is generally fairly low, chemical decomposition of $NO_2^-$ formed by nitrification might be an important pathway for NO loss (Laudelout, *et al.*, 1977). Likewise for savannas, which consist of aerated, well drained acid soils, self-decomposition of $NO_2^-$ and subsequent emission of NO has been suggested as a process for loss of nitrogen (Pereira, 1982). The NO emission rates from these areas are assumed to be lower than the emission from cultivated land. The release of NO from deserts was assumed to be zero. All the NO fluxes given in Table VII are of course very uncertain since they are based on only three sets of measurements which have been extrapolated to very large areas.

The global flux obtained here, 0.2 to 3 Tg N a$^{-1}$ and a median value of ~1 Tg a$^{-1}$, is appreciably lower than earlier estimates. The median value is less than 10% of the estimates reported earlier and about 5% of the anthropogenic emission by combustion of fossil fuels, which is around 20 Tg N a$^{-1}$ (Söderlund and Svensson, 1976). According to Table VII the global flux from cultivated land represents about 30% of the total release of NO from soils. Given the annual production of nitrogen fertilizers, 55 Tg N (FAO, 1979), and the maximal estimated loss of fertilizer nitrogen (0.35%) obtained in this paper, the resultant contribution of 0.19 Tg N a$^{-1}$ to the global natural NO flux is about 20% of the median global flux. With increased demands for land used in agriculture and global use of artificial nitrogen fertilizers this source of $NO_x$ in the atmosphere is expected to increase.

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