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NITROUS OXIDE EMISSIONS FROM EXCRETA APPLIED IN A SIMULATED GRAZING PATTERN

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Summary—N_2O emissions were measured from cattle dung and urine applied to six separate experimental areas over a period of 15 months, to represent distinct components of a grazing season. Application of livestock excreta increased N_2O emissions significantly over that measured from control (untreated) plots and fluxes up to 290 μg N m^{-2} h^{-1} from dung and 192 μg N m^{-2} hr^{-1} from urine were measured. No significant correlations were observed between N_2O fluxes and environmental factors, such as rainfall and soil mineral-N. This was attributed to the specific physical and biogeochemical processes in the excreta that might override other environmental factors at our plots. Total N_2O—N losses from dung and urine patches over 100 d represented up to 0.53% and 1% respectively, of the N excreted. The average annual N_2O fluxes were approximately five times greater from the urine patches than from the dung, and from the excreta deposited during wet conditions (autumn) than during dry conditions (summer). Our results suggest that excreta deposited on grassland from grazing animals is an important source of N_2O, and can contribute up to 22% of the total N_2O emission from U.K. grassland. © 1998 Elsevier Science Ltd. All rights reserved

INTRODUCTION

Nitrous oxide (N_2O) is an important intermediate product of microbial nitrification and denitrification in soils (e.g. Davidson, 1991). Although chemically relatively inert in the troposphere, with an atmospheric lifetime of about 150 y, it has been implicated in the destruction of stratospheric ozone and in contributing to global warming (e.g. Warneck, 1988). While estimates of the global N_2O emissions from many of the major sources of this gas have been made (Bouwman, 1994), fluxes from grassland are still poorly defined (INDITE, 1994; Bouwman and Asman, 1996; Lee *et al*., 1996). About 70% of the total U.K. land surface area is grassland (Waters, 1994), which may be an important source because of inputs of nitrogen in fertilizer and excreta, often to soils which have anaerobic characteristics for a good deal of the year. Applications of N fertilizer can increase N_2O emissions from soils (e.g. Harrison *et al*., 1995) so that the potential for emissions from intensive managements may be large (Jarvis *et al*., 1996). While estimates of N_2O emissions derived from N fertilizer have received much attention (Eichner, 1990), information on the influence of animal excreta on N_2O production is still limited.

Inputs of excretal N to grassland are generally in forms other than NO_3^-. Thus an initial nitrification step is important to provide the essential substrate (NO_3^-) for the microbial denitrification processes, as well as providing the opportunity for release of N_2O from the nitrification process itself. There is competition for the removal of substrates for nitrification and denitrification. Leaching, absorption by plants or utilization by microorganisms indirectly influence the production of N_2O. Clough *et al*. (1996) examined the fate of synthetic urine-N from a field lysimeter experiment and found that up to 47% of the inorganic-N was lost by leaching and up to 35% by plant uptake, depending on soil type and treatment. Therefore soil and meteorological factors may strongly control N_2O production, transport and emissions to the atmosphere, directly and indirectly. The two processes responsible for N_2O production may be closely linked or coupled, can occur concurrently and have common intermediates.

It has been estimated that the 250 kt NH_4^+–N stored in livestock wastes can account for more than 12% of the total N_2O—N emissions from all terrestrial sources in the U.K. (INDITE, 1994) when applied to land. Excreta deposited while animals are grazing may also be a source. However, data on N_2O emissions from animal excreta in grazed grasslands are few and interactions of animal excreta with environmental conditions and the global N_2O flux (Flessa *et al*., 1996) are still uncertain. Allen *et al*. (1996) showed that there were some major differences in N_2O emissions from dung and urine, depending on when these were deposited

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and on which type of soil. This research has been extended in the present study to measure N_2O emission from cattle dung and urine applied through a simulated seasonal grazing pattern.

MATERIAL AND METHODS

Site description and treatments

The experiments were taken on a long-term grass sward (Burrows Field) at the Institute of Grassland and Environmental Research, Devon, in S.W. England. The soil, Halstow series, was a poorly-drained silty clay loam with an approximate composition of 22% sand, 47% silt and 31% clay (Harrod, 1981) with a pH of about 5.5. The site is slightly sloping (3° WSW) and the sward, comprising perennial ryegrass and a mixture of other grasses, had not been fertilized or grazed for at least 12 months prior to the measurements. The grass was cut and removed periodically during the grazing season.

Lantinga *et al.* (1987) have indicated that the average animal excreta-N deposition is approximately 500 kg N ha^{-1} on each occasion that dung is dropped for cattle with a herbage intake of 16 kg dry matter d^{-1}. This is equivalent to 6 g N of dung and 1.5 g N of urine per each 20 cm area used in this study. Typical dung and urine properties are shown in Table 1. From the N content of the excreta (Table 1), it was estimated that a single excretion will be equivalent to 1400 g of dung and 180 g urine. However, because of the different daily intake of the animals used to provide excreta in our study, it was suggested that 1200 g of dung and 200 ml of urine would be a reasonable overall estimation (Allen *et al.*, 1996). Applications at these rates were repeated six times on separate experimental areas over a period from July 1994 to September 1995 to simulate excreta deposition during different grazing seasons. For convenience, these repeated applications will be referred to as experiments 1–6. The dates of treatment applications in each experiment during 1994 were 1 September, 16 September and 21 October (experiments 1–3, respectively) and during 1995 were 31 May, 17 July and 4 September (experiments 4–6, respectively).

During each experiment, a uniform area was divided into three sub-plots each of about 40 m^2, two being used for dung and urine applications and one left untreated as a control. To each of the dung and urine plots, 60 samples (reduced to 48 during experiments 4–6) of dung (1.2 kg) or urine (200 ml) were applied to 20 cm dia. areas each separated by approximately 70 cm. N_2O emission rates and soil variables (available NH_4^+ and NO_3^- and soil moisture) were then measured from each application within each experiment, immediately after the application and periodically up to November 1995.

Excreta collection and analysis

All excreta samples were collected directly from dairy cows while tethered at milking. To obtain sufficient dung and urine for each of the experimental runs, samples were collected for a period of up to one week and kept refrigerated at 4°C. Before each treatment, samples were mixed and applied as mentioned previously. The cows were fed on grass silage and concentrate diets and on one occasion (experiment 6) on grass silage and kale to supplement their diets at grazing. Fresh samples of dung and urine were also analysed for the total N and C content before the applications (Table 1). The total N and C of the dung and urine was measured using a combustion C/N analyser (NA 1500, Carlo Erba).

Flux measurements and analysis

N_2O flux measurements were carried out using the closed-chamber technique. Chambers were made from galvanized steel cylinders, each of about 40 cm dia. with a 2 cm flange on the top covered with a rubber gasket. Each chamber lid was made from perspex, fitted with a 3-way stopcock for gas sampling and a rubber seal which fitted tightly onto the chamber gasket. Before each measurement, six chambers per treatment (reduced to three during experiments 4–6) were positioned carefully to avoid disturbance, over a dung or urine patch, and inserted into the soil to a depth of 5 cm to leave an enclosed headspace volume of *ca.* 31 1. The area covered by the chamber was equivalent to approximately five times the area treated with excreta to take account of the total area affected because of diffusion of N from the excreta in the soil (Lantinga *et al.*, 1987). Care was taken to ensure that the individual dung or urine patches were selected randomly for each flux measurement, in order to take account of the spatial variability within each plot.

During each flux measurement (generally between 9.00 and 12.00 h), chambers were closed for 40 min and duplicate gas samples of 40 ml of the chamber atmosphere at 0 and 40 min were taken by polypropylene syringes. The syringes were immediately used

Table 1. Properties of typical excreta samples (1.2 kg dung and 200 ml urine). Calculations are on w/w basis for dung and w/v for urine

			Fresh weight		Dry weight			
	g N $sample^{-1}$	g C $sample^{-1}$	N%	C%	N%	C%	C to N ratio	Fresh weight moisture %
Dung	5.19	77.34	0.43	6.45	2.88	42.97	14.97	85
Urine	1.69	2.83	0.84	1.41	—	—	1.70	—

to fill (under atmospheric pressure) 20 ml vials fitted with PTFE-silicon septa. The vials were then loaded into an automated Hewlett-Packard Headspace sampler (type 19395A) connected to a GC (ATI-Unicam 610) with electron capture detector for N_2O analysis. The GC had a pre-flush system with a stripper column of porapak N (1.0 m × 3mm) and an analytical column of Hay Sep Q (2.0 × 3.mm), both 80–100 mesh, and held at 50°C oven temperature. The detector temperature was 300°C and the carrier gas was N_2(40 ml min^{-1}). The GC was attached to an integrator (ATI-Unicam 4815), and N_2O contents of the samples were determined within 8 h with a precision of 1.5% for repeated analysis of ambient N_2O concentration. The flux was determined from N_2O concentrations above ambient by the following equation:

$$FN_2O = k(C_t - C_0)(273/T)(V/A)$$

where F is the flux of N_2O (μg N_2O–N m^{-2} h^{-1}), k is a unit conversion factor (1.25 μg N μl^{-1}), C_t–C_0 is the change in N_2O concentration measured at time 0 and t respectively (μ l l^{-1} h^{-1}), T is the temperature within the chamber (K), V is the air volume within the chamber (l) and A is the soil area enclosed by the chamber (m^2).

Soil sampling and analysis

To reduce the influence of the spatial heterogeneity during soil sampling within each treatment, dung and urine patches were selected randomly during each experiment to provide sites for soil sampling. Similar sites were also selected within the control plot. From each of the selected dung or urine patches, three soil cores (each 25 mm dia) were collected, one from the centre of the patch, one from the edge and one from the outside of the patch at a distance equivalent to the radius of the flux chamber (i.e. 20 cm). Soil cores from each treatment were collected from the top 10 cm of the soil and divided into two separate horizons (i.e. 0–5 cm and 5–10 cm) and the fractions from each horizon from the three cores were mixed together for processing and analysis.

Soil available NH_4^+ and NO_3^- were determined by extracting 50 g of soil with 100 ml 1M KCl and subsequent analysis of these ions using standard continuous flow colorimetric methods. Soil moisture content was determined gravimetrically by the weight loss of fresh soil samples on drying at 105°C overnight. The pH was measured in 1:2.5 soil-to-water suspensions.

Data for ambient air, soil temperature at 10 and 39 cm depth and rainfall during the entire experimental period were obtained from a meteorological station approximately 100 m from the experimental plots. Soil temperature showed little variation between 10 and 30 cm depth and ranged from 2 to 20°C during the period when the experiments were undertaken. Variations in rainfall were very large. The total cumulative rainfall for 30 d after excreta application during experiments 1, 3 and 6 was up to four times greater than the corresponding cumulative amounts during experiments 2, 4 and 5. Variation in the soil moisture (% dry weight) was generally much smaller and was between 25 and 45% during the corresponding 30 d after application of excreta in all the experiments. Detailed information on typical meteorological conditions (rain, temperature, wind direction, etc.) at our site is given by Allen *et al.* (1996).

Statistical analysis

Linear regression was used to identify the relationships between N_2O emissions and meteorological and soil variables. Analysis of variance was used to assess the effect of treatment and time of application on N_2O emissions.

RESULTS

N_2O flux from dung pats

An example of the N_2O fluxes from the dung and urine patches and from the control plots during experiments 4 and 5 are shown in Fig. 1. A number of general points relating to all experiments can be made, firstly N_2O emission rates from the dung treatments showed large variation from all applications and throughout all measurement periods. Fluxes ranged from -41 ± 29.4 (of the mean) to 290 ± 81.1 μg N m^{-2} h^{-1} during all experiments. Generally, emission rates peaked within 40 d of application. Variations in the peak emission rates between the experiments were high and ranged from 80 ± 17.9 (3 d after the dung application in July 1994, experiment 5) to 290 ± 81.1 μg m^{-2} h^{-1} (124 d after the application in September 1994, experiment 2).

Correlations between N_2O emission rates and soil NH_4^+ or NO_3^- were not significant. Also, in most of the experiments, no correlations were observed between N_2O emission rates and soil temperature, moisture or rainfall. However, correlations were observed with the soil surface temperature (10 cm) during experiment 2 ($P < 0.01$) and with rainfall ($P < 0.1$) during experiments 2 and 3.

Figure 2 shows a typical example of concentrations of the available NH_4^+ and NO_3^- measured after applications of dung and urine during experiment 6, in top 0–5 cm soil horizons where most of the variation ocurred. The background concentration of the available NH_4^+–N (i.e. average concentrations measured from the control plots) was 4.6 μg N g^{-1} dry soil. The corresponding NO_3^- concentration was 1.6 μg N g^{-1} dry soil. The total available-N ($NH_4^+ + NO_3^-$) contents in the soil after application of dung were much smaller relative to the urine, but remained significantly higher than

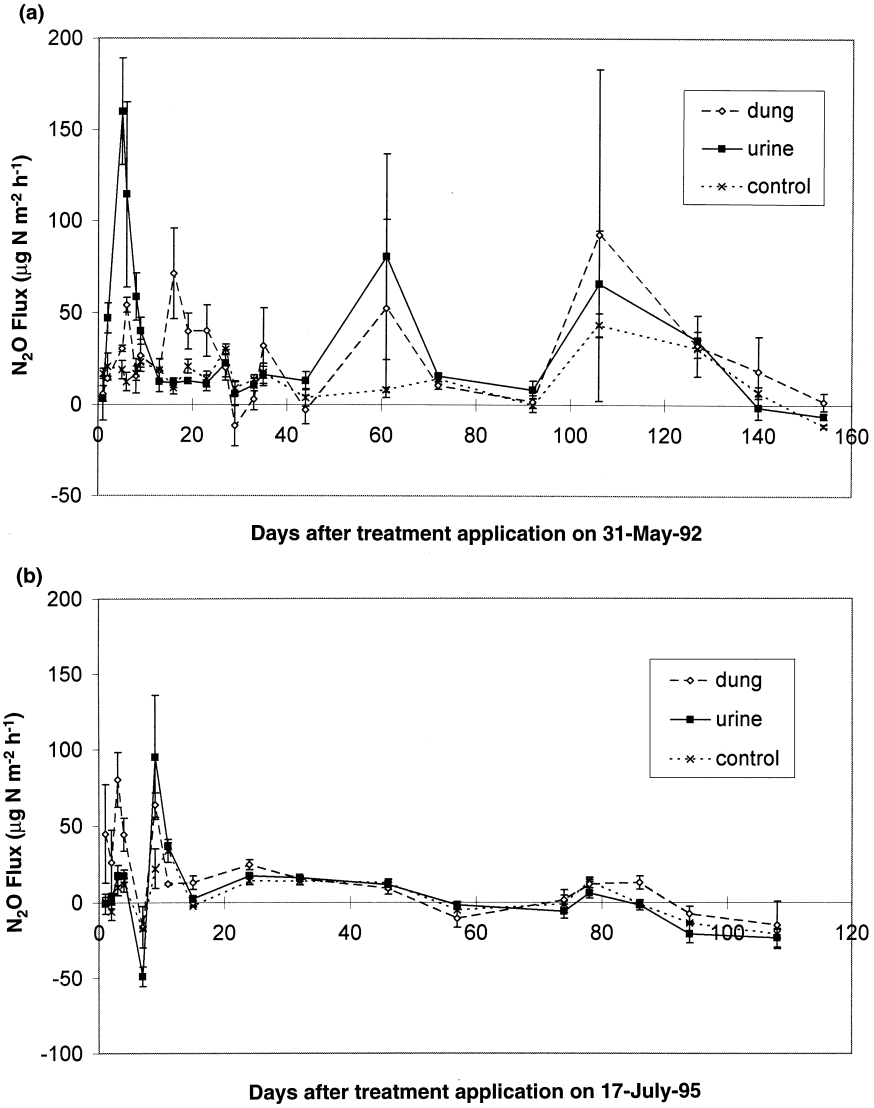

Fig. 1. Examples of the N_2O fluxes measured from the dung, urine and control plots during experiment 4 (a) and 5 (b). Data are means for three replicate chambers. Vertical bars are SE for means.

those observed in the urine treatments when these had returned to the background concentrations. All treatments, including the control plots, had high mineral-N contents in the top 0–5 cm soil horizon than in the lower 5–10 cm horizon and higher concentrations of NH_4^+ than NO_3^-.

N_2O flux from urine patches

Variation in N_2O emission rates from the urine treatment was large both between each experimental period and also within each experiment. N_2O fluxes did not differ substantially from those for dung and often followed similar trends (Fig. 1) with fluxes ranging from 90 (124 d after application in September 1994, experiment 2) to 192 ± 62.5 μg N m^{-2} h^{-1} (7 d after application in October 1994, experiment 3).

Correlations between N_2O emission rates and the soil NH_4^+ or NO_3^- were generally not significant and where they did exist were generally stronger with NH_4^+ than with NO_3^-. Emission rates of N_2O from the urine treatment generally showed weak correlations ($P < 0.1$) with soil temperature and rainfall. Application of urine increased the total available-N ($NH_4^+ + NO_3^-$) in the soil significantly over that from the dung application, but values decreased sharply thereafter, generally reaching a background content within 15 d (Fig. 2). When urine was applied during dry conditions, the available NH_4^+–N concentration in the top 0–5 cm soil horizon measured within the first few days after application was generally much higher than that measured in the lower 5–10 cm horizon. During wet conditions, the concentration of NH_4^+–N was ap-

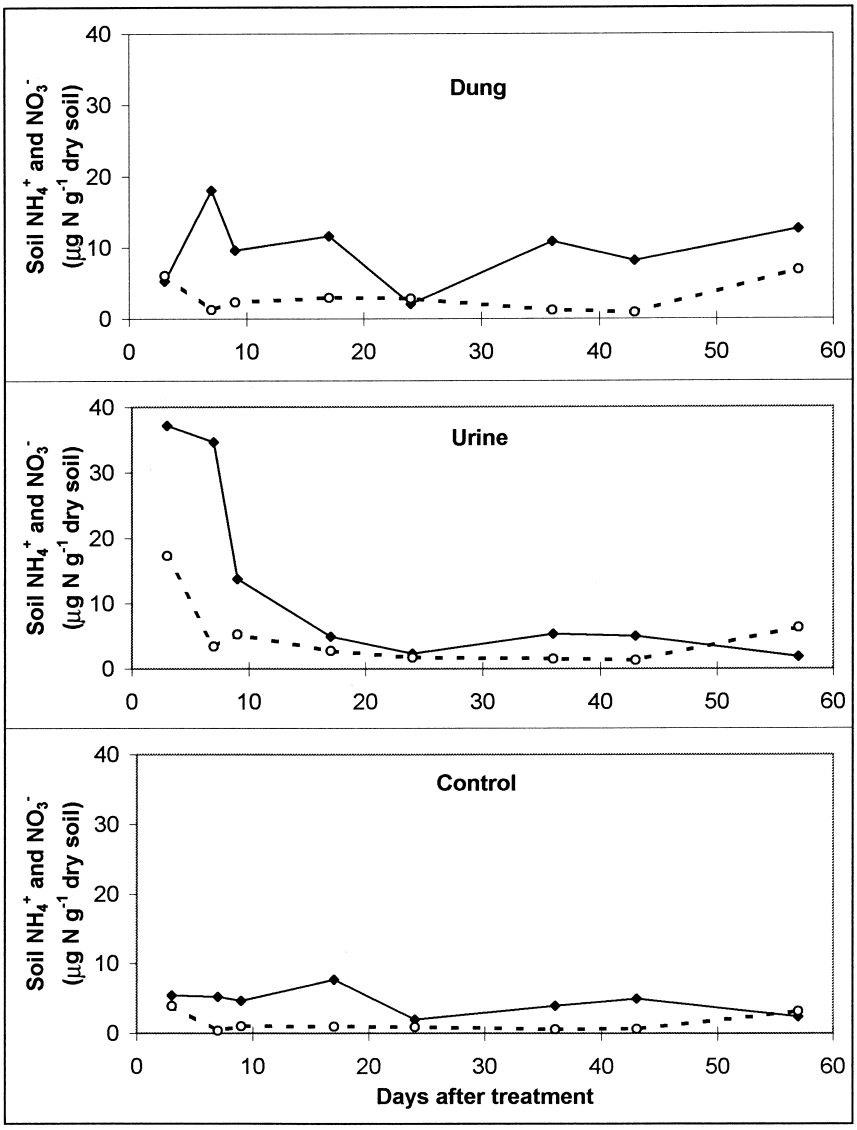

Fig. 2. Example of typical soil available NH_4^+ and NO_3^- measured from the dung, urine and control plots during experiment 6. Data points are averages of two analytical samples. -◆- NH_4^+ and -⊖- NO_3^-.

proximately equal in both soil horizons. In contrast, the initial NO_3^-–N concentration in both soil horizons generally increased to its maximum only when urine was applied during wet conditions. These observations, as expected, could be due to leaching to the deeper soil layer of the NO_3^- originally present in the soil and that derived from nitrification of the NH_4^+ in urine. This in turn could be an important factor affecting the volatilization of ammonia and the production and emission of N_2O from the mineral-N remaining in the soil.

Cumulative N_2O fluxes from excreta

The total cumulative flux measured over the whole observation period in each of the experiments (ranging from 60 to 417 d) from the dung and urine patches and from the control plots (determined from the area under the curves in Fig. 1) are shown in Fig. 3. However, to enable a comparison between the different experiments, the total cumulative flux from the excreta patches and the control plots in all the experiments were also measured over 100 d (except for experiment 6, in which fluxes were measured over 60 d) after applications (Fig. 3), where the overall effect of treatments was significant ($P < 0.001$). The total N_2O flux over the whole observation period during experiments 1–3 (approximately over 1 y) from the dung patches were 31, 45 and 25 mg N treatment^{-1} and from the urine patches were 30, 26 and 33 mg N, respectively. The corresponding fluxes over 100 d of observation were substantially lower with 13, 25 and 20 mg N treatment^{-1} from the dung and 15, 12 and 26 mg from the urine. The total flux of N_2O was generally

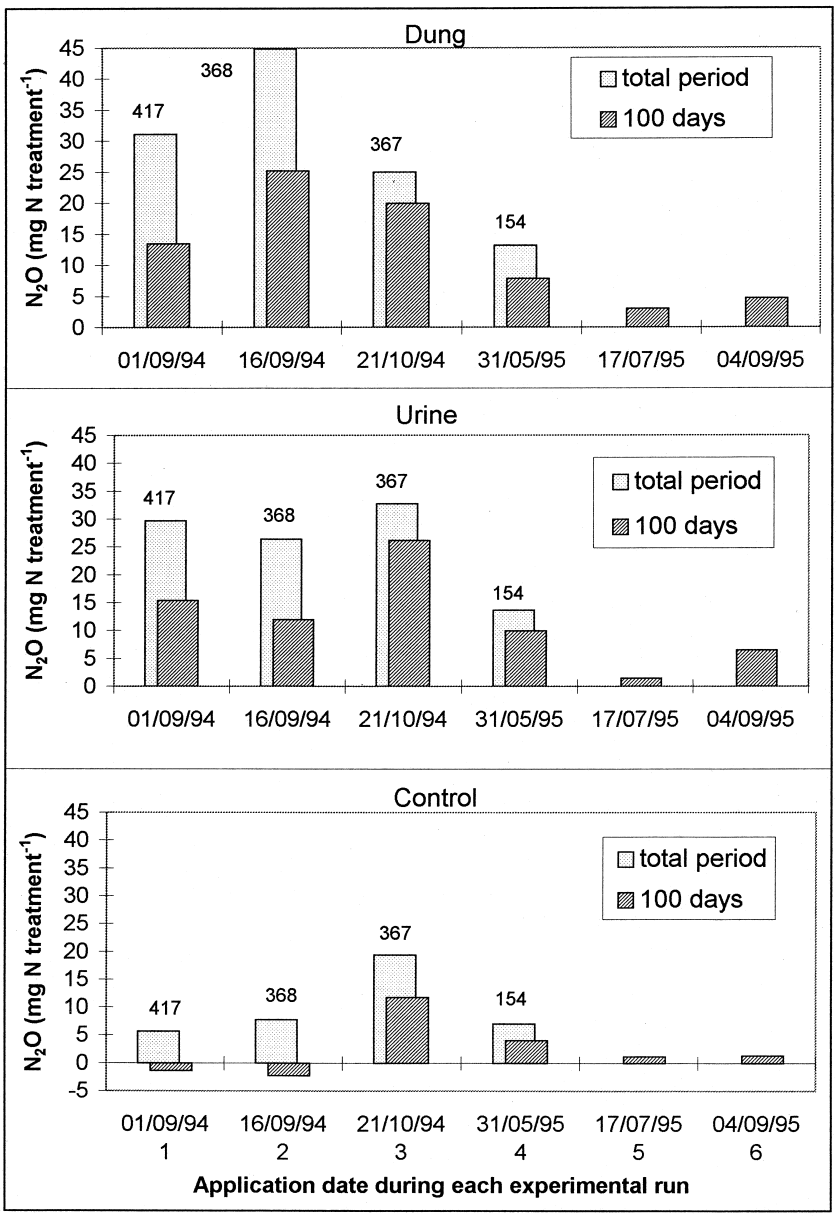

Fig. 3. The total cumulative flux of N_2O from the dung, urine and control plots during each experiment, calculated for the total days of observation (numbers on histograms) and for 100 d after application.

higher from excreta deposited during autumn (September and October) than from that deposited during summer (June and July), and varied from trace amounts to more than 25 mg N_2O–N Table 2 summarizes the excreta-induced N_2O-N emissions for a period of 100 d from excreta application from each experiment (cumulative flux from treatments for 100 d-control) expressed as a percentage of the dung or urine N deposited (Table 1). Percentage loss of N_2O-N from the dung-N varied from 0.004 in July to 0.53 in September over the 100 d. Corresponding higher percentage losses were observed from the urine and ranged from 0.02 to 1.

DISCUSSION

N_2O fluxes from excreta

There are relatively few studies on the effect of urine on N_2O emissions and the work carried out has often either used artificial urine (deKlein and van Logtestijn, 1994: Clough *et al.*, 1996) or controlled laboratory conditions (Monaghan and Barraclough, 1993). Field information on emissions of N_2O from excreta deposited as dung are even scarcer (e.g. Allen *et al.*, 1996; Flessa *et al.*, 1996).

Analysis of variance showed that the overall effect of the treatments on N_2O fluxes, over 100 d in all the experiments, differed significantly

($P = < 0.009$) with time of application. These differences cannot be explained by variation in treatment application as the experimental handling procedures and the application rates did not differ between the experiments and should not have directly influenced fluxes. Interactions between the dung or urine patches and the environmental factors was the most likely factor affecting N_2O emission rates during the various experiments. The specific influence of measured environmental factors on N_2O emission processes, however, was not clear. Correlations between N_2O emissions and available NH_4^+ and NO_3^- or meteorological factors were generally not significant. Multiple linear regression analysis between N_2O fluxes and both soil (NH_4^+, NO_3^- and moisture) and meteorological (temperature and rain) factors also showed weak correlations. Other studies have also shown that N_2O fluxes from grassland systems are poorly correlated with soil variables (e.g. Velthof *et al.*, 1996). Similarly, Allen *et al.* (1996) found no correlations between N_2O emissions and soil mineral-N, either in laboratory experiments or in field measurements during spring and summer under similar experimental conditions to our studies. However, Allen *et al.* (1996) did observe significant correlations during an autumn and winter experiment. Interactions between soil variables and other specific influences of excreta, such as increasing soil pH when NH_3 volatilizes from urine and decreased gaseous diffusion as the dung pats dry out, may over-ride other environmental effects, particularly within a few days of excreta deposition. Determination of variables in bulk samples may also not represent accurately the integrated effect of soil interactions which may occur within microsites (Velthof *et al.*, 1996). Therefore several factors could obscure relationships between N_2O emissions and soil and meteorological variables, because of the large number of processes involved, presumably all with limiting factors.

The contributions of microbial processes involved in N_2O emissions after excreta are deposited can not be easily separated. Higher emissions were generally observed during and after rainfall periods. Peak emissions from dung and urine patches occurred after the soil NH_4^+ peak had declined. However, the available NO_3^- concentrations measured during the experiments were always low (i.e. $< 20\ \mu g\ N\ g^{-1}$ dry soil) probably indicating a rapid reduction of the indigenous NO_3^- by denitrification. Oenema *et al.* (1997) indicated that nitrification is potentially an important and lasting source of N_2O in urine and dung patches. This is because a temporary accumulation of NO_2^- in urine patches could be expected due to nitrite-oxidizing bacteria being more rapidly inhibited than ammonia-oxidizing bacteria (Monaghan and Barraclough, 1993). This temporary accumulation of NO_2^- could increase the release of N_2O via nitrification (Oenema *et al.*, 1997). Whilst some studies have shown that denitrification can be an important source of N_2O emission following urine application (deKlein and van Logtestijn, 1994), others have found a strong involvement of the nitrification process (Jarvis *et al.*, 1994; Allen *et al.*, 1996).

Loss rate of excreta-N as N_2O

To quantify the influence of excreta as a global source of N_2O, it is necessary to calculate the percentage loss of N within the deposited excreta (dung or urine) as N_2O (Table 2) calculated for the 100 d period. The results generally showed that the proportion lost as N_2O was highly dependent on the time of the grazing season when excreta were deposited. Higher losses were observed during autumn than during summer, except during experiment 6 which showed relatively lower losses when excreta was applied in October 1995 in comparison to those observed during October 1994 (experiments 1 and 2). This could be due to the fact that fluxes were only measured for a period of 60 d during experiment 6, which might underestimate the total loss. The total cumulative N_2O emission for a 1 y period of observation after excreta application, could be double of that measured over only 100 d (Fig. 3). The period of observation is therefore a very important factor and should be addressed in future studies of losses particularly from excreta. However, the lower N_2O loss rates observed during experiment 6 could also be related to the different environmental conditions during excreta applications. The cumulative rainfall during 7 d before excreta application during experiment 6 (application on 4 September 1995), for example, was only 4.9 mm in comparison to 32 mm rain during experiment 1 (application on 1 September 1994). In contrast, the corresponding cumulative rainfall after excreta application in both experiments was 44 and 22.9 mm, respectively.

Recent values of the percentage loss of N_2O–N from urine, of 0.1–2.4 from Belgium (Vermoesen *et al.*, 1996), < 1–3 from New Zealand (Clough *et al.*, 1996) and 0.5 from the Netherlands (Velthof and Oenema, 1994), fall within the range in our study. Poggemann *et al.* (1995) and Flessa *et al.* (1996) from Germany, have also reported similar values of 0.4 and 0.47%, respectively, for dung and 0.4–1.3% and 3.8%, respectively, for urine. The higher percentage loss of N_2O from urine measured by Flessa *et al.* (1996) could be due to the higher estimated deposition rate of urine per animal to that used in our study. Comparing the percentage loss of N_2O–N from excreta to that from NH_4NO_3 fertilizer of 0.93% measured from arable land (Yamulki *et al.*, 1995) and 0.82% measured from adjacent grassland plots under similar conditions to the current measurements (Yamulki and Jarvis, 1996), shows

Table 2. Summary of the integrated N_2O fluxes and excreta-induced emissions during the first 100 d after dung or urine application from each experiment and estimation of the annual emission from excreta return by grazing animals, based on a stocking rate of two animals ha^{-1} (cattle) and 180 grazing d y^{-1}

Experimental runs	Date of treatment application	Treatment-induced emission*		Percent loss of treatment-N as N_2O-N		Annual flux g N_2O-N ha^{-1} y^{-1}			Estimated U.K. emissions from excreta Tg N_2O–N y^{-1}
		Dung	Urine	Dung	Urine	Dung	Urine	Total	
1	01-09-94	14.92	16.89	0.29	1.00	89.5	608.2	697.7	0.0035
2	16-09-94	27.61	14.32	0.53	0.85	165.7	515.6	681.3	0.0034
3	21-10-94	8.13	14.38	0.16	0.85	48.8	517.6	566.3	0.0028
4	31-05-95	3.76	5.87	0.07	0.35	22.6	211.4	234.0	0.0012
5	17-07-95	1.85	0.25	0.04	0.02	11.1	9.2	20.3	0.0001
6**	04-09-95	3.36	5.18	0.07	0.31	20.3	186.5	206.8	0.0010
Mean Total		9.94	9.48	0.19	0.56	59.7	341.4	401.1	0.0020

*Expressed as mg N_2O-N $patch^{-1}$ of dung or urine (20 cm area).
**Measurements for 60 d only

that the contribution of excreta-N to the global N_2O emission is large and can be compared, per unit area, to that from fertilizer.

The total average N_2O–N emissions from dung treatment (9.9 mg N_2O–N $patch^{-1}$, Table 2) was equal to that from the urine (9.5 mg N_2O–N $patch^{-1}$). However, the average percentage loss from the urine was much higher than from the dung. The fact that about three times more N was added with dung than with urine on area basis, indicates that losses were related to the readily available form of N in urine rather than total N. It is also possible that the higher carbon content added with dung (Table 1) may promote further reduction of the N_2O to N_2 (Firestone and Davidson, 1989). This is similar to effects observed by Allen *et al.* (1996) from field experiments made on the same experimental site as in our study during autumn and winter and from laboratory experiments. However, their results indicated a higher percentage loss from dung applications than urine during spring and summer.

Annual N_2O emissions from excreta return by grazing animals

Assuming a daily deposition per animal (cow) of 10×2 kg each of dung and urine, a stocking rate of two animals ha^{-1} and 180 grazing d y^{-1} (Allen *et al.*, 1996), it was estimated that 7200 kg ha^{-1} y^{-1} are deposited as dung or urine during grazing by cattle. Based on this assumption, the total annual fluxes of N_2O were calculated. Annual N_2O emissions of up to 608 g N_2O–N ha^{-1} y^{-1} from the urine deposited and up to 166 g N_2O–N ha^{-1} y^{-1} from the dung were calculated. The total annual N_2O–N emission from the urine patches was much higher than that from dung during autumn, perhaps as a result of the larger amounts of mineral-N available with urine. This could also be due to effects of moisture on the physical properties of dung (Marsh and Campling, 1970), and different trends in mineralization and immobilization. Therefore, during autumn NH_3 volatilization is expected to be much lower from the urine patches, because of lower temperatures (Hales and Drewes, 1979), reducing the competition between the grass and soil microorganisms for the available N and thus increasing the potential for emissions.

Assuming the total area of land used for grazing in the U.K. is 5 Mha (MAFF, 1989), a mean total flux of 0.002 Tg N_2O–N y^{-1} (2 kt N_2O–N y^{-1}) can be estimated for the whole of the U.K. from excreta (dung and urine) return by grazing animals (Table 2). This estimate is based on a limited database and to provide a measure of the uncertainty requires measurements from other soils over a longer period. The total N_2O emission rates from U.K. grassland is estimated to be 16 kt N_2O–N y^{-1} (Fowler *et al.*, 1996). This indicates that up to

21.9% (12.5% on average) of the total N_2O emission from U.K. grassland is due to excreta return by grazing animals. Our results also show that the average emission from excreta during autumn (538 g N_2O–N ha^{-1} y^{-1}), is more than four times greater than during summer (127 g N_2O–N ha^{-1} y^{-1}). Measurements from New Zealand (Müller *et al*., 1997) have shown similar results, in which the total emission from urine applied in autumn (1850 g ha^{-1} y^{-1}) was approximately four times greater than during summer (430 g ha^{-1} y^{-1}) and a total emission rate of 1470 g ha^{-1} y^{-1} during spring. Our results also showed that the average emission for the entire experiment from the urine patches was more than five times greater than that from the dung (Table 2). However, these estimates could vary substantially with soil type, the dietary intake of the grazing animals (i.e. the C-to-N ratio of the deposited excreta) and the environmental conditions during and after grazing. Estimates of the U.K. emission during winter (not measured in this study) are likely to be higher due to more anaerobic soil conditions which favour denitrification. However, most of the cattle in the U.K. are housed at that time giving no excreta returns through grazing. Therefore our total estimates are applicable to the full year.

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REFERENCES

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

Allen A. G., Jarvis S. C. and Headon D. M. (1996) Nitrous oxide emissions from soils due to inputs of nitrogen from excreta return by livestock on grazed grassland in the U.K.. *Soil Biology & Biochemistry* **28,** 597–607.

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

Bouwman, A. F. and Asman, W. A. H. (1997) Scaling of nitrogen gas fluxes from grasslands. In Gaseous Nitrogen Emissions from Grasslands, eds. S. C. Jarvis, and B. F. Pain, pp. 311–330. CABI, Wallingford.

Clough T. J., Sherlock R. R., Cameron K. C. and Ledgard S. F. (1996) Fate of urine nitrogen on mineral and peat soils in New Zealand. *Plant and Soil* **178,** 141–152.

Davidson E. A. (1991) Fluxes of nitrous oxide and nitric oxide from terrestrial ecosystem. In *Microbial Production and Consumption of Greenhouse Gases: Nitrogen Oxide, and Halomethanes*, ed. J. E. Rogers and W. B. Whitman, pp. 219–235. American Society for Microbiology, Washington.

deKlein C. A. M. and van Logtestijn R. S. P. (1994) Denitrification in the top soil of managed grasslands in The Netherlands in relation to soil type and fertilizer level. *Plant and Soil* **138,** 33–44.

Eichner M. J. (1990) Nitrous oxide emissions from fertilized soils: summary of available data. *Journal of Environmental Quality* **19,** 272–280.

Firestone M. K. and Davidson E. A. (1989) Microbiological basis of NO and N_2O production and consumption. In *Exchange of Trace Gases Between Terrestrial Ecosystems and the Atmosphere*, ed. O. M. Andreae and D. S. Schimel, pp. 7–21. John Wiley, Chichester.

Flessa H., Dorsch P., Beese F., Konig H. and Bouwman A. F. (1996) Influence of cattle wastes on nitrous oxide and methane fluxes in pasture land. *Journal of Environmental Quality* **25,** 1366–1370.

Fowler D., Skiba U. and Hargreaves K. J. (1997) Emissions of NO and N_2O from grassland. In Gaseous Nitrogen Emissions from Grasslands, eds S. C. Jarvis and B. F. Pain, pp. 147–164. CABI, Wallingford.

Hales J. M. and Drewes D. R. (1979) Solubility of ammonia in water at low concentrations. *Atmospheric Environment* **13,** 1133–1147.

Harrison R. M., Yamulki S., Goulding K. W. T. and Webster C. P. (1995) Effect of fertilizer application on NO and N_2O fluxes from agricultural fields. *Journal of Geophysical Research* **100,** 25923–25931.

Harrod T. R. (1981) *The soils of North Wyke and Rowden*. Unpublished report, Soil Survey of England and Wales to Grassland Research Institute, Harpenden.

INDITE (1994) Impacts of nitrogen deposition in terrestrial ecosystems, October 1994. *Report of The United Kingdom Review Group on Impacts of Atmospheric Nitrogen*. Prepared on behalf of the Department of the Environment, London.

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.

Jarvis S. C., Wilkins R. J. and Pain B. F. (1996) Opportunities for reducing the environmental impact of dairy farming managements: a systems approach. *Grass and Forage Science* **51,** 21–31.

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?*, ed. H. G. Van der Meer, R. J. Unwin, T. A. Van Dijk and G. C. Ennik, pp. 103–117. Martinus Nijhoff, Dordrecht.

Lee D. S., Bouwman A. F., Asman W. A. H., Dentener F. J., van der Hoek K. W. and Olivier J. G. J. (1997) Emissions of nitric oxide, nitrous oxide and ammonia from grasslands on a global scale. In Gaseous Nitrogen Emissions from Grasslands, eds S. C. Jarvis and B. F. Pain, pp. 353–371. CABI, Wallingford.

Marsh R. and Campling R. C. (1970) Fouling of pastures by dung. *Herbage Abstracts* **40,** 123–130.

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

Müller C., Sherlock R. R. and Williams P. H. (1997) Factors influencing N_2O emissions from dung pasture soil affected by animal urine or ploughing. *European Journal of Soil Science* (in press).

Oenema O., Velthof G. L., Yamulki S. and Jarvis S. C. (1997) Nitrous oxide emissions from grazed grassland. *Soil Use and Management* **13**, 288–295.

Poggemann S., Weisbach F. and Künzel U. (1995) N_2O-Freisetzung von Exkrementflecken auf dem Weideland. *VDLUFA* Schriftenreihe, **40**, 901–904.

Velthof G. L. and Oenema O. (1994) Effect of nitrogen fertilizer type and urine on nitrous oxide flux from grassland in early spring. In *Grassland and Society, Proceeding of the 15th General Meeting of European Grassland Federation*, ed. L. 't Mannetje and J. Frame, pp. 458–462. Wageningen Pers, Wageningen.

Velthof G. L., Jarvis S. C., Stein A., Allen A. G. and Oenema O. (1996) Spatial variability of nitrous oxide fluxes in mown and grazed grassland on a poorly clay soil. *Soil Biology & Biochemistry* **28,** 1215–1225.

Vermoesen A., Van Cleemput O. and Hofman G. (1997) Contribution of urine patches to the emission of N_2O. In *Gaseous Nitrogen Emissions from Grasslands*, eds S. C. Jarvis and B. F. Pain, pp. 189–194. CABI, Wallingford.

Warneck P. (1988) *Chemistry of the Natural Atmosphere*. Academic Press, San Diego.

Waters G. R. (1994) Current government policy and existing instruments: balance in the countryside, In *Grassland Management and Nature Conservation*, eds. R. J. Hagger, S., Peel Proceedings of a joint meeting organized by the Britich Grassland Society and British Ecological Society, Leeds University, 27–29 September 1993, Occasional Symposium No. 28, pp. 3–9. The British Grassland Society, Reading.

Yamulki S. and Jarvis S. C. (1997) Nitrous oxide emissions from excreta from a simulated grazing pattern and fertilizer application to grassland. In *Gaseous Nitrogen Emissions from Grasslands*, eds S. C. Jarvis and B. F. Pain, pp. 195–199. CABI, Wallingford.

Yamulki S., Goulding K. W. T., Webster C. P. and Harrison R. M. (1995) Studies on NO and N_2O fluxes from a wheat field. *Atmospheric Environment* **29,** 1627–1635.