

# Methane Emission and Uptake from Soils as Influenced by Excreta Deposition from Grazing Animals

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## ABSTRACT

There have been only a few recent field-based measurements to estimate the impact of excreta from grazing animals on methane ( $\text{CH}_4$ ) exchange. We report measurements of  $\text{CH}_4$  exchange from cattle dung and urine deposited in a simulated seasonal grazing pattern. Dung and urine samples were deposited on six separate experimental plots at different times of the year to simulate different stages of normal grazing under UK conditions to study the effects of environmental factors on  $\text{CH}_4$  exchange rates. During all the experiments, dung pats were the main emitter of  $\text{CH}_4$  with an estimated mean daily rate of 0.72 (range 0.21 to 1.46) g  $\text{CH}_4\text{-C}$  per cow. Both net emission and consumption of  $\text{CH}_4$  from urine patches, however, were observed during the different experiments with a negligible mean daily emission of 0.02 (range -0.05 to 0.15) g  $\text{CH}_4\text{-C}$  per cow. Significant variations were observed between the experiments, despite the similarity of application rates and the composition of the excreta. These variations were mainly due to climatic differences when excreta were deposited: ambient temperature and rainfall were the most important factors. The estimated total annual emission of  $\text{CH}_4$  from cattle excreta deposition during the grazing season in the UK ranged between 0.42 and 3.26 (mean = 1.57) kt  $\text{CH}_4\text{-C}$ . Our work confirms other studies that suggest that excreta patches are insignificant sources of  $\text{CH}_4$  compared to the rumens of grazing cattle, and that climatic factors have a significant impact on  $\text{CH}_4$  emissions.

IT IS ESTIMATED that the total source strength of  $\text{CH}_4$  present in the atmosphere is  $505 \pm 105 \text{ Tg yr}^{-1}$  (Crutzen, 1991). Approximately 80% ( $405 \text{ Tg yr}^{-1}$ ) of the total  $\text{CH}_4$  emission comes from biogenic sources of which ruminant animals represent approximately 20% ( $80 \text{ Tg yr}^{-1}$ ) and the rest ( $325 \text{ Tg yr}^{-1}$ ) comes from landfills, rice (*Oryza sativa* L.) fields, natural wetlands, biomass burning, oceans, and insects (Crutzen, 1991). However, Steed and Hashimoto (1994) have estimated that the total  $\text{CH}_4$  emission from excreta and manure can account for approximately  $35.2 \text{ Tg yr}^{-1}$ , that is, about 9% of the total biogenic source. Although it is clear that excreta and manure can be an important source for  $\text{CH}_4$ , the total source strength is still in debate. In the UK about 31% of the total annual  $\text{CH}_4$  emissions is from agricultural sources (Williams, 1993), most of which originates from animal production (Jarvis et al., 1995).

Many ruminant production systems involve grazing, where interaction between soil, plant, and animals may influence net  $\text{CH}_4$  emission (Lockyer and Jarvis, 1995). Thus many factors will affect the overall production and/or oxidation of  $\text{CH}_4$  making estimation of an accurate budget of  $\text{CH}_4$ , particularly from grazing animals, a diffi-

cult task. For example, Jarvis et al. (1995) found a strong exponential relationship between  $\text{CH}_4$  release and the C/N ratio of dung, which in turn depended on the dietary intake of the animals. Methane production is highly temperature sensitive, with little being formed below  $10^\circ\text{C}$  (Steed and Hashimoto, 1994). Production of  $\text{CH}_4$  in soils requires strict anaerobic conditions. Well-drained aerated soil in contrast, acts as a sink for atmospheric  $\text{CH}_4$ . Thus under field conditions, variations in meteorological conditions, particularly rainfall and temperature during and after dung deposition, can be expected to strongly influence the overall  $\text{CH}_4$  emissions. Jarvis et al. (1995) found no effect of urine on  $\text{CH}_4$  emission, although the large amount of water and to some extent the soluble C in the urine might have been expected to increase the soil's methanogenic potential.

Although making only a small contribution to net emissions, it is important that accurate estimates of the impact of excreta from grazing animals on  $\text{CH}_4$  emission are made. There have been only a few field-based measurements on which to base accurate assessments (e.g., Jarvis et al., 1995; Flessa et al., 1996; Holter, 1997). The aim of the present work was therefore to study the effect of environmental factors on  $\text{CH}_4$  fluxes from the soil and the impact of excretal returns from grazing cattle in a simulated pattern of dung and urine deposition during a grazing season.

## MATERIALS AND METHODS

### Experimental Site and Treatments

The site used was a long-term pasture comprising perennial ryegrass and a mixture of other grasses at the Institute of Grassland and Environmental Research (IGER), in Devon, SW England. The sward had not been fertilized or grazed for at least 12 mo prior to the experiment and was cut and removed periodically during the grazing season. The soil was a poorly permeable silty clay loam (Halstow series: gleyic cambosol: ochrept) with an approximate composition of 22% sand, 47% silt, and 31% clay (Harrod, T.R. 1981. The soils of North Wyke and Rowden. Unpublished report. Soil Survey of England and Wales for Grassland Research Institute) and a pH of about 5.5.

In order to simulate the excreta return from grazing cattle on the sward, 48 samples of dung (1.2 kg) and 48 samples of urine (200 mL), representative of typical animal excreta deposition rates on a mass per area basis (Allen et al., 1996), were applied precisely to 20 cm diam. areas located within two respective plots, each of which was approximately  $40 \text{ m}^2$ . Full details concerning the estimation of the dung and urine sample application rates used during this study are given elsewhere (Yamulki et al., 1998). Another similar ( $40 \text{ m}^2$ ) adjacent plot was left untreated to act as a control. These application rates were repeated six times on separate experimental plots

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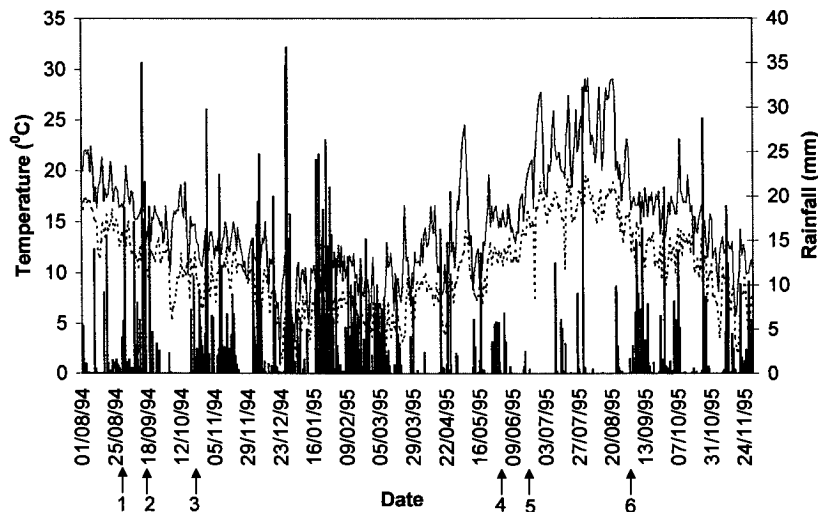


Fig. 1. Soil temperature at 10 cm (dotted line), air temperature (solid line), and rainfall data (column) for all the six experiments. Arrows indicate dates of treatment applications (to separate plots) during each experiment.

over a period of approximately 15 mo between July 1994 and September 1995 to simulate excreta deposition during different stages of a normal grazing pattern. For convenience, these repeated applications will be referred to as Exp. 1 to 6. In Exp. 1, 2, and 3, excreta were applied on 1 and 16 Sept. and 21 Oct. 1994, respectively, and on 31 May, 17 July, and 4 Sept. 1995 for Exp. 4, 5, and 6. Dung and urine samples were collected for a period of up to 7 d, directly from dairy cows fed on grass silage and concentrate diets and on one occasion (Exp. 6) on grass silage and kale fed as supplements to grazing; the collected excreta were mixed and kept refrigerated at 4°C before each treatment and then applied as described.

The average daily precipitation, ambient air temperature and soil temperature at 10 cm depth during each experiment are shown in Fig. 1. Generally, variations between soil temperature at 10 and 30 cm depth were minimal, but much larger variations were observed between the soil and the air temperature particularly during summer. Daily precipitation followed the normal seasonal pattern with higher rainfall during autumn and winter months. The mean daily sunshine varied from 0 h to approximately 15 h during the study.

### Gas Measurement Techniques

Methane fluxes were measured from three replicate excreta patches within each plot (i.e., within the 48 samples of dung or urine patches) and within the control plot, using a closed-chamber technique, in which galvanized steel cylinders (each 40 cm diam.) were positioned over the patches and carefully inserted into the soil (5 cm depth). Care was taken during each flux measurement to ensure that the three individual dung or urine patches were selected randomly, to take account of the spatial variability within each plot. The area covered by each chamber was equivalent to approximately five times the area of the excreta patch to take account of the total area affected because of diffusion of mobile materials from the excreta into the surrounding soil (Lantinga et al., 1987). The top of each chamber was covered with a rubber gasket to allow a perspex lid, fitted with a three-way stopcock valve for gas sampling and a rubber seal, to be fitted tightly onto the chamber gasket. During each flux measurement (generally taken between 0900 and 1200 h), chamber lids were closed for about 40 min after which gas samples were transferred by polypropylene syringes to 20 mL headspace sampler vials fit-

ted with polytetrafluoroethylene-silicon septa. The vials were then loaded into an automated Hewlett-Packard Headspace sampler (type 19395A) connected to a GC (ATI-Unicam 610) with FID for  $\text{CH}_4$  analysis. The GC had a pre-flush system with a stripper column of porapak N (1.0 m by 1/8 in) and an analytical column of Hay Sep Q (2.0 m by 1/8 in), both 80 to 100 mesh, and held at 50°C oven temperature. The GC was attached to an integrator (Konik instruments SP 4290), and  $\text{CH}_4$  contents of the samples were determined within 8 h with a precision of 1% of repeated analysis of  $\text{CH}_4$  at the ambient background level. Generally  $\text{CH}_4$  flux measurements were made more frequently directly after application of the treatments and reduced when flux values were similar to that from the control plot.

### Soil Sampling and Analysis

Soil samples were collected during each experiment from six randomly allocated dung and urine patches within each plot and also from within the control plot to reduce the influence of the spatial heterogeneity during sampling. From each allocated patch area, three soil cores were collected periodically, one from the center of the patch, one from the edge and one from the outside at a distance equivalent to the radius of the flux chamber (i.e., 20 cm). Soil cores from each treatment were collected from the 0 to 5 cm and 5 to 10 cm layers and the contents from each layer from the three cores were mixed together for processing and analysis for pH,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and moisture content. The pH was measured in a 1:2.5 soil/water suspension. The soil-available  $\text{NH}_4^+$  and  $\text{NO}_3^-$  ( $\text{mg N kg}^{-1}$  dry soil) were determined by extraction with 1 M KCl solution (50 g of soil, 100 mL solution) and subsequent analysis of these ions using standard continuous flow colorimetric methods (Searle, 1984; Kamphake et al., 1967). Soil moisture content was determined gravimetrically by the weight loss on drying at 105°C overnight. Soil temperatures at 10 and 30 cm depth and rainfall during all the experiments were obtained from a meteorological station within 25 m of the site.

Samples of dung and urine were also analyzed for their total N and C contents before their application (Table 1). Duplicate samples of approximately 10 mg of freeze-dried dung samples and 100  $\mu\text{L}$  of urine absorbed on a chromosorb W/AW (80–100 mesh) were analyzed for the total N and C using a combustion C/N analyzer (Carlo Erba, NA 1500).

All data were subjected to statistical analysis. Linear regres-

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

	g N/sample	g C/sample	N%	C%	C/N ratio	Moisture % Fresh wt.
Dung	5.19	77.34	2.88	42.97	14.97	85
Urine	1.69	2.83	—	—	—	—

sion was used to identify relationships between  $\text{CH}_4$  fluxes and meteorological and soil variables. Analysis of variance was used to assess the effect of treatment and time of application on  $\text{CH}_4$  fluxes.

## RESULTS

### Methane Fluxes from the Dung Pats

A typical example of  $\text{CH}_4$  fluxes from the dung, urine, and the control plots measured during Exp. 4 (treatment applied on 31-05-95) is shown in Fig. 2. During all the experiments, significant  $\text{CH}_4$  emissions ( $P < 0.001$ ) were observed from the dung pats immediately after applica-

tion. Peak emissions were observed within 24 h after application during Exp. 1, 2, 3, and 4 with a highest peak rate of  $4520 (\pm 189)$ , standard error of the mean)  $\mu\text{g C m}^{-2} \text{ h}^{-1}$  observed from Exp. 4. However, during Exp. 5 and 6,  $\text{CH}_4$  emissions peaked 48 and 72 h after application, respectively; the lowest peak emission rate of  $1290 (\pm 155)$   $\mu\text{g C m}^{-2} \text{ h}^{-1}$  was observed in Exp. 5. Emission rates declined to the background levels of the control plots approximately 15 d after application of the dung and both deposition (oxidation) and emissions of  $\text{CH}_4$  were observed thereafter. Fluxes of  $\text{CH}_4$  from the dung pats from all the experiments showed that 62% of the values indicated net  $\text{CH}_4$  emissions and, typically, fluxes were in the range of  $-10$  to  $+10$   $\mu\text{g C m}^{-2} \text{ h}^{-1}$  for the background soil and rose to more than  $1000$   $\mu\text{g C m}^{-2} \text{ h}^{-1}$  immediately after dung application. Seasonal effects, that is, effect of time of application on  $\text{CH}_4$  fluxes, were greater immediately after dung applications and negligible thereafter during all the experiments.

Soil mineral N concentrations ( $\text{NH}_4^+$  and  $\text{NO}_3^-$ ) at 0

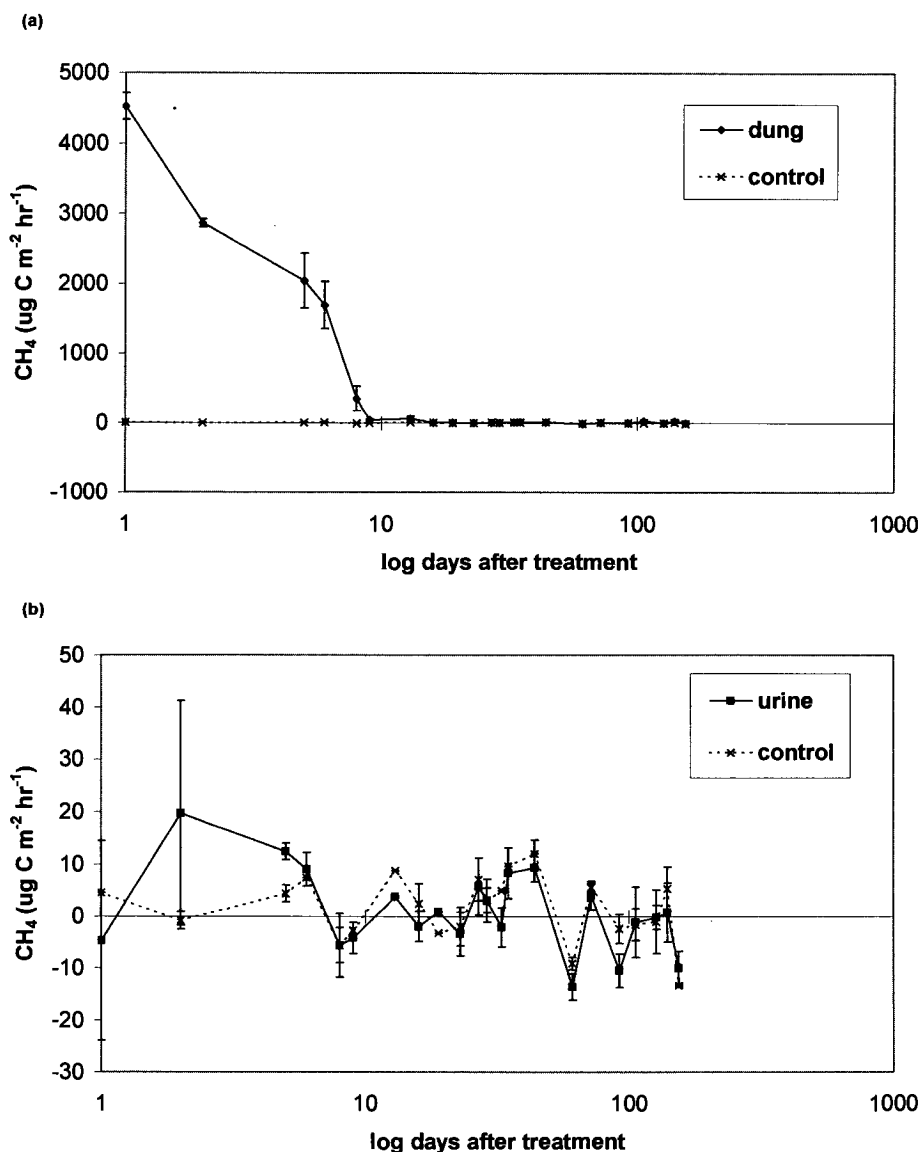


Fig. 2. A typical example of  $\text{CH}_4$  fluxes measured from the dung (a), urine (b) and the control plots during Exp. 4.

**Table 2.** Summary of the soil mineral N ranges (mg NH<sub>4</sub>NO<sub>3</sub> kg<sup>-1</sup> dry soil) and the soil moisture content (dry basis) measured during Exp. 4, 5, and 6.† Other details are given below.‡

Experiment	Soil depth	Dung	Urine	Control	Soil moisture
	cm				%
4	0–5	5.1–16.7	3.9–160.5	2.5–12.6	17.5–287.0
	5–10	2.4–8.7	2.2–29.3	1.7–6.8	17.1–176.6
5	0–5	4.6–27.1	5.5–58.5	3.4–14.0	17.7–49.2
	5–10	2.6–11.2	3.6–70.9	2.2–9.2	16.5–36.3
6	0–5	4.9–19.5	4.0–54.5	2.9–9.4	34.3–67.4
	5–10	2.5–6.1	2.8–43.5	2.1–5.3	22.7–69.4

† The periods of study during Exp. 4, 5, and 6 were from 31 May 1995, 17 July 1995, and 4 Sept. 1995, respectively, to 3 Nov. 1995.

‡ Total pore space % = 69, bulk density = 0.82 g cm<sup>-3</sup>, and particle density = 2.65 g cm<sup>-3</sup>.

to 5 cm and 5 to 10 cm depths measured after the application of dung during Exp. 4, 5, and 6 (where data were available for the whole experimental period) did not increase substantially over those in the control plots until approximately 10 d and then remained higher afterwards. Generally, concentrations of NH<sub>4</sub><sup>+</sup> were higher than NO<sub>3</sub><sup>-</sup> and the total mineral N (NH<sub>4</sub><sup>+</sup> + NO<sub>3</sub><sup>-</sup>) content was higher at the top 0 to 5 cm soil layer (Table 2). The total N concentrations measured during all the experiments ranged from 4.9 to 27.1 mg N kg<sup>-1</sup> dry soil in the 0 to 5 cm layer and from 2.4 to 11.2 mg N kg<sup>-1</sup> dry soil at 5 to 10 cm. The corresponding concentrations measured in the control plots ranged from 2.5 to 14.0 and 1.7 to 9.2 mg N kg<sup>-1</sup> dry soil, respectively. More details of the soil available N concentrations and patterns of change during the experiments are given by Yamulki et al., 1998. Linear regression analysis between CH<sub>4</sub> fluxes and the measured soil variables in the two soil layers, during each experiment, did not show significant correlations. Correlations between CH<sub>4</sub> fluxes and meteorological factors during each experiment were also generally weak and although in some of the experiments there were significant positive correlations between CH<sub>4</sub> fluxes and some of the variables, this was not always the case: these differences could not be easily explained. There were, however, significant positive correlations between CH<sub>4</sub> fluxes and rainfall when dung was applied during autumn in Exp. 2, 3, and 6 ( $P < 0.05$ ).

### Methane Fluxes from the Urine Patches

During each experiment, both emission and deposition of CH<sub>4</sub> were observed throughout the measurement

period (see example in Fig. 2b). In most of the experiments, exchanges of CH<sub>4</sub> were not affected significantly by urine, and generally there was no peak in CH<sub>4</sub> flux after application. One exception was during Exp. 3 when a peak CH<sub>4</sub> emission of 58.2 µg C m<sup>-2</sup> h<sup>-1</sup> was observed immediately after urine application. Typically, CH<sub>4</sub> fluxes ranged between -20 and 10 µg C m<sup>-2</sup> h<sup>-1</sup> from the urine patches and the control plots, during all the experiments. Approximately 56% of the flux data implied deposition rather than emission of CH<sub>4</sub> from both the urine patches and the control plots.

Total soil N measured within the first 48 h after urine application during the experiments ranged between 54.5 and 160.5 mg N kg<sup>-1</sup> dry soil, respectively, in the top 0 to 5 cm soil (Table 2). The corresponding concentrations at the 5 to 10 cm layer were 29.3 and 70.9 mg N kg<sup>-1</sup>. The total available N in the soil decreased sharply thereafter and reached background contents approximately 15 d after applications. Generally, in all the experiments there was a higher content of soil-available NH<sub>4</sub><sup>+</sup> than of NO<sub>3</sub><sup>-</sup> and higher total N in the 0 to 5 cm soil layer than at 5 to 10 cm. Linear regression analysis between CH<sub>4</sub> fluxes and soil variables in the two soil layers during each experiment did not show any significant relationships. Similarly, only weak correlations were observed between CH<sub>4</sub> fluxes and rainfall and ambient temperature. However, correlations were observed between CH<sub>4</sub> fluxes from the urine plots and CH<sub>4</sub> fluxes from the control plots in all the experiments ( $P < 0.01$ ). Generally fluxes of CH<sub>4</sub> from the urine patches were negligible compared to those from the dung patches, and a clear seasonal variation in the exchange pattern of CH<sub>4</sub> was not observed.

### Cumulative Methane Fluxes from Excreta

Fluxes of CH<sub>4</sub> from the dung and urine patches during Exp. 1 to 6 were measured for periods of 417, 368, 367, 154, 100, and 60 d, respectively. However, to enable a comparison between the different experiments, the total cumulative flux (estimated from the area under the flux curves less that from the control) was calculated over 60 d after application (Table 3). The total cumulative CH<sub>4</sub> flux from the dung ranged between 12.6 to 87.7 mg C patch<sup>-1</sup> (100–696 mg C m<sup>-2</sup>), and from the urine between -0.47 to 1.5 mg C patch<sup>-1</sup> (-3.8 to 12.1 mg C m<sup>-2</sup>), assuming an average affected area of 0.126 m<sup>2</sup> in

**Table 3.** Summary of the integrated CH<sub>4</sub> fluxes and estimation of annual emissions from excreta return by grazing animals during each experiment.

Experimental run	Date of treatment application	Total cumulative flux† mg CH <sub>4</sub> -C per patch		Annual flux g CH <sub>4</sub> -C per animal			Estimated UK‡ emissions from excreta kt CH <sub>4</sub> -C yr <sup>-1</sup>
		Dung	Urine	Dung	Urine	Total	
1	01-09-94	30.6 ± 2.2	1.5 ± 0.3	91.9	27.4	119.3	1.42
2	16-09-94	53.9 ± 15.4	-0.5 ± 0.3	161.8	-8.5	153.3	1.82
3	21-10-94	87.7 ± 14.1	0.6 ± 0.3	263.1	11.1	274.2	3.26
4	31-05-95	46.6 ± 1.6	-0.2 ± 0.2	139.7	-4.2	135.5	1.61
5	17-06-95	12.6 ± 0.4	-0.1 ± 0.2	37.8	-2.6	35.2	0.42
6	04-09-95	25.6 ± 2.7	-0.1 ± 0.2	76.8	-1.8	75.1	0.89
Mean		42.8 ± 6.1	0.2 ± 0.2	128.5	3.6	132.1	1.57

† Calculated on a 60 d basis, ± SE.

‡ Assuming that observations made during a particular experimental run apply to complete grazing season.

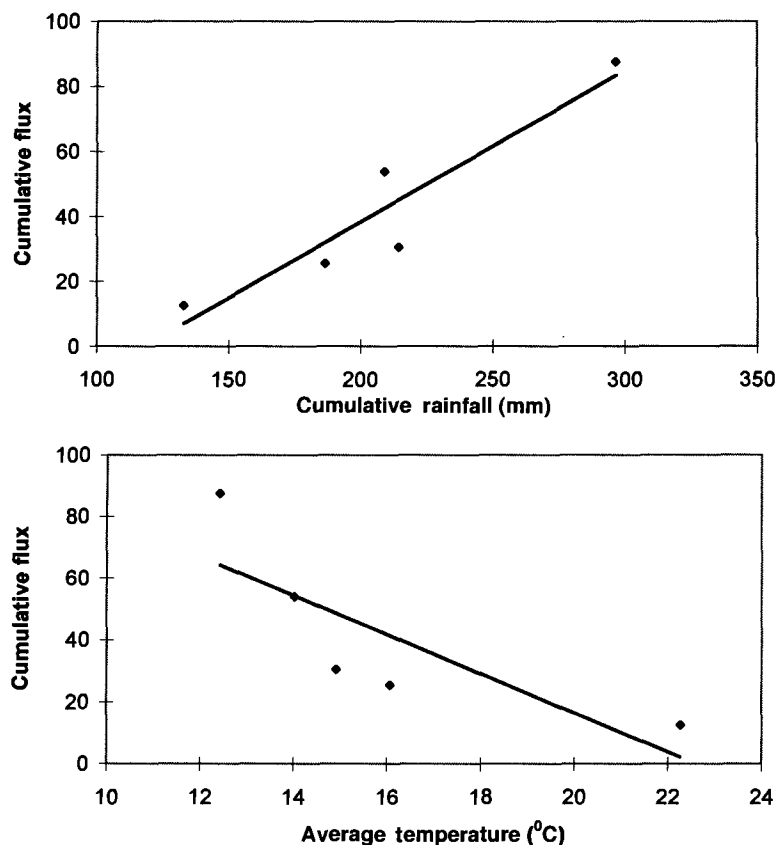


Fig. 3. The effect of ambient temperature and rainfall on CH<sub>4</sub> emissions (mg CH<sub>4</sub>-C patch<sup>-1</sup>) from the dung pats during each experiment. Lines represent regression lines.

the field (Allen et al., 1996). The overall mean of CH<sub>4</sub> flux from the dung for all the experiments was 42.8 mg patch<sup>-1</sup>. For urine, however, the cumulative flux calculated for each experiment showed both emissions and uptakes with a negligible net CH<sub>4</sub> emission of 0.2 mg C patch<sup>-1</sup>. Taking account of CH<sub>4</sub> exchange over a longer period had little effect on the cumulative flux: for example, emissions for Day 61 to 417 in Exp. 1 were equivalent to a further 2 and -0.7 mg C patch<sup>-1</sup> (15.8 and -5.4 mg C m<sup>-2</sup>), for dung and urine, respectively. The net exchange with the background site (control plot) was equivalent to -4.48 mg C m<sup>-2</sup> on average over the 60 d periods.

The data showed that the overall cumulative flux of CH<sub>4</sub>-C from the dung patches for the 60 d observation during all the experiments, with the exception of that during Exp. 4, was correlated (Fig. 3) with the corresponding cumulative rainfall ( $r^2 = 0.896$ ,  $P < 0.01$ ) and average air temperature ( $r^2 = 0.659$ ,  $P < 0.1$ ). During Exp. 4, the specific climatic conditions, that is, dry with an average temperature of 20°C and a total cumulative rainfall of only 53 mm observed during the 60 d of observation, may have been the reason for this divergence and that other factors might have controlled over the CH<sub>4</sub> flux. Other significant correlations between both meteorological and soil factors and fluxes of CH<sub>4</sub> during each experiment from the dung and urine patches were not observed. This might be due to the overriding interactions that occur between both meteorological and soil factors and the controls over fluxes of CH<sub>4</sub>.

## DISCUSSION

### Dung

The fact that a significant CH<sub>4</sub> emission was observed from all the experiments immediately after dung application and that emission rates declined generally within the first 48 h after application implies that most of the emitted CH<sub>4</sub> came from the dung itself. This is not surprising as freshly voided dung patches have potential for CH<sub>4</sub> production, that is, large microbial population, readily available C supply (total C was 43% dry wt., Table 1) and a relatively high moisture content (85% fresh wt., Table 1) and temperature (Jarvis et al., 1995). Furthermore, anaerobic conditions seem to build up almost immediately in the fresh dung patches after deposition (Holter, 1997). These conditions and the fact that CH<sub>4</sub> production by methanogenic bacteria is limited to strictly anaerobic conditions (Tiedje et al., 1984; Cicerone and Oremland, 1988), will stimulate the production of CH<sub>4</sub>. During all the experiments, more than 80% of the total CH<sub>4</sub> was emitted within the first week after application. During this period metabolization of the organic matter in the dung is expected to be low, because of the anoxic conditions present (Holter and Hendriksen, 1988). This suggests that movement of materials and interactions between the dung and the soil in influencing CH<sub>4</sub> was relatively small. Similar results were also indicated by Jarvis et al., (1995). This might explain why no correlations were observed between CH<sub>4</sub> fluxes and soil moisture. During grazing, variation in the emission

rate of  $\text{CH}_4$  from individual dung patches is expected to be very large ranging from zero to more than  $100 \mu\text{g min}^{-1}$  (Williams, 1993), and the total cumulative flux varied significantly ( $P < 0.001$ ) between the experiments in the present study. One reason for large variations in  $\text{CH}_4$  emissions in the field could be due to variation in the C/N ratio of dung patches as the result of varying diets (Jarvis et al., 1995). However, during all the experiments in this study, dung was collected from cows that had similar dietary intake (C/N and moisture content) and was applied to the soil at similar rates. Therefore, the variation in the total cumulative flux should only be attributed to meteorological factors, particularly temperature and rain by influencing anaerobic conditions and the microbial activity within the patch, respectively. The physical effect of rainfall can also significantly increase emissions by degassing the  $\text{CH}_4$  trapped within the patch. However, high temperatures could stimulate early formation of crust, reducing  $\text{CH}_4$  volatilization from the surface thus increasing the potential for  $\text{CH}_4$  oxidation. Similar results were also observed from slurry by Husted (1993) who indicated that the presence of surface crust reduced  $\text{CH}_4$  emission by a factor of 11 to 12. Our data showed that despite the above complexity, climatic difference between the experiments was an important factor affecting the total  $\text{CH}_4$  emissions from the dung. Higher cumulative fluxes of  $\text{CH}_4$  were observed during the experiments with relatively higher rainfall and lower ambient temperature.

### Urine

Estimates of the total cumulative flux of  $\text{CH}_4$  measured from the urine patches showed large variation between the experiments. While the total cumulative flux showed a small net emission of  $\text{CH}_4$  during Exp. 1 and 3, an uptake of  $\text{CH}_4$  was observed from the other experiments. Recently, Flessa et al. (1996) reported that urine affected areas were sinks for  $\text{CH}_4$  showing slightly higher  $\text{CH}_4$  uptake rates compared with the soil not influenced by excreta. In the present study,  $\text{CH}_4$  uptake was generally observed when urine was applied during a relatively high temperature and low rainfall period (Exp. 4, 5, and 6) and emission during periods of high rainfall and low temperature (Exp. 1 and 3). Whalen and Reeburgh (1996) have recently found a similar combination effect of both temperature and moisture on  $\text{CH}_4$  fluxes from laboratory experiments. The effect of moisture in reducing  $\text{CH}_4$  uptake is generally attributed to the more anaerobic conditions which favor  $\text{CH}_4$  production and to the slower diffusion of  $\text{CH}_4$  to the surface of methanotrophic bacterial cells, reducing the overall oxidation. High temperatures, while increasing the biological activity and  $\text{CH}_4$  production would also cause physiological changes to the methanotrophs, resulting in higher oxidation rates (Boeckx et al., 1996). During Exp. 2, the total cumulative flux of  $\text{CH}_4$  was reduced markedly despite a heavy precipitation of 46 mm deposited in the first week after application of urine, followed by drier conditions (8.5 mm rain) and a relatively higher temperature during the following 26 d. The heavy pre-

cipitation immediately after the urine application might remove the essential substrates for methanogenesis by washing them into the ground (Jarvis et al., 1995), therefore reducing  $\text{CH}_4$  emissions.

During all the experiments, the flux pattern and the uptake or emission rates of  $\text{CH}_4$  were generally similar in both the urine and the control plots. This is clearly demonstrated by the significant ( $P < 0.01$ ) correlation observed between  $\text{CH}_4$  fluxes measured from the urine and the control plots during each experiment. This indicates that environmental factors had similar effects on  $\text{CH}_4$  fluxes from the urine and the control plots and that the large inputs of  $\text{NH}_4^+$  to the soil in the urine generally did not have any specific effect on  $\text{CH}_4$  fluxes. Similar results have also been observed from other studies with urine (Jarvis et al., 1995; Flessa et al., 1996), urea- $\text{NH}_4\text{NO}_3$  fertilizer (Flessa et al., 1995) and from cattle slurry (Hansen et al., 1993). However, other studies have shown that the application of inorganic N either from the atmosphere (e.g., Melillo, 1989) or from fertilizer suppresses  $\text{CH}_4$  oxidation rate in soils (e.g., Sitaula et al., 1995; Hansen et al., 1993; Hütsch, 1996). This reduction has been attributed to the specific effect of  $\text{NH}_3$  monooxygenase of nitrifiers competing with the  $\text{CH}_4$  monooxygenase of methanotrophs for the same active site on the monooxygenases (Knowles, 1993). Bronson and Mosier (1993) and, more recently, Dobbie and Smith (1996) have indicated that only the initial application of N fertilizer will have an inhibitory effect on  $\text{CH}_4$  oxidation rate and any further addition does not inhibit the process further. At the start of the present experiment, the soil mineral N measured from the control plot showed relatively high concentrations (up to  $15 \text{ mg NH}_4 + \text{NO}_3 \text{ kg}^{-1}$  soil) and may have similarly limited the inhibitory effect of urine on  $\text{CH}_4$  oxidation.

## CONCLUSIONS

### Annual Methane Fluxes from Excreta

The total cumulative flux of  $\text{CH}_4$  for the dung and urine patches had mean values of  $42.8$  and  $0.2 \text{ mg CH}_4\text{-C patch}^{-1}$ , respectively. Similar rates have also been measured by Jarvis et al. (1995) from dung of dairy cows, when compared on an equivalent average area, with  $\text{CH}_4$  emission ranging between  $17$  to  $48 \text{ mg CH}_4\text{-C patch}^{-1}$  with an overall mean of  $23.1$ , however, no effect of urine was noted. Although large fluxes of  $\text{CH}_4$  were observed, the highest rate of emission from the dung patch ( $87.7 \text{ mg CH}_4\text{-C}$ ) and from the urine ( $1.52 \text{ mg CH}_4\text{-C}$ ) represented only  $0.1$  and  $0.05\%$  of the total C added in the dung and urine. Based on the assumption of a daily deposition per cow of  $2 \text{ kg}$  of dung and  $2 \text{ kg}$  of urine on each of  $10$  occasions (Lantinga et al., 1987; Marsh and Campling, 1970), the daily  $\text{CH}_4$  fluxes from the dung and urine patches can be estimated. The  $\text{CH}_4$  fluxes as estimated for  $60 \text{ d}$  from the different experiments varied between  $0.21$  to  $1.46$  (mean =  $0.72$ )  $\text{g CH}_4\text{-C per animal per day}$  from the dung. The corresponding fluxes from the urine patches ranged between  $-0.047$  to  $0.152$  (mean =  $0.02$ )  $\text{g CH}_4\text{-C per animal per day}$ . Flessa et al. (1996) have also measured  $\text{CH}_4$

emissions from dung in Germany over 78 d and estimated a daily emission of 0.778 g CH<sub>4</sub>-C per animal, that is, similar to our mean estimates. If cattle are grazing for 180 d per year, then the total annual flux of CH<sub>4</sub> will range between 37.8 and 263 (mean = 129) g CH<sub>4</sub>-C from the dung and -8.46 to 27.4 (mean = 3.6) g CH<sub>4</sub>-C from the urine (Table 3) (assuming that all cattle behave in the same way). Earlier estimates by Jarvis et al. (1995) gave an annual mean value of 165 g CH<sub>4</sub>-C per cow when excreta were applied to the same soil as in the present work. Recently Holter (1997) has also estimated the total amount of CH<sub>4</sub> emitted from cow dung in Denmark from three experimental periods and gave annual estimates (based on the assumptions made in our study) which ranged between 92 and 302 g CH<sub>4</sub>-C per cow. On a national scale, it can be estimated that the total annual output of CH<sub>4</sub> from excretal return from grazing cattle ranges between 0.42 and 3.26 kt CH<sub>4</sub>-C (mean = 1.57) assuming a total population of 11.9 M cattle in the UK (MAFF, 1996).

The large range in emission with a factor of approximately seven can be attributed, as suggested by Holter (1997), to climatic differences at the time of deposition of the dung. At the upper end of the range (3.26 kt CH<sub>4</sub>-C), this source of CH<sub>4</sub> emission is greater than the total annual CH<sub>4</sub>-C emission from slurry of 1.53 kt in the UK (Sneath et al., 1997). However, our results also confirm other studies (Jarvis et al., 1995; Flessa, et al., 1996) that this excretal source is insignificant compared to the enteric source within the rumen of cattle in the UK (about 229.5 kt CH<sub>4</sub>-C; Sneath et al., 1997).

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#### REFERENCES

- Allen, A.G., S.C. Jarvis, and D.M. Headon. 1996. Nitrous oxide emissions from soils due to inputs of nitrogen from excreta return by livestock on grazed grassland in the UK. *Soil Biol. Biochem.* 28:597-607.
- Boeckx, P., O. Van Cleemput, and I. Villaralvo. 1996. Methane emission from a landfill and the methane oxidising capacity of its covering soil. *Soil Biol. Biochem.* 28:1397-1405.
- Bronson, K.F., and A.R. Mosier. 1993. Nitrous oxide emissions and CH<sub>4</sub> consumption in wheat and corn-cropped systems in northeastern Colorado. p. 133-144. *In* L.A. Harper et al. (ed.) *Agricultural ecosystem effects on trace gases and global climate change*. ASA Spec. Publ. 55. ASA, CSSA, and SSSA, Madison, WI.
- Cicerone, R.J., and R.S. Oremland. 1988. Biogeochemical aspects of atmospheric methane. *Global Biogeochem. Cycles* 2:299-327.
- Crutzen, P.J. 1991. Methane sinks and sources. *Nature (London)* 350:380-381.
- Dobbie, K.E., and K.A. Smith. 1996. Comparison of CH<sub>4</sub> oxidation rates in woodland, arable and set aside soils. *Soil Biol. Biochem.* 28:1357-1365.
- Flessa, H., P. Dörsch, and F. Beese. 1995. Seasonal variation of N<sub>2</sub>O and CH<sub>4</sub> fluxes in differently managed arable soils in southern Germany. *J. Geophys. Res.* 100(D11):23115-23124.
- Flessa, H., P. Dörsch, F. Beese, H. König, and A.F. Bouwman. 1996. Influence of cattle wastes on nitrous oxide and methane fluxes in pasture land. *J. Environ. Qual.* 25:1366-1370.
- Hansen, S., J.E. Mæhlum, and L.R. Bakken. 1993. N<sub>2</sub>O and CH<sub>4</sub> fluxes in soil influenced by fertilization and tractor traffic. *Soil Biol. Biochem.* 25:621-630.
- Holter, P. 1997. Methane emissions from Danish cattle dung pats in the field. *Soil Biol. Biochem.* 29:31-37.
- Holter, P., and N.B. Hendriksen. 1988. Respiratory loss and bulk export of organic matter from cattle dung pats: A field study. *Holarctic Ecol.* 11:81-86.
- Husted, S. 1993. Seasonal variation in methane emission from stored slurry and solid manures. *J. Environ. Qual.* 23:585-592.
- Hütsch, B.W. 1996. Methane oxidation in soils of two long-term fertilization experiments in Germany. *Soil Biol. Biochem.* 28:773-782.
- Jarvis, S.C., R.D. Lovell, and R. Panayides. 1995. Pattern of methane emission from excreta of grazing animals. *Soil Biol. Biochem.* 27:1581-1588.
- Kamphake, L.J., S.A. Hannah, and J.M. Cohen. 1967. Automated analysis for nitrate by hydrazine reduction. *Water Res.* 1:205-216.
- Knowles, R. 1993. Methane processes of production and consumption. p. 145-156. *In* L.A. Harper et al. (ed.) *Agricultural ecosystem effects on trace gases and global climate change*. ASA Spec. Publ. 55. ASA, CSSA, and SSSA, Madison, WI.
- Lantinga, E.A., J.A. Keuning, J. Groenwold, and P.J.A.G. Deenen. 1987. Distribution of excreted nitrogen by grazing cattle and its effects on sward quality, herbage production and utilization. p. 103-117. *In* H.G. Van der Meer et al. (ed.) *Animal manure on grassland and fodder crops*. Martinus Nijhoff Publ., Dordrecht, the Netherlands.
- Lockyer, D.R., and S.C. Jarvis. 1995. The measurement of methane losses from grazing animals. *Environ. Pollut.* 90:383-390.
- Ministry of Agriculture, Fisheries, and Food. 1996. The digest of agricultural census statistics. MAFF, the Stationery Office, London.
- Marsh, R., and R.C. Campling. 1970. Fouling of pastures by dung. *Herb. Abstr.* 40:123-130.
- Melillo, J.M., P.A. Steudler, J.D. Aber, and R.D. Bowden. 1989. Atmospheric deposition and nutrient cycling. p. 263-280. *In* M.O. Andreae and D.S. Schimel (ed.) *Exchange of trace gases between terrestrial ecosystems and the atmosphere*. John Wiley & Sons, New York.
- Searle, P.L. 1984. The Berthelot or Indophenol reaction and its use in the analytical chemistry of nitrogen. *Analyst* 109:549-565.
- Sitaula, B.K., L.R. Bakken, and G. Abrahamsen. 1995. CH<sub>4</sub> uptake by temperate forest soil: Effect of N input and soil acidification. *Soil Biol. Biochem.* 27:871-880.
- Sneath, R.W., D.R. Chadwick, V.R. Phillips, and B.F. Pain. 1997. A UK inventory of methane emissions from farmed livestock. Report submitted to MAFF (Commissions WA 0604 and WA 0605). May 1997. Silsoe Res. Inst., UK.
- Steed, J., and A.G. Hashimoto. 1994. Methane emissions from typical manure management systems. *Bioresour. Technol.* 50:123-130.
- Tiedje, J.M., A.J. Sexstone, T.B. Parkin, N.P. Revsbech, and D.R. Shelton. 1984. Anaerobic processes in soil. *Plant Soil* 76:197-212.
- Whalen, S.C., and W.S. Reeburgh. 1996. Moisture and temperature sensitivity of CH<sub>4</sub> oxidation in Boreal soils. *Soil Biol. Biochem.* 28:1271-1281.
- Williams, D.J. 1993. Methane emissions from manure of free-range dairy cows. *Chemosphere* 26:179-187.
- Yamulki, S., S.C. Jarvis, and P. Owen. 1998. Nitrous oxide emissions from excreta applied in a simulated grazing pattern. *Soil Biol. Biochem.* 30:491-500.