Pergamon

Soil Biol. Biochem. Vol. 28, No. 4/5, pp. 597–607, 1996

PII: S0038-0717(95)00186-7
0038-0717/96 $15.00 + 0.00

# NITROUS OXIDE EMISSIONS FROM SOILS DUE TO INPUTS OF NITROGEN FROM EXCRETA RETURN BY LIVESTOCK ON GRAZED GRASSLAND IN THE U.K.

A. G. ALLEN,[1] S. C. JARVIS* and D. M. HEADON
Istitute of Grassland and Environmental Research, North Wyke, Okehampton, Devon, EX20 2SB, U.K.

*(Accepted 15 November 1995)*

**Summary**—Nitrous oxide ($N_2O$) emissions from different soils under grass were measured after treatment with cow dung and urine in field trials conducted during two separate seasons and in laboratory incubation experiments. $N_2O$ emission rates were much higher during autumn–winter than during spring–summer, and in the case of well-drained soil were substantial for both excreta types (207 mg $N_2O$-N $kg^{-1}$ of deposited dung and 197 mg $N_2O$-N $kg^{-1}$ of urine in autumn–winter). The corresponding data for poorly-drained soil were 0.2 mg (dung) and 148 mg (urine). Emissions continued over much longer periods ( ~ 60 days) from sandy and stony loams than from a silty clay loam ( ~ 30 days) under both field and laboratory conditions, and were not solely dependent on soil $NO_3^-$ or $NH_4^+$ status but also related to other factors including soil moisture, rate of plant growth and carbon availability. Results suggest that $N_2O$ production occurred during both nitrification and denitrification processes. Emission rates of up to ~1590 μg $N_2O$-N $h^{-1}$ $m^{-2}$ occurred in the field, while small rates of deposition to the soil were occasionally observed. Under laboratory conditions, similar treatments produced large emissions from loam soils having pH of 4.5–6.5 and zero emissions from a peat soil with pH of 3.8. The ratio of nitrogen released as $N_2O$ to the amount of N excreted by the livestock varied from ~ 0% (summer) to 0.8–2.3% (winter), consistent with loss rates observed for mineral fertilizers. Copyright © 1996 Elsevier Science Ltd

## INTRODUCTION

Nitrous oxide ($N_2O$) is a radiatively active atmospheric trace gas, currently accounting for 2–4% of total Greenhouse Warming Potential (GWP) (Watson *et al.*, 1992). It can also deplete stratospheric ozone following photolytic oxidation to nitric oxide (NO) (Crutzen, 1976). A current concentration of 310 nl $l^{-1}$ is increasing at a rate of 0.3% $y^{-1}$ due to an imbalance (of 30%) between sources (largely from biological processes in natural systems, both terrestrial and oceanic) and sinks (stratosphere and soil), coupled with a long atmospheric lifetime of 150 y (Khalil and Rasmussen, 1992).

On a global scale, grasslands probably contribute 10% to the total flux of $N_2O$ to the atmosphere, although uncertainty exists in estimates of the relative contributions of natural and anthropogenically managed systems (Bouwman *et al.*, 1993). In the U.K. agricultural livestock production results in large inputs of fertilizer and returns of excretal N to grassland soils (Jarvis, 1993), with subsequent loss of N to the atmosphere (as $N_2$, $N_2O$, NO and $NH_3$) as well as to ground and surface waters (mainly as $NO_3^-$).

$N_2O$ is produced as a result of microbial processes occurring in soils, especially by denitrification, i.e. the sequential reduction of $NO_3^-$ and $NO_2^-$ to NO, $N_2O$ and $N_2$ (Firestone *et al.*, 1980; Firestone and Davidson, 1989), but also through nitrification, i.e. the oxidation of $NH_4^+$ to $NO_2^-$ and $NO_3^-$ (Bremner and Blackmer, 1978; Lipschultz *et al.*, 1981) Ryden (1981) suggested that denitrification may be the dominant mechanism influencing $N_2O$ production in fertilized grassland under oxygen-limited or anaerobic conditions. However, this process is often dependent on an initial nitrification step and is inhibited under aerobic conditions. In many studies a close relationship has been observed between $N_2O$ flux and soil moisture content (Mosier *et al.*, 1981), although this may not necessarily reflect an increase in denitrification alone. Nitrification may become the major mechanism responsible for $N_2O$ production under moderately aerobic conditions (Bremner and Blackmer, 1981; Klemedtsson *et al.*, 1988).

Animals return large quantities of mineral N in their excreta which have profound effects on soil N transformation and loss processes. Whilst there is a growing database for denitrification under grazing management, there is only limited information on the specific influence of animal excreta on $N_2O$ production (e.g. Parton *et al.*, 1988; Colbourn, 1992; Spatz *et al.*, 1992; Monaghan and Barraclough,

*Author for correspondence.
[1]Current address: Institute of Public and Environmental Health, University of Birmingham, Edgbaston, Birmingham B15 2TT, U.K.

1993). Estimates of $N_2O$ emissions derived from various components of animal production are still uncertain. Here we present the results of measurements of $N_2O$ fluxes which occurred after application of cattle dung and urine to different soils in the field and the laboratory, and attempt to identify the major controls and mechanisms responsible for $N_2O$ production under these conditions.

## MATERIALS AND METHODS

### *Site description and soil types*

Field experiments were conducted on grassland at the Institute of Grassland and Environmental Research, North Wyke Research Station farm in Devon. This southwest region of the U.K. has a cool maritime climate with predominantly westerly airflows. A mean annual rainfall of 1050 mm is fairly evenly distributed throughout the year, with wettest months being November–January. Annual mean air temperature is 9.5°C, and coldest and warmest months are February (4.3°C) and July (15.3°C), respectively. Mean annual sunshine is 1430 h, with an average of 1.6 h $d^{-1}$ in December–January and 6.4 h $d^{-1}$ in June. Soil temperature and rainfall data for the periods during which field trials were undertaken (i.e. 23 November 1993–9 February 1994 and 18 April 94–14 June 1994) are illustrated in Fig. 1. The autumn–winter period was characterized by cool, very wet conditions, and the spring–summer period by steadily increasing soil temperature and

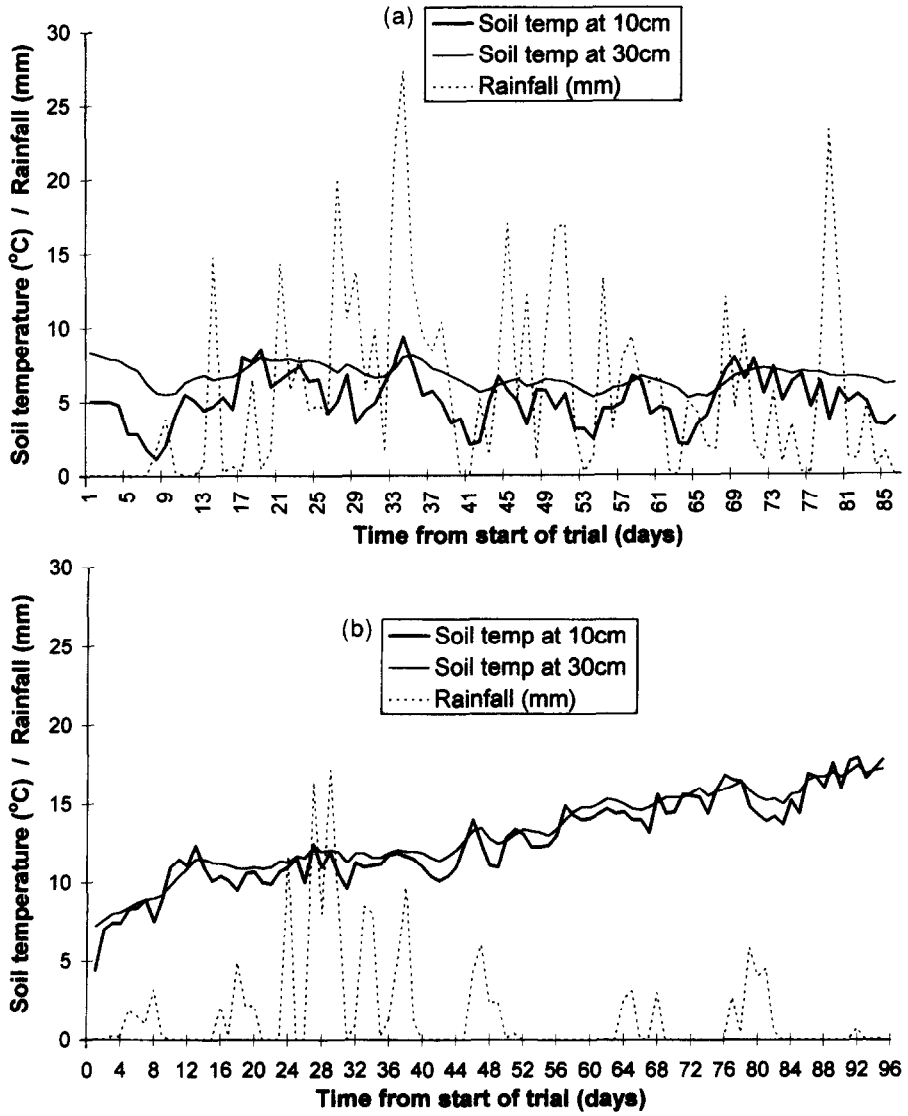


Fig. 1. Soil temperature and rainfall data for experimental sites at North Wyke during (a) November 1993–February 1994 and (b) April–July 1994.

limited rainfall (which mostly occurred between days 20–40 of the trial).

Five different soil types were used; the field measurements were based on soils A and B, all soils were used for the laboratory studies.

A. Poorly-drained silty clay loam (Halstow series), pH 4.9. Perennial ryegrass plus other grasses and white clover sward.

B. Moderately well-drained stony loam (Crediton series), pH 5.8. Perennial ryegrass plus white clover sward.

C. Well-drained sandy loam (Cuckney series), pH 6.5. Perennial ryegrass sward.

D. Raw oligomorphous peat, perennially waterlogged (Crowdy series), pH 3.8. Mixed heather–grass sward.

E. Slowly permeable clay-loam under deciduous woodland, (Hallsworth series), pH 4.5. Ground cover of leaf litter and bark plus limited grass.

*Field experiments*

Samples of excreta (1.2 kg dung or 200 ml urine) collected directly from dairy cows fed on grass silage diets were each applied to 20 cm dia areas in field plots located on soils A and B. These application rates were representative of typical animal excreta deposition rates, on a mass per area basis, and allowed the entire area of soil affected by the excreta, i.e. the area covered plus that into which excretal derived material diffuses ( $\sim 2 \times$ the area of excreta, according to Lantinga *et al.*, 1987) to be contained within the circumference of the cover chambers used for $N_2O$ measurements. At both sites each treatment was applied to four separate plots, at a spacing of 3 m, on areas which had not been grazed or had fertilizers applied for at least 12 months prior to the start of the experiments: herbage had been cut and removed during the course of the previous year. Dung and urine applications were made to the soils in November 1993 and April 1994, and fertilizer ($NH_4NO_3$ at a rate equivalent to 120 kg N $ha^{-1}$) was applied both alone and in combination with dung and urine in April 1994. Measurements of $N_2O$ fluxes and soil mineral N (as $NO_3^-$ and $NH_4^+$) were then continued for up to 4 subsequent months after application.

*Laboratory incubations*

Incubation experiments were designed to investigate the influence of variables such as soil temperature, soil type and excretal type on $N_2O$ generation. Dung and urine treatments were applied to soil blocks (17 × 16 × 10 cm) removed from the field sites, placed into polypropylene containers, and incubated after treatment with measurements of $N_2O$ and soil $NO_3^-$ and $NH_4^+$ for up to several months. Evaporation from the soil blocks was restricted by partially covering the containers with plastic lids and no further water was applied. Two separate experiments were conducted, in which the amounts of dung and urine added to the soil area covered were proportional to those used in the field studies:

Expt 1: (1). Duplicate blocks for each soil–temperature–treatment combination for soils A and B were collected from the field on 2 March 1994, allowed to stabilize for 2 days and then treated with dung (220 g) and urine (37 ml) from cows on either silage–concentrate (Type 1) or silage-only (Type 2) diets, and then incubated at either 5 or 16°C. There were also duplicate blank soils at each temperature.

Expt 2: (2). Triplicate blocks of soils A–E were collected from the field on 13 May 1995, allowed to stabilize for 3 days and then treated with 37 ml of urine from cows on silage–barley diets and incubated at 16°C. Blanks without added urine were run in duplicate.

*Measurement techniques*

In the field, $N_2O$ flux was measured using a static chamber technique (Mosier, 1989). Galvanized steel cylinders (40 cm dia × 30 cm height) were driven into the soil to a depth of 5 cm around each individual excretal patch, and covered with a Perspex lid (fitted with a 3-way valve) sealed onto the top of the chambers during short (30–40 min) periods of measurement (one per sampling day, usually obtained between 10.00 and 13.00 h). Gas samples were transferred to 20 ml headspace sampler vials fitted with PTFE-silicone septa using 60 ml polypropylene syringes. $N_2O$ contents of the samples were determined within 12 h.

In the laboratory experiments, $N_2O$ concentrations in the container headspace were measured after sealing the containers with tightly fitting lids for $\sim 20$ min. The containers had been previously checked for possible leakage by filling them with known concentrations of $N_2O$ and analysing the contents after periods of $< 3$ h storage; no loss of $N_2O$ was observed. The stability of $N_2O$ in the headspace vials was also investigated; losses of $< 10\%$ were observed after 24 h and $< 50\%$ after 6 days, necessitating analysis within at least 6 h after collection.

$N_2O$ analysis was by an automated ATI–Unicam 610 g.c. equipped with an e.c.d. and a column backflush system. The columns (2.0 m × 1/8″) contained HayeSep Q and HayeSep N (80–100 mesh). Nitrogen was used as carrier (40 ml $min^{-1}$) and $CO_2$ as a detector quenching agent to enhance sensitivity (0.5 ml $min^{-1}$). Detector and oven temperatures were maintained at 300 and 80°C, respectively. Gas samples were loaded into the system using a Hewlett–Packard type 19395A Headspace Analyser and the output signal measured on a strip chart recorder. The detection limit of the technique ($3\sigma$) was 12 nl $N_2O$ $l^{-1}$ with a relative standard deviation of 1.5% for measurements at ambient background concentrations.

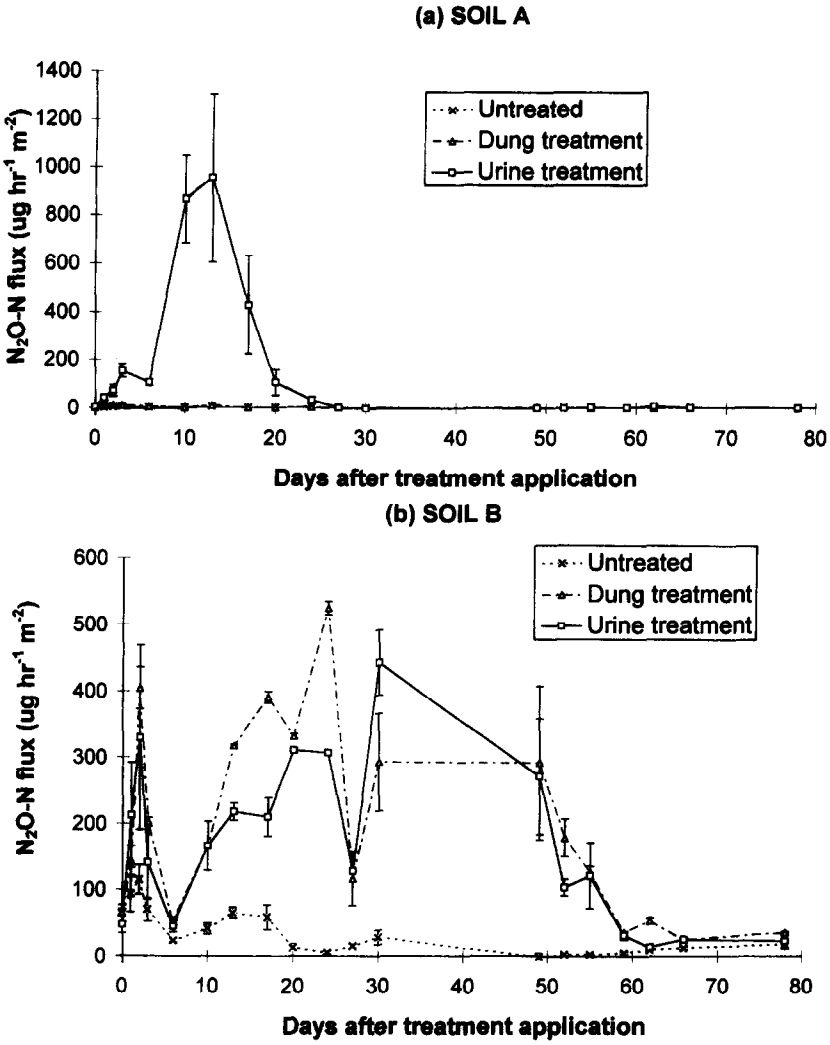


Fig. 2. $N_2O$ fluxes from field soils treated with either dung or urine, or left untreated, during autumn–winter. A — poorly-drained clay; B — moderately-drained loam. Day 0 = 23 November 1993. Vertical bars are SE for means.

Samples of soil (25 mm dia cores) were periodically removed from three separate horizons in the field experiment (i.e. 0–5 cm, 5–10 cm, 10–20 cm). The separate horizon fractions from at least four cores removed from each treatment were combined to reduce the potential influence of spatial heterogeneity. In the laboratory incubation studies, soil was sampled by removing representative material to the depth of the whole block. All soils were extracted immediately after collection with 1M KCl (ratio 1:2 moist soil:KCl). Soil moisture content was determined gravimetrically after drying representative samples of fresh soil at 105°C overnight. After filtering through clean Whatman no. 41 filters, $NO_3^-$ and $NH_4^+$ in the filtrate were analysed using a segmented flow technique, on a Skalar autoanalyser, with colorimetric detection (Hendriksen and Selmer Olsen, 1970; Verdouw *et al.*, 1977). Soil extracts were frozen and stored for up to several weeks whenever immediate analysis was impracticable.

Linear regression was used to identify the relationships between soil variables and $N_2O$ emissions. Student's t-test was used to identify differences between the two temperature regimes used in the laboratory experiment.

## RESULTS AND DISCUSSION

### *Field experiments*

*Autumn–winter effects.* During autumn–winter the two soils showed large differences in response after excreta application (Fig. 2). The total $N_2O$ flux (determined from the area under the curves) from the dung treatment was much higher for soil B than for soil A (Table 1), while urine treatment produced similar overall fluxes from both soils even though the

Table 1. Total nitrous oxide flux, mg $N_2O$-N (RSD[1]) due to application of 1 kg dung or urine to soils A and B under field conditions

| Treatment | Soil A Poorly drained | | Soil B Well drained | |
|---|---|---|---|---|
| Autumn/winter (applied in November) | | | | |
| | mean | RSD | mean | RSD |
| Dung[2] | 0.2 | 82 | 207 | 20 |
| Urine[2] | 148 | 65 | 197 | 38 |
| Spring/summer (applied in April) | | | | |
| | mean | RSD | mean | RSD |
| Dung[2] | 7.1 | 80 | 12.4 | 42 |
| Urine[2] | −17.0 | 85 | −4.3 | 61 |
| Dung + fertilizer[3] | 5.4 | 9 | 14.8 | 4 |
| Urine + fertilizer[3] | −3.5 | 9 | −40.6 | 52 |

[1] RSD: relative standard deviations, as % (h = 4)[2].
[2] Determined after subtracting the flux measured from untreated plots.
[3] Determined after subtracting the flux measured from fertilized-only plots.

duration of the emission periods was much longer for soil B. Peak rates of emission, on the other hand, were highest for the poorly-draining soil (A). These effects were probably due, at least in part, to different soil moisture regimes in the two soils. Soil moisture content varied between 30–50% (soil A) and 18–25% (soil B) during the course of the experiment, decreasing with depth in both cases. Soil A frequently reached or approached field capacity. On occasions, there was a small uptake of $N_2O$ by soil A (i.e. a reduction in headspace $N_2O$ concentration after the container was closed) which may have been related to greatly reduced rates of diffusion through the soil and enhanced rates of reduction to $N_2$ (Ryden, 1981; Keller *et al.*, 1986). It had been found that although denitrification may be enhanced at low $O_2$ concentrations, $N_2O$ production may be reduced as anaerobic conditions develop and the ratio of $N_2$ to $N_2O$ increases (Firestone and Davidson, 1989). $N_2$ may be the only gaseous product of denitrification when the ability of the soil to reduce $N_2O$ is greater than the rate of $N_2O$ production.

Differences in the $N_2O$ emissions observed for soil A cannot be explained by between-treatment variation in soil $NO_3^-$-N, as similar low concentrations (e.g. 0.5–1.0 $\mu$g $NO_3^-$-N $g^{-1}$ dry soil) and temporal trends occurred in all cases, including untreated plots. The peak after 12 days was therefore apparently due to environmental factors rather than a treatment-specific effect. Conversely, $NH_4^+$-N increased up to 70 days in untreated (from 5 $\mu$g $NH_4^+$-N $g^{-1}$ dry soil to 12 $\mu$g $NH_4^+$-N $g^{-1}$) and dung-treated (from 6 $\mu$g $NH_4^+$-N $g^{-1}$ dry soil to 18 $\mu$g $NH_4^+$-N $g^{-1}$) areas, while a very high initial concentration (140 $\mu$g $NH_4^+$-N $g^{-1}$) in urine-treated soil declined to background amounts after 70 days. Neither $NO_3^-$-N nor $NH_4^+$-N concentration maxima occurred at times of highest rates of $N_2O$ evolution, although statistically significant relationships occurred between $N_2O$ and $NO_3^-$-N, and between $N_2O$ and $NH_4^+$-N (Table 2). $NH_4^+$ contents provided the better correlations with soil A but not with soil B under field conditions. Although this indicates a strong involvement of nitrification the mechanisms, either direct or indirect, are not clear. Better correlations of $N_2O$ with $NH_4^+$ than $NO_3^-$ have also been found when animal slurry was added to a peat soil (Jarvis *et al.*, 1994).

Higher concentrations of $NO_3^-$-N were always found in soil B than soil A, although between-treatment differences were small, with a background concentration of 0–1 $\mu$g $NO_3^-$-N $g^{-1}$ dry soil and peak amounts (after 21 days) of 2–5.5 $\mu$g $NO_3^-$-N $g^{-1}$ (highest for urine treatments). Significant correlation

Table 2. Values for the correlation coefficient (r) obtained between $N_2O$ emission rates and soil $NO_3^-$ and $NH_4^+$ concentrations, in (a) autumn/winter, (b) spring/summer and (c) field and laboratory experiments

| | SOIL A | | SOIL B | |
|---|---|---|---|---|
| Horizon (cm) | $NO_3^-$ | $NH_4^+$ | $NO_3^-$ | $NH_4^+$ |
| (a) | | | | |
| 0–5 | 0.64*** | 0.58*** | 0.43** | 0.06 |
| 5–10 | 0.21 | 0.75*** | 0.49** | 0.21 |
| 10–20 | 0.27 | 0.66*** | 0.24 | 0.38* |
| All horizons (0–20) | 0.40* | 0.65*** | 0.43** | 0.15 |
| (b) | | | | |
| 0–5 | 0.16 | 0.00 | −0.33 | 0.19 |
| 5–10 | 0.02 | 0.07 | −0.29 | −0.29 |
| 10–20 | −0.24 | −0.03 | −0.18 | 0.06 |
| All horizons (0–20) | 0.06 | 0.07 | −0.29 | −0.10 |
| (c) | | $NO_3^-$ | | $NH_4^+$ |
| Soil A (5°C) | | 0.21 | | 0.01 |
| Soil A (16°C) | | −0.11 | | −0.26 |
| Soil B (5°C) | | 0.49*** | | −0.20 |
| Soil B (16°C) | | 0.19 | | 0.26 |

Significant at: *P < 0.1, **P < 0.05, ***P < 0.01.

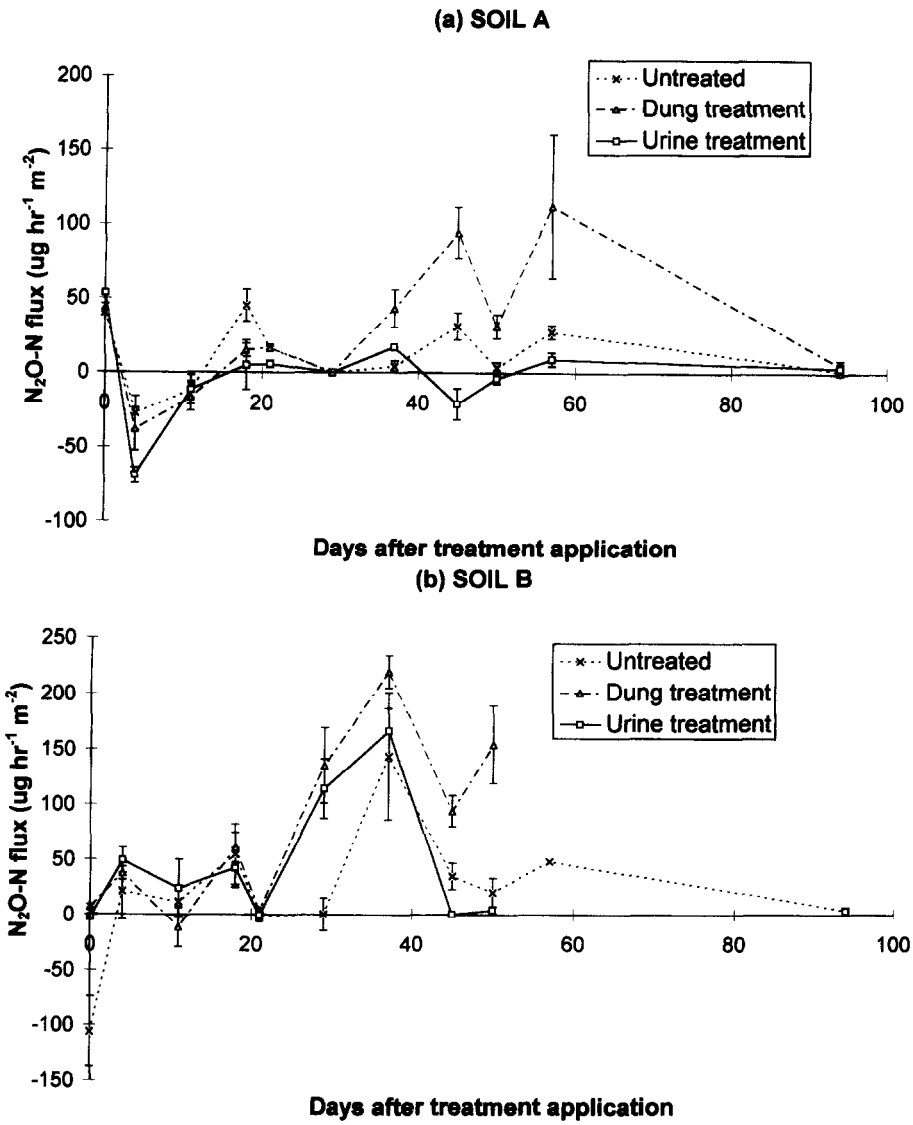


Fig. 3. $N_2O$ fluxes from field soils treated with either dung or urine, or left untreated, during spring–summer. A — poorly drained clay; B — moderately drained loam. Day 0 = 18 April 1994. Vertical bars are SE for means.

was obtained between $N_2O$ and soil $NO_3^-$-N (Table 2). $NH_4^+$-N was present at concentrations above background in both dung and urine-treated soils throughout the trial, with $NH_4^+$-N derived from the treatments remaining in the upper soil horizons despite heavy rainfall. Differences in rates of oxidation of $NH_4^+$ between the two soils may explain the between-soil differences in $N_2O$ production in dung-treated plots. It remains unclear, however, why such differences were restricted to added dung and were not observed with urine (other variables such as soil moisture content, temperature and grass cover were the same for both treatments), although the presence of dung may have impeded $O_2$ transfer into the soil.

*Spring–summer effects.* $N_2O$ fluxes were much lower in spring–summer than in autumn–winter for all treatments (Fig. 3). Highest emissions were measured over the dung plots, perhaps due to more anaerobic conditions, different trends in mineralization–immobilization or a lower rate of N uptake by the growing grass, and consequent increased N available for soil microbial processes. Enhanced soil $NO_3^-$-N only occurred with fertilizer + dung treatments although $NO_3^-$ was always present after 40 days. $N_2O$ emissions from soil B dung and urine-treated plots increased sharply at 30–40 days post-treatment, in line with rainfall events which occurred between days 23 and 37; untreated control plots also showed an increase in emissions during this period. Soil A, which retained a higher soil moisture content (26–30%) than soil B (15–25%) during the entire experiment, did not show any equivalent response. $NH_4^+$-N was always present in both of the

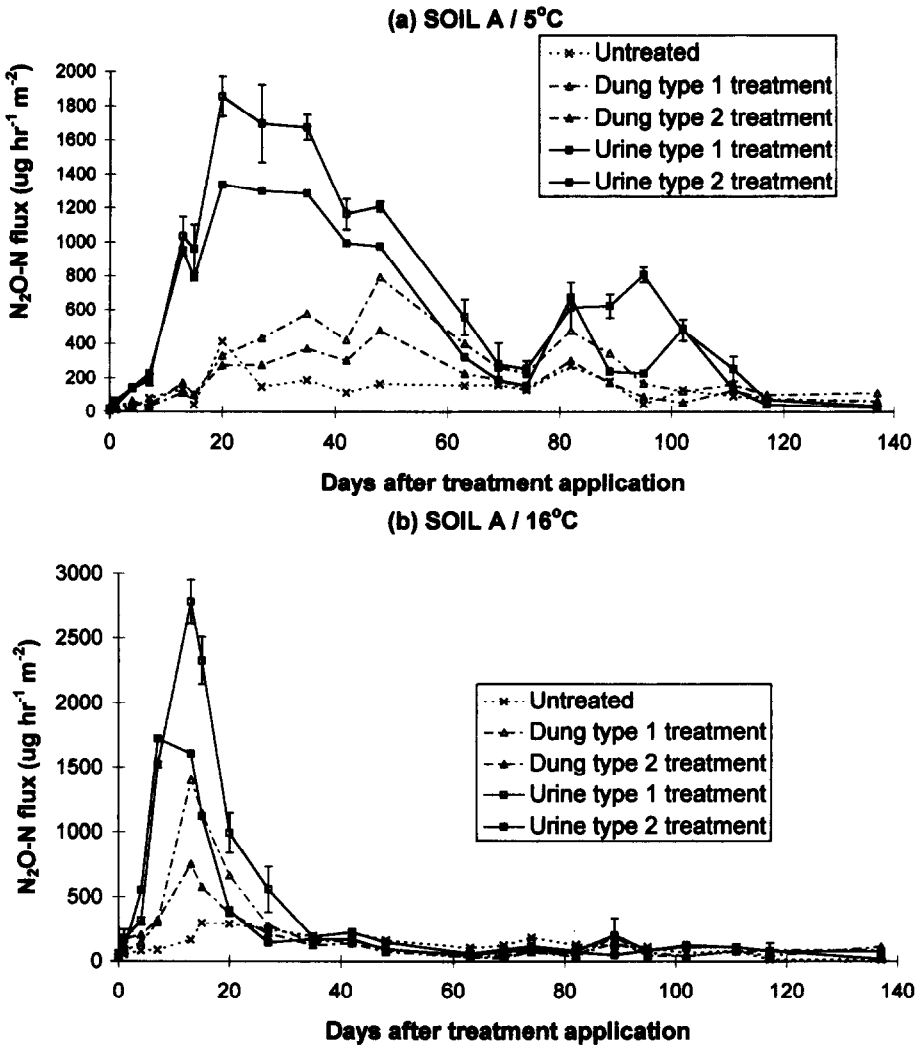


Fig. 4. Emissions of $N_2O$ obtained after treatment with dung and urine in laboratory experiments, as influenced by soil type and incubation temperature and excretal source (type 1 = milking cows fed on silage + concentrates; type 2 = barren cows fed on silage only). Vertical bars are SE for means.

soils and in all treatments, existing mainly in the upper soil horizons, probably due to continued mineralization. Low rates of $N_2O$ generation occurred at low soil moisture contents (i.e. no denitrification) rather than to mineral N deficiency, although active cycling of N at the root–soil interface during rapid grass growth may have reduced $NO_3^-$ availability for microbial processes likely to generate $N_2O$.

Although positive fluxes were generally measured over both treated and untreated areas, the net effect of urine applications to both soils in the summer was to reduce $N_2O$ emissions relative to untreated areas, perhaps due to changes in the ratio of $N_2O$-to-$N_2$ evolved or other direct effects on soil microbiota physiology and composition. Similarly, lower emissions were measured from fertilizer + urine treatments than from fertilizer-only treatments (Table 1).

Table 3. Total nitrous oxide flux, mg $N_2O$-N (RSD[1]) arising from application of 1 kg dung or urine to soils under laboratory conditions (Excretal type 1 from milking cows fed on silage + concentrates; excretal type 2 from barren cows fed on silage only)

| | Soil A | | | | Soil B | | | |
|---|---|---|---|---|---|---|---|---|
| Treatment | 5°C | RSD | 16°C | RSD | 5°C | RSD | 16°C | RSD |
| Dung 1 | 64 | 38 | 32 | 7 | 202 | 68 | 116 | 41 |
| Dung 2 | 26 | 23 | 3 | 11 | 72 | 42 | 58 | 12 |
| Urine 1 | 1496 | 33 | 550 | 4 | 1015 | 33 | 757 | 5 |
| Urine 2 | 1015 | 54 | 258 | 24 | 843 | 24 | 1049 | 16 |

[1] RSD: relative standard deviations, as %.

The effect of fertilizer alone was to approximately double the background emission rate to 2.2 and 9.2 g $N_2O$-N $kg^{-1}$ of applied N for soils A and B, respectively; correlations with soil mineral N were poor (Table 2).

*Laboratory incubation experiments*

$N_2O$ emissions from soils A and B during Experiment (1) were greater than those in the field for all treatments (Fig. 4). Total emissions were higher from soils at the lower temperature (5°C) for all soil-treatment combinations except soil B + urine 2 (Table 3), although the differences were not statistically significant. The effect of temperature is unlikely to be reflected in overall rates of denitrification, as in temperate soils this process is generally reduced at low temperatures (Jacobson and Alexander, 1980). However, the influence of temperature on the ratio of gaseous products was not determined in our work. Urine always produced higher fluxes than dung, and animal diet had a clear influence on emissions which, with the single exception of soil B + urine 2, were always highest where concentrates had been included in the feed and thus likely to have increased N excretion (no analysis was made of excretal material). Emission rates from soils at 16°C peaked after 10 days with soil A and 15 days with soil B, and the flushes continued for 30 and 60 days, respectively. At 5°C, emissions continued for 120 days in both cases.

As occurred during the winter field measurements, the greatest differential between emissions resulting from either dung or urine occurred with soil A. However, large differences were also observed for soil B, even though treatments were applied at the same proportional rate as in field trials and dung had been similarly incorporated into the soil in the incubation box by the action of earthworms and other macrofauna by the end of the experiment.

Although the cow urine used in the incubation experiments would have contained twice as much total N as dung (on a fresh weight basis) (Van Vuuren and Meijs, 1987), the different quantities used (i.e. ratio of dung-to-urine, by weight, of 6) would have resulted in approximately 3 times more N being applied with dung than with urine. The greater fluxes of $N_2O$ from urine treatments therefore indicate that the immediate losses are related to the mobile forms of N, rather than the total amount of N. The various urines used contained 0–6 mg $NO_3^-$-N $l^{-1}$ and 110–160 mg $NH_4^+$-N $l^{-1}$, and the fresh dungs 2 mg $NO_3^-$-N $l^{-1}$ and 16–19 mg of $NH_4^+$-N $l^{-1}$. The N in urine is largely present in the form of relatively easily-metabolized compounds such as urea and hippuric acid which can be readily converted to $NH_4^+$ which can be oxidized (nitrification) and then rapidly denitrified under appropriate conditions. Soil $NO_3^-$-N concentrations generally increased towards the end of the incubation, usually after peak $N_2O$ emission rates had been reached, with significant amounts ( $< 250$ $\mu g$ $NO_3^-$-N $g^{-1}$ dry soil) remaining in the soil when gaseous emissions were at very low rates. Correlation analysis showed that in only one of the soil type–incubation temperature subsets (soil B at 5°C) was there a significant relationship between $N_2O$ emissions and corresponding soil $NO_3^-$-N concentrations (Table 2). No significant correlations were obtained between $N_2O$ and $NH_4^+$-N. These data suggest that, under our conditions, $NO_3^-$ was not limiting for $N_2O$ emission. $NO_3^-$ accumulation in soils is known to occur when there is C limitation to denitrifiers (Hutchinson and Davidson, 1993). The reduced fluxes may, therefore, have been due to reduced C availability (Eaton and Patriquin, 1989) or, perhaps, the cessation of nitrification although both of these seem unlikely because of the additions made in the excreta. In most cases, $NH_4^+$-N was exhausted in soils by the end of the incubation, at which time most of the mineral N was present as $NO_3^-$. Plant uptake of N was unlikely to have been significant because plant growth was inhibited by low illumination, although immobilization into microbial biomass may have provided a potentially large sink for both $NO_3^-$ and $NH_4^+$. Although major changes in soil moisture contents during the course of the experiment were avoided by partially covering the containers to prevent evaporation while permitting gaseous exchange to occur, some change in overall moisture content and distribution would have taken place and this may have had direct effects on denitrification.

In Experiment 2 the differences between soils A–E were investigated with untreated and urine-treated subsamples at 16°C. Despite all the soil samples being collected from field sites in the same geographical region on the same day, moisture contents at the time of collection were 47, 21, 19, 80 and 49% for soils A–E, respectively, reflecting differing water holding capacities and drainage. The rates and patterns of $N_2O$ production from soils A and B were similar to those obtained during Experiment 1, with emissions declining to very low rates after 40 days (Fig. 5), while soil C showed a general trend similar to soil B (as expected for well drained soils having comparable physical texture) although both peak emission rate and total $N_2O$ production were lower. Soils D and E exhibited quite different characteristics from the other soils. Soil D (Dartmoor peat) failed to produce any significant quantities of $N_2O$, in marked contrast to all other soils studied despite, in principle, possessing an abundance of anaerobic sites which should be favourable for denitrification. Unlike in the other soils, $NO_3^-$ remained at very low concentrations throughout the incubation, while $NH_4^+$ remained high, presumably due to either a complete lack of available $O_2$ in the soil, a small population of nitrifiers or high acidity. Failure to produce $N_2O$ in soil D emphasizes the importance of a coupling between nitrification and denitrification processes. Denitrification is strongly inhibited under

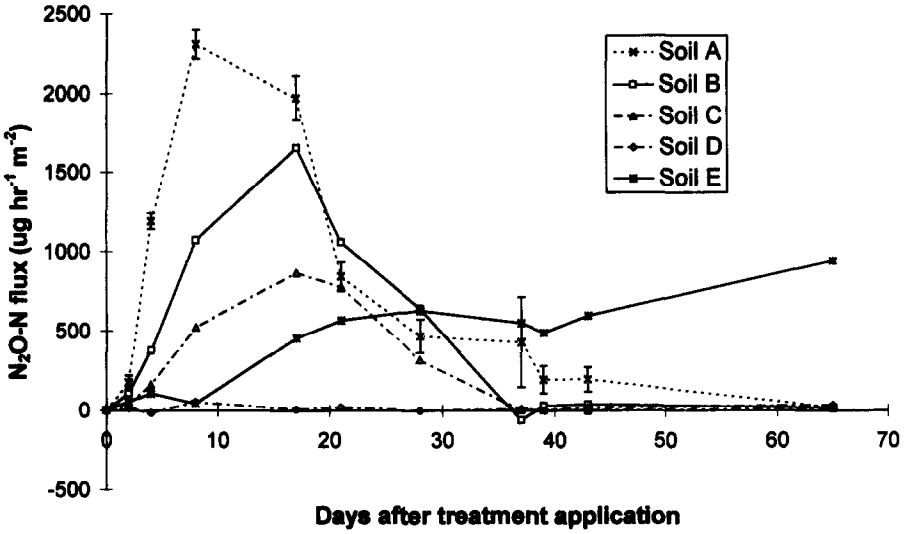


Fig. 5. Comparative emissions of $N_2O$ under laboratory conditions from five soil types treated with urine (A = silty clay loam, B = loam, C = sandy loam, D = peat, E = silty clay loam (woodland)) in lowland U.K. Vertical bars are SE for means.

continuously anaerobic conditions due to suppression of nitrification by $O_2$ non-availability (Sahrawat and Keeney, 1986), and is also reduced at low pH (Knowles, 1982). $N_2O$ production from soil E occurred over an extended period, with peak rates reached after ~ 65 days: slow rates of nitrification and nitrate production are typical of forest soils. Total emissions were therefore potentially greater for this soil than for the other soils, even though peak evolution rates were lower. It is possible that leaf litter and plant debris in this soil provided a plentiful source of metabolisable C, which in the other soils may have become limiting at an early stage. This seems unlikely since all the soils were grassland soils with much C accumulated in the soil including recently senesced sward materials. Again there is the possibility of a strong coupling of nitrification and denitrification processes in soil E.

*Estimate of $N_2O$ production following deposition of excreta by grazing domestic livestock on U.K. lowland grassland*

The total annual emission of $N_2O$ due to excretal patches on lowland grassland in the U.K. has been calculated after making several initial assumptions, these being: (i) the area of land involved is 5 Mha (Agricultural Statistics, U.K., 1989); (ii) a daily deposition per animal (cattle) on the sward of 2 kg

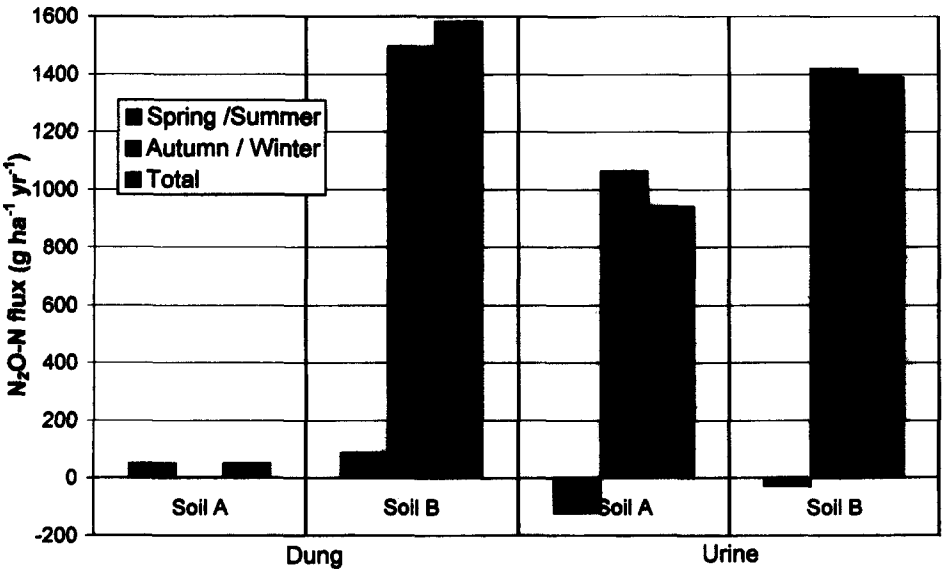


Fig. 6. Calculated annual fluxes of $N_2O$ resulting from field-deposited excreta, on the basis of a stocking density of 2 animals $ha^{-1}$ (cattle) and 180 grazing d $y^{-1}$.

each of dung and urine and each on 10 occasions (Lantinga *et al.*, 1987); (iii) a stocking rate of 2 animals $ha^{-1}$ and 180 grazing days $y^{-1}$; and (iv) a similar influence, per unit area, on $N_2O$ fluxes resulting from cattle and sheep excreta (the latter were not employed in this work).

The ratio of N released as $N_2O$ to the estimated amounts of N excreted by the grazing livestock was highly dependent on season, ranging from $< 0.1\%$ in summer (i.e. April–July in our case) to 0.8–2.3% in winter (depending on soil type). Equivalent flux rates for either dung or urine, were 53 to 1583 g $N_2O$-N $ha^{-1}$ $y^{-1}$ (Fig. 6). A mean total flux for the U.K. of 0.0025–0.0075 Tg $N_2O$-N $y^{-1}$ (equivalent to 0.50–1.50 kg $N_2O$-N $ha^{-1}$) corresponds to 0.2–0.6% of the global $N_2O$ flux from grasslands (excluding the direct contribution from mineral N fertilizers), according to current estimates (Bouwman, 1993). Bouwman (1994) estimated a global emission of 1.0 Tg $N_2O$-N $y^{-1}$ due to all forms of domestic animal excreta, assuming a similar loss as for mineral fertilizers (1% of added N). The grazing component of lowland livestock production in the U.K. then contributes 0.25–0.75% of this global total. On the basis of the present data, the actual emission is probably towards the lower end of the quoted range since much of the grazing influence of the British herd will be during spring–summer rather than during autumn–winter. However, the quantity of excreta deposited during the autumn appears to be particularly critical. Further work is required to refine the above estimates, and to take account of different effects during other times of the grazing season. The seasonally variable interactions between excretal returns and $N_2O$ efflux also need to be determined for a range of soil types.

*Acknowledgement*—The work described was commissioned by the Ministry of Agriculture, Fisheries and Food, London.

## REFERENCES

Agricultural Statistics, U.K. (1989) pp. 1–96, Ministry of Agriculture, Food and Fisheries, HMSO.

Bouwman A. F. (1993) The global source distribution of nitrous oxide. In *Proceedings of the IPCC Workshop on Methane and Nitrous Oxide* (A. R. van Amstel, Ed.), pp. 261–269. RIVM, 3720 BA Bilthoven.

Bouwman A. F. (1994) Estimated global source distribution of nitrous oxide. In *$CH_4$ and $N_2O$. Global Emissions and Controls from Rice Fields and Other Agricultural and Industrial Sources* (K. Minami, A. Mosier and R. Sass, Eds), pp. 147–159. National Institute of Agro-Environmental Sciences, Tsukuba. Yokendo Publishers, Tokyo.

Bouwman A. F., Fung I., Matthews E. and John J. (1993) Global analysis of the potential for $N_2O$ production in natural soils. *Global Biogeochemical Cycles* **7**, 557–597.

Bremner J. M. and Blackmer A. M. (1978) Nitrous oxide: emission from soils during nitrification of fertilizer nitrogen. *Science* **199**, 295–296.

Bremner J. M. and Blackmer A. M. (1981) Terrestrial nitrification as a source of atmospheric nitrous oxide. In *Denitrification, Nitrification and Atmospheric Nitrous Oxide* (C. C. Delwiche, Ed.), pp. 151–170. John Wiley, New York.

Colbourn P. (1992) Denitrification and $N_2O$ production in pasture soil. The influence of nitrogen supply and moisture. *Agriculture Ecosystems and Environment* **39**, 267–278.

Crutzen P. J. (1976) The influence of nitrogen oxides on the atmospheric ozone content. *Quarterly Journal of the Royal Meterological Society* **96**, 320–325.

Eaton L. J. and Patriquin D. G. (1989) Denitrification in lowbush blueberry soils. *Canadian Journal of Soil Science* **69**, 303–312.

Firestone M. K., Firestone R. B. and Tiedje J. M. (1980) Nitrous oxide from soil denitrification: factors controlling its biological production. *Science* **208**, 749–751.

Firestone M. K. and Davidson E. A. (1989) Microbiological basis of NO and $N_2O$ production and consumption in soil. In *Exchange of Trace Gases between Terrestrial Ecosystems and the Atmosphere* (M. O. Andreae and D. S. Schimel, Eds), pp. 7–21. John Wiley, New York.

Hendriksen A. and Selmer Olsen A. R. (1970) Automatic methods for determining nitrate and nitrite in water and soil extracts. *Analyst* **95**, 514–518.

Hutchinson G. L. and Davidson E. A. (1993) Processes for production and consumption of gaseous nitrogen oxides in soil. In *Agricultural Ecosystem Effects on Trace Gases and Global Climate Change*, pp. 79–93. American Society of Agronomy, Madison.

Jacobson S. N. and Alexander M. (1980) Nitrate loss from soil in relation to temperature, carbon source and denitrifier populations. *Soil Biology & Biochemistry* **12**, 501–505.

Jarvis S. C. (1993) Nitrogen cycling and losses from dairy farms. *Soil Use and Management* **9**, 99–104.

Jarvis S. C., Hatch D. J., Pain B. F. and Klarenbeek J. V. (1994) Denitrification and the evolution of nitrous oxide after the application of cattle slurry to a peat soil. *Plant and Soil* **166**, 231–241.

Keller M., Kaplan W. A. and Wofsy S. C. (1986) Emissions of $N_2O$, $CH_4$ and $CO_2$ from tropical forest soils. *Journal of Geophysical Research* **91**, 11 791–11 802.

Khalil M. A. K. and Rasmussen R. A. (1992) The global sources of nitrous oxide. *Journal of Geophysical Research* **97**, 14 651–14 660.

Klemedtsson L., Svensson B. H. and Rosswall T. (1988) Relationships between soil moisture content and nitrous oxide production during nitrification and denitrification. *Biology and Fertility of Soils* **6**, 106–111.

Knowles R. (1982) Denitrification. *Microbiology Review* **46**, 43–70.

Lantinga E. A., Keuning J. A., Groenwold J. and Deenen P. J. A. G. (1987) Distribution of excreted nitrogen by grazing cattle and its effects on sward quality, herbage production and utilization. In *Animal Manure on Grassland and Fodder Crops. Fertilizer or Waste?* (H. G. van der Meer, R. J. Unwin, T. A. van Dijk and G. C. Ennik, Eds), pp. 103–117. Martinus Nijhoff Publishers, Dordrecht.

Lipschultz F., Zafiriou O. C., Wofsy S. C., McElroy M. B., Valois F. W. and Watson S. W. (1981) Production of NO and $N_2O$ by soil nitrifying bacteria. *Nature* **294**, 641–643.

Monaghan R. M. and Barraclough D. (1993) Nitrous oxide and dinitrogen emissions from urine-affected soil under controlled conditions. *Plant and Soil* **151**, 127–138.

Mosier A. R., Stillwell M., Parton W. J. and Woodmansee R. G. (1981) Nitrous oxide emissions from a native shortgrass prairie. *Soil Science Society of America Journal* **45**, 617–619.

Mosier A. R. (1989) Chamber and isotope techniques. In *Exchange of Trace Gases between Terrestrial Ecosystems and the Atmosphere* (M. O. Andreae and D. S. Schimel, Eds), pp. 175–187. John Wiley, New York.

Parton W. J., Mosier A. R. and Schimel D. S. (1988) Rates and pathways of nitrous oxide production in a shortgrass steppe. *Biogeochemistry* **6**, 45–58.

Ryden J. C. (1981) $N_2O$ exchange between a grassland soil and the atmosphere. *Nature* **292**, 235–237.

Sahrawat K. L. and Keeney D. R. (1986) Nitrous oxide emission from soils. In *Advances in Soil Science*, Vol. 4 (B. A. Stewart, Ed.), pp. 103–148. Springer-Verlag, New York.

Spatz G., Neuendorff J., Pape A. and Schröder C. (1992) Nitrogen dynamics under excrement patches in pastures. (In German.) *Zeitschrift für Pflanzenernährung und Bodenkunde*. **155**, 301–305.

Van Vuuren A. M. and Meijs J. A. C. (1987) Effects of herbage composition and supplement feeding on the excretion of nitrogen in dung and urine by grazing dairy cows. In *Animal Manure on Grassland and Fodder Crops. Fertilizer or Waste*? (H. G. van der Meer, R. J. Unwin, T. A. van Dijk and G. C. Ennik, Eds), pp. 17–25. Martinus Nijhoff Publishers, Dordrecht.

Verdouw H., van Echteld C. J. A. and Dekkers E. M. J. (1977) Ammonia determination based on indophenol formation with sodium salicylate. *Water Research* **12**, 399–402.

Watson R. T., Meira Filho L. C., Sanhueza E. and Janetos A. (1992) Sources and sinks. In *Climate Change* 1992, *The Supplementary Report to the IPCC Scientific Assessment* (J. T. Houghton, B. A. Callander and S. K. Varney, Eds), pp. 25–46. Cambridge University Press, Cambridge.