PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Measurements of CH$_{4}$ and N$_{2}$O Fluxes at the Landscape Scale Using Micrometeorological Methods

D. Fowler, K. J. Hargreaves, U. Skiba, R. Milne, M. S. Zahniser, J. B. Moncrieff, I. J. Beverland, M. W. Gallagher, P. Ineson, J. Garland and C. Johnson

Phil. Trans. R. Soc. Lond. A 1995 **351**, 339-356
doi: 10.1098/rsta.1995.0038

Measurements of CH_4 and N_2O fluxes at the landscape scale using micrometeorological methods

By D. Fowler[1], K. J. Hargreaves[1], U. Skiba[1], R. Milne[1], M. S. Zahniser[2], J. B. Moncrieff[3], I. J. Beverland[3] and M. W. Gallagher[4]

[1]*Institute of Terrestrial Ecology, Bush Estate, Penicuik, Midlothian EH26 0QB, U.K.*

[2]*Aerodyne Research, Inc., 45 Manning Road, Billerica, Massachusetts 01821, U.S.A.*

[3]*University of Edinburgh, Institute of Ecology and Resource Management, Darwin Building, Kings Buildings, Mayfield Road, Edinburgh EH9 3JU, U.K.*

[4]*University of Manchester, Institute of Science and Technology, P.O. Box 88, Manchester M60 1QD, U.K.*

Flux gradient, eddy covariance and relaxed eddy accumulation methods were applied to measure CH_4 and N_2O emissions from peatlands and arable land respectively. Measurements of N_2O emission by eddy covariance using tunable diode laser spectroscopy provided fluxes ranging from 2 to 60 μmol N_2O m^{-2} h^{-1} with a mean value of 22 μmol N_2O m^{-2} h^{-1} from 320 h of continuous measurements.

Fluxes of CH_4 measured above peatland in Caithness (U.K.) during May and June 1993 by eddy covariance and relaxed eddy accumulation methods were in the range 70 to 120 μmol CH_4 m^{-2} h^{-1} with means of 14.7 μmol CH_4 m^{-2} h^{-1} and 22.7 μmol CH_4 m^{-2} h^{-1} respectively.

Emissions of CH_4 from peatland changed with water table depth and soil temperature; increasing from 25 μmol CH_4 m^{-2} h^{-1} at 5% pool area to 50 μmol CH_4 m^{-2} h^{-1} with 30% within the flux footprint occupied by pools. A temperature response of 4.9 μmol CH_4 m^{-2} h^{-1} $^{\circ}C^{-1}$ in the range 6–12 $^{\circ}$C was also observed.

The close similarity in average CH_4 emission fluxes reported for wetlands in Caithness, Hudson Bay and Alaska in the range 11 to 40 μmol CH_4 m^{-2} h^{-1} suggests that earlier estimates of CH_4 emission from high latitude wetlands were too large or that the area of high latitudes contributing to CH_4 emission has been seriously underestimated.

1. Introduction

The presence of CH_4 in the atmosphere was identified in 1948 by Migeotte (1948) and in the following three decades concentrations were considered to be approximately constant at about 2 ppmv (Khalil 1993). However, by the early 1980s measurements were available to show that global CH_4 concentrations were increasing rapidly (Rasmussen & Khalil 1981). It was also recognized that in addition to the

Phil. Trans. R. Soc. Lond. A (1995) **351**, 339–356
Printed in Great Britain

role of CH_4 as an important radiatively active gas, second only to CO_2, atmospheric CH_4 plays a major role in the tropospheric chemistry of O_3 and OH and as a source of H_2 and H_2O in the stratosphere (Ehhalt 1974). The ambient concentrations during the period 1000–1800 AD were about 700 ppbv but increased almost exponentially between 1800 and the present day to a concentration of about 1750 ppbv (Raynaud *et al.* 1988). A much longer record from Greenland and Antarctic ice cores (Stauffer *et al.* 1988; Raynaud *et al.* 1988, respectively) show that during the last glacial–interglacial climatic cycle the atmospheric concentration of CH_4 was closely coupled to global temperature and this has directed attention towards the two major natural components of the atmospheric CH_4 cycle – emissions from natural wetlands and atmospheric oxidation by OH radicals.

Natural wetlands are believed to contribute 100 ± 30 Tg CH_4 to the annual atmospheric budget, approximately 20% of all sources and a much larger fraction of natural sources (Matthews 1993). The peat wetland source areas, primarily located at high latitudes in the Northern Hemisphere between 50° and 80° N have been estimated to occupy between 3 and 9.0×10^{12} m^2 (Sebacher *et al.* 1986; Matthews & Fung 1987).

Rates of CH_4 emission from peatlands have been shown to increase by a factor of 3 or more over the temperature range 5–15 °C (Bartlett *et al.* 1992). As the temperature increases resulting from radiative forcing over the next 50 years are predicted to be largest at high latitudes (IPCC 1992), it is possible that emission from natural wetland sources of CH_4 in these regions may increase substantially.

The emission of CH_4 from wetlands shows very large spatial and temporal variability as a result of interactions between production and oxidation processes and the conditions which regulate them, especially the redox potential, water table and temperature (Harriss *et al.* 1985; Wahlen & Reeburgh 1988; Morrisey & Livingston 1992). The extrapolation of fluxes to landscape and regional scales from chamber measurements has therefore been a speculative exercise (Matthews 1993) and the choice of appropriate values to characterise annual emission fluxes from wetland areas for global modelling of the CH_4 budget appears rather arbitrary.

Nitrous oxide (N_2O) in the atmosphere, in addition to contributing to radiative forcing is also involved in the chemistry of the stratosphere where it represents a major source of nitrogen oxides which are important in regulating stratospheric ozone. Concentrations in the atmosphere have been increasing throughout the past four decades and current global average concentrations of about 310 ppbv exceed the pre-industrial concentration (285 ppbv) by about 9%, the annual increase being between 0.2 and 0.3% (IPCC 1992). The sources of N_2O appear to be dominated by soil emission following biological denitrification but significant emissions are also contributed by oceans and industrial processes (Bouwman 1990). The spatial variability in N_2O emission from arable land is as large or larger than that for CH_4 for wetlands, with differences in emission of several orders of magnitude reported over a transect of 20–30 m in arable soils (Smith *et al.* 1994).

2. Application of micrometeorology

Micrometeorological methods have been applied to bridge the gap between the scales over which fluxes are required, and the scale over which most measurements have been made. However, for the radiatively active gases CH_4 and N_2O, micrometeorological methods have been applied only recently. The Arc-

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

tic Boundary Layer Expedition (ABLE 3A) during July and August 1988, provided the first micrometeorological measurements of CH_4 emission fluxes over sub-Arctic tundra (Fan *et al.* 1992). Fluxes measured using the eddy covariance method provided approximately 100 h of data. The mean emission rate for the area surrounding the instrument tower was 65 μmol CH_4 m^{-2} h^{-1} with larger emission from the wet meadow tundra (75 μmol CH_4 m^{-2} d^{-1}) than the areas of dry tundra (28 μmol CH_4 m^{-2} h^{-1}). A study of CH_4 emission over the Hudson Bay lowlands as part of the Canadian Northern Wetlands Study (NOWES) also used eddy covariance methods (Edwards *et al.* 1994). The average emission was 41 μmol CH_4 m^{-2} h^{-1} with daytime exceeding nighttime fluxes by 25%.

The application of micrometeorology to provide large area estimates of the emission of N_2O from soils has been reported by Hargreaves *et al.* (1994) from a collaborative study by a range of chamber and micrometeorological methods at a grassland site in the central lowlands of Scotland (Smith *et al.* 1994).

In this paper, the methods and instrumentation to measure CH_4 and N_2O emission fluxes at the field scale by micrometeorology are briefly described. We report measurements of CH_4 fluxes above a blanket bog in the flow country of northern Scotland and N_2O fluxes over arable land in Denmark. The degree to which the measurements can be used to gain an understanding of the processes which regulate emissions is also considered using measurements of CH_4 emission from peatlands.

3. Flux gradient methods

The vertical transport of a gas towards an absorbing surface may be written as

$$F_\chi = -\rho K_\chi \frac{\partial \chi}{\partial z}, \tag{3.1}$$

where K_χ is the transfer (diffusion) coefficient for the gas a and $\partial\chi/\partial z$ is the vertical gradient in concentration in the constant flux layer. By convention a flux towards the surface is negative and vice versa. The diffusion processes within the atmosphere are dominated by turbulent exchange: K_χ, the turbulent exchange coefficient is assumed identical for all gases and sensible heat but in unstable conditions differs from that for momentum K_m (Thom 1975). The application of flux gradient aerodynamic methods in which K_χ is provided by wind velocity and temperature profiles, and corrections for stability effects are described by Fowler & Duyzer (1989).

Flux-profile relationships have been shown to provide satisfactory methods of measuring the net exchange of momentum, sensible and latent heat flux and a range of trace gases over short vegetation. However, over tall vegetation and forests in particular, flux-profile relationships are more complex (Raupach 1989). These complexities may lead to large systematic differences in measured fluxes using different methods over forests if general flux-profile relationships are used (Thom *et al.* 1975). In such cases, it is still possible to obtain fluxes from profiles, but it is necessary to establish the form of the flux-profile function at the site of the study. This is most readily achieved by measuring the sensible heat flux directly using an eddy covariance method, establishing the flux-profile function for sensible heat and, making the assumption of similarity of transfer and sources and sinks for sensible heat and trace gases. Site-specific flux gradient functions

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

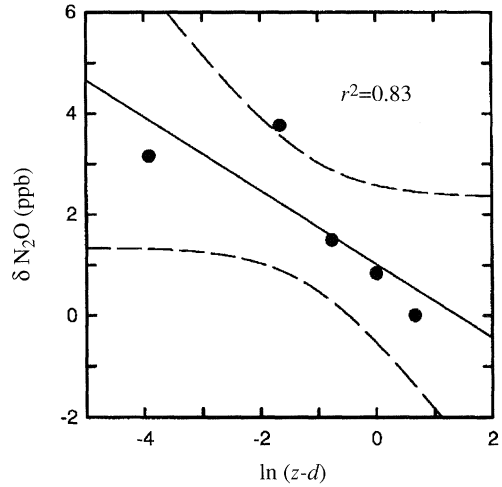


Figure 1. Example of N_2O concentration profile measured above fertilized grassland during the early afternoon. N_2O flux = 18 μmol m^{-2} h^{-1}), calculated using the aerodynamic method (from Hargreaves *et al.* 1994). z is the height above surface (m), d is the zero plane displacement (m), δN_2O is the deviation of N_2O concentration from ambient (ppb).

may then be applied for the trace gas of interest. This approach has recently been applied to measure NH_3 exchange over a forest in the Netherlands (Duyzer *et al.* 1994).

Alternatives to the use of wind and temperature profiles to obtain the eddy diffusivity are provided by the energy balance or Bowen ratio method (Monteith & Unsworth 1990). The application of these methods for the measurement of CH_4 and N_2O is, in principle, straightforward. In practice, there are few sensors for these gases capable of detecting the very small gradients in concentration in the large background. Taking typical emission fluxes from the literature of 22 μmol CH_4 m^{-2} h^{-1} and 2 μmol N_2O m^{-2} h^{-1} over wetland and agricultural grassland respectively, the expected gradients in the lowest 3 m, are typically 3 μg CH_4 m^{-3} and 5 μg N_2O m^{-3}. These concentration differences represent between 0.2 and 1.0% of the ambient concentrations. For such measurements an important correction, for the effects of heat and water vapour on the measured gradients of CH_4 and N_2O arises if the air sampled is not dried and brought to a common temperature (Webb *et al.* 1980). The error in the flux estimated is large for any trace gas flux that is small relative to its ambient concentration and may exceed 100% for N_2O fluxes.

Both CH_4 and N_2O fluxes have been measured using flux gradient methods. Measurements of N_2O concentration gradients were made over a height range of 3.0 m above a recently fertilized grassland in the Stirling experiment (Smith *et al.* 1994). An example of the vertical profile is shown in figure 1. Also included on the figure is an estimate of the emission flux for this period using the measured sensible and latent heat fluxes and an aerodynamic method. The flux of 18 μmol m^{-2} h^{-1} was within the range measured using enclosure methods at the same site (Smith *et al.* 1994).

For the measurement of CH_4 emission from wetlands, gradient methods have also successfully been applied using a flame ionization detector fitted with a catalyst to destroy all of the non-methane hydrocarbons without significantly

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

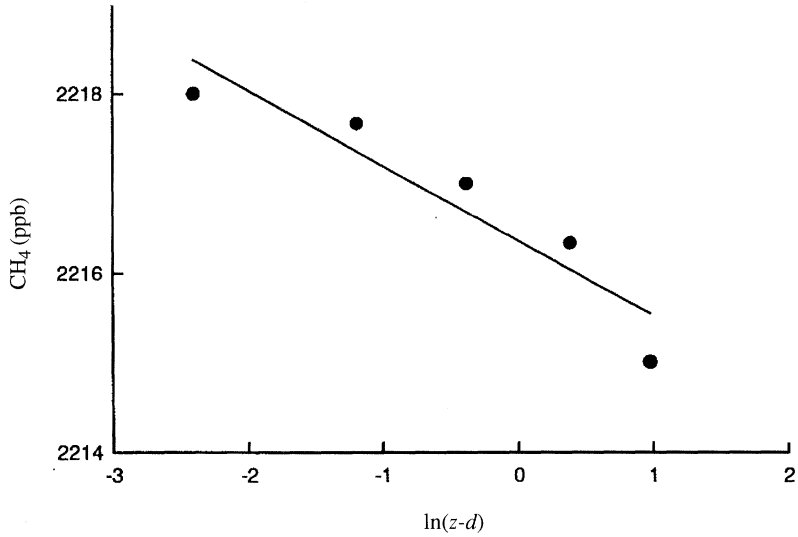


Figure 2. Example of a CH_4 concentration profile measured above peatlands in northern Scotland 02:00–02:30 GMT. CH_4 flux = 12 μmol $m^{-2}h^{-1}$, calculated using the aerodynamic method.

influencing the CH_4 signal. The vegetation at the site in Sutherland over blanket bog was dominated by *Calluna vulgaris* and *Erica* sp. on the drier (hummock) areas, by *Sphagnum* species in the pool and wet 'lawn' areas. An example of the profile data and the flux calculated are shown in figure 2.

The application of aerodynamic methods to estimate fluxes of CH_4 and N_2O is straightforward given adequate sensitivity in the gas analysis system, a suitable site and atmospheric conditions meeting uniformity and stationarity requirements.

4. Eddy covariance methods

This method, also known as 'eddy correlation' is the simplest and most elegant of the micrometeorological methods, in which the vertical flux density F_χ of a trace gas may be written as

$$F_\chi = w\rho_\chi, \tag{4.1}$$

where w is the vertical wind velocity and ρ_χ is the density of the trace gas. The term $w\rho_\chi$ may be considered as the sum of two components, the first being the product of the mean vertical windspeed (w) and mean trace gas density (ρ_χ), while the second component is the mean product of the fluctuations about the mean for the same two components (w' and ρ'_χ):

$$F_\chi = \overline{w}\,\overline{\rho_\chi} + \overline{w'\rho'_\chi}. \tag{4.2}$$

In practice, the three components of the turbulent structure are all measured and resolved in such a way that mean vertical windspeed is zero, eliminating the first term in equation (4.2). To sample the spectrum of turbulent eddies transporting the flux, the instrumentation must be capable of detecting the high frequency structure in vertical wind velocity and concentration of the trace gas, requiring a response time of 0.1 s or better.

A sonic anemometer mounted 5 m above the surface on an aluminium lattice

tower provided the three-component wind velocity and air temperature. Concentrations of CH_4 and N_2O were measured using tunable diode-laser (TDL) absorption spectroscopy (Zahniser *et al.*, this volume). The laser, operating at liquid nitrogen temperatures in the range 75–100 K and tuned to an absorption line centred on 3017.46 cm^{-1} for CH_4 and 2204.71 cm^{-1} for N_2O provided an r.m.s. noise of typically 1% and 0.1% for both CH_4 and N_2O at ambient concentrations sampling at 20 Hz and 1.0 Hz respectively.

The flux measurements were obtained from the covariance of vertical wind and concentrations with the coordinate system rotated to provide the fluctuating component of w perpendicular to the streamlines of flow as described by McMillan (1986), using the software 'Eddysol' (Moncrieff *et al.* 1994). The fluxes were calculated following the subtraction of a 20 min running mean from the 20 Hz measurements of w and χ to provide 10 min mean fluxes.

The spectral density of the variations in ambient CH_4 was calculated from blocks of 4096 points from the TDL recorded at 20 Hz. Within each hour of data, 17 consecutive blocks of this size were analysed by calculating the power spectrum. The 2048 raw spectral estimates for each data block were ensemble-averaged with the others for the hour and grouped logarithmically to provide 54 spectrum estimates ranging from 5×10^{-3} to 9.8 Hz. The spectrum for a typical hour is shown in figure 3 plotted against non-dimensional frequency, $f = nz/u$, where n is the frequency (Hz), z is the height above zero plane (m), and u is the mean horizontal wind speed. The spectrum has been normalized with respect to the variance in the methane concentration signal and scaled by multiplying by frequency (n). The spectrum decays above $f = 1$ at a slope of approximately $-2/3$ indicating that the TDL had a response fast enough to record the high frequency variations in CH_4 concentrations.

The measured eddy fluxes of methane covered approximately 600 h during May and June of 1993 and May–June 1994 over a blanket bog at Loch More in Caithness, U.K. The data (figure 4) show values approximately normally distributed about a mean of 14.7 μmol CH_4 m^{-2} h^{-1} with an s.d. of 14.6 μmol m^{-2} h^{-1} for the 1993 measurements.

Eddy covariance fluxes of N_2O were measured over a recently harvested wheat field on land close to the coast of the Danish island of Zealand during August–September 1993, using the same TDL instrument as for CH_4. Fluxes over wheat stubble were measured for 180 h between 18 and 28 August 1993.

The measurements again appear normally distributed about a mean value of 24.2 μmol N m^{-2} h^{-1} with an s.d. of 14 μmol N m^{-2} h^{-1} (figure 5).

5. Relaxed eddy accumulation (conditional sampling)

The eddy accumulation method described by Desjardins (1977), requires the detection of upward (+ve) and downward (−ve) components of vertical wind velocity w' and the sampling of trace gases associated with up- and downdraughts respectively into two reservoirs. The practical difficulties in achieving satisfactory sampling characteristics prevented the widespread application of this method, which in principle, obviates the need for rapid response gas analysis. Businger & Oncley (1989) proposed a simplification of the eddy accumulation method relaxing the requirement for sampling at rates that are proportional to w'. They proposed a measurement system in which the gases are sampled at a constant

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

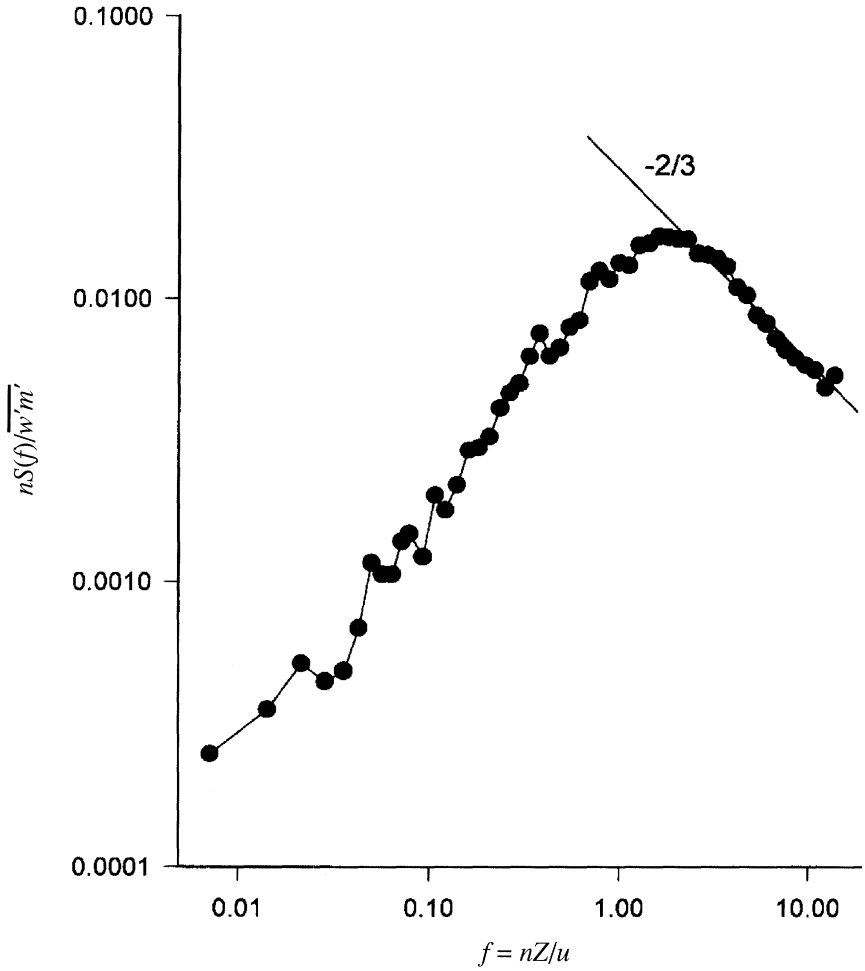


Figure 3. Spectral density of variations in the CH_4 concentration 5 m above peat wetland. u is the mean horizontal windspeed (m s^{-1}) for data block, Z is the height above surface (m), n is the frequency (Hz), f is the normalized frequency, $S(f)$ is the spectral density, $\overline{w'm'}$ mean methane flux for data block.

rate during up and down draughts which, as before, are fed to separate reservoirs during the measurement period. The flux is then provided as

$$F_\chi = \beta\sigma_w(\chi_u - \chi_d), \tag{5.1}$$

where σ_w is the standard deviation of the vertical windspeed, χ_u and χ_d are the concentrations of the trace gas within the up and down draught reservoirs and β is a dimensionless constant determined by measurement. As in the case of eddy covariance methods, the measurement system must be capable of detecting the high frequency structure in w' and the valve and gas sampling system must be capable of providing rapid switching and sampling of air during up and down draughts. In practice a time resolution of 0.1 s is required for the gas sampling system.

A relaxed eddy accumulation system developed by Beverland *et al.* (1995) was

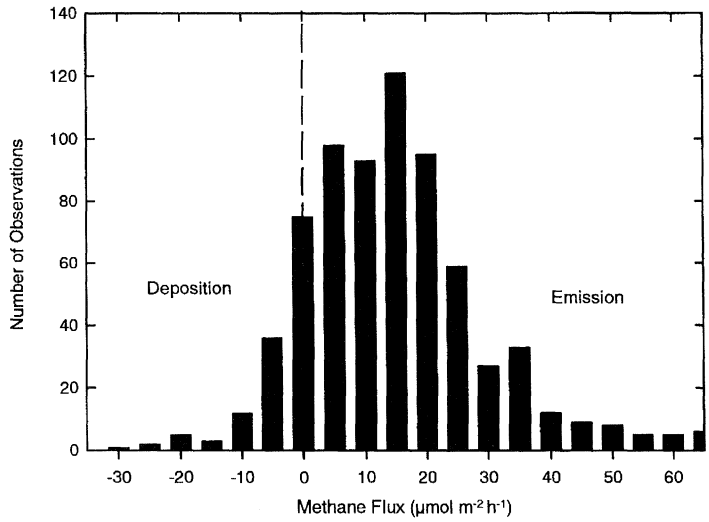


Figure 4. Frequency distribution of 300 hourly averages of CH_4 fluxes measured by eddy covariance above peatland at Loch More, Caithness, June 1993.

used to measure CH_4 exchange over the peat wetlands in Caithness at the same site as that described earlier for the eddy covariance measurements.

The 1993 study of methane emission at Loch More provided 130 hours of flux measurement, mainly during daytime giving a CH_4 mean emission flux of 22.7 μmol m^{-2} h^{-1} similar to that for eddy covariance (14.7 μmol m^{-2} h^{-1}), but with a rather larger range (-70 to $+120$ μmol CH_4 m^{-2} h^{-1}) (figure 6).

The extent to which the methods may be applied to investigate the underlying process which regulate trace gas fluxes may now be examined. For this purpose, measurements of CH_4 exchange over wetlands are used to illustrate the process.

The CH_4 emission fluxes appear similar to those reported for chamber studies. The key question is whether variability within the data may be quantitatively associated with properties of the atmosphere or wetland surface.

6. Methane emission and deposition

The data presented in figure 4, imply a very broad range in hourly fluxes with a significant proportion representing CH_4 uptake, which may be interpreted as CH_4 oxidation. However, the flux measurements 5 m above the surface are subject to flux divergence during periods when CH_4 concentrations are changing systematically during the measurement. The process leads to accumulation (or loss) of CH_4 in the layer of the atmosphere between the measurement level and the surface and can be estimated from figure 7. The flux divergence for the site conditions during the Loch More experiment is approx. 1.8 μmol CH_4 m^{-2} h^{-1} for a 10 ppb h^{-1} change in concentration (or 5% of a typical flux). Applying similar approaches to estimate the likely magnitude of flux divergence as a consequence of horizontal gradients in CH_4 concentration (advection) leads to a value of 3 μmol CH_4 m^{-2} h^{-1} (or 10% of a typical flux) for a horizontal gradient of 1 ppb km^{-1}. It may be shown that for the mean flux from several hundred hours of measurement, the storage errors cancel. However, a substantial fraction of the

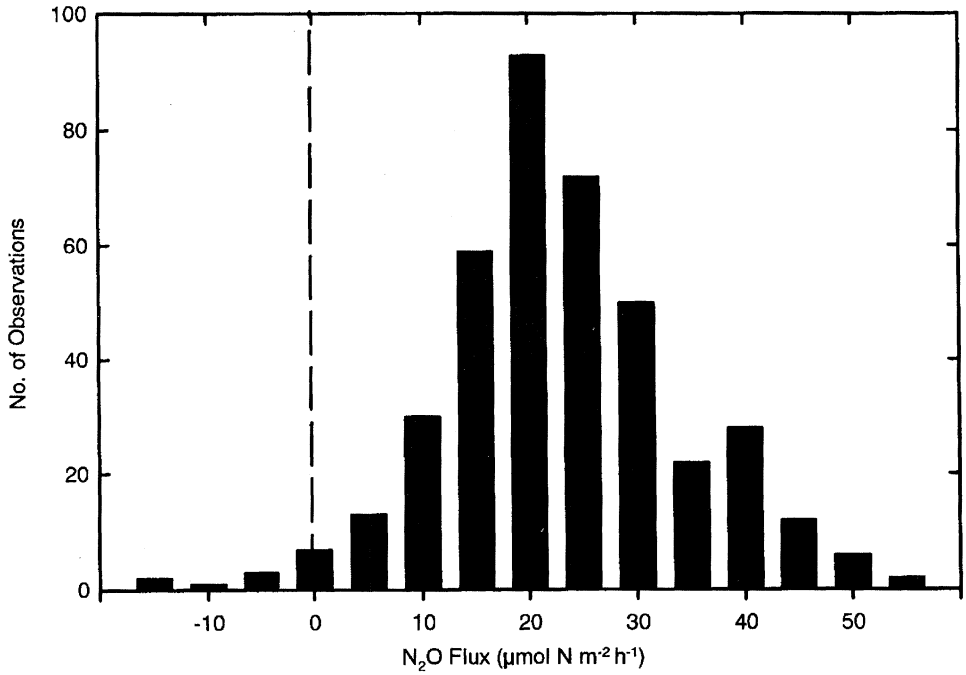


Figure 5. Frequency distribution of 200 hourly averages of N_2O fluxes measured by covariance above a recently harvested wheat field in Zealand during August 1993.

observed deposition fluxes in figure 4 and the very large emission fluxes, result from changes in storage during the measurement period.

7. Effects of water table on CH_4 emissions

Micrometeorological measurements of CH_4 fluxes over tundra by Fan *et al.* (1992) show a clear sector dependence of methane emission. For wind directions over a fetch containing the largest area of lakes and pools, fluxes at 75 μmol m^{-2} h^{-1} were a factor of two larger than those over the dry tundra with a water table classified as below 5 cm. In the measurements over blanket bog in Caithness, the flux measuring tower was placed at a boundary between a large area of pools extending through 160° to the west while to the east of the mast, the fetch was uniformly much drier with very few pools and mean water tables along 120 m transects from the mast consistently deeper than 10 cm (figure 8).

The emission flux is closely linked with the presence of pool areas as fluxes from the sectors between 240 and 360° of 35–50 μmol CH_4 m^{-2} h^{-1} are a factor of two to three larger than those over the dry easterly sectors with lower water table. The relationship between the sector mean methane flux and pool area in the sector is almost linear (figure 9), providing a very valuable tool for the extrapolation of fluxes using geographical information on the areal distribution of pools to provide landscape methane emissions.

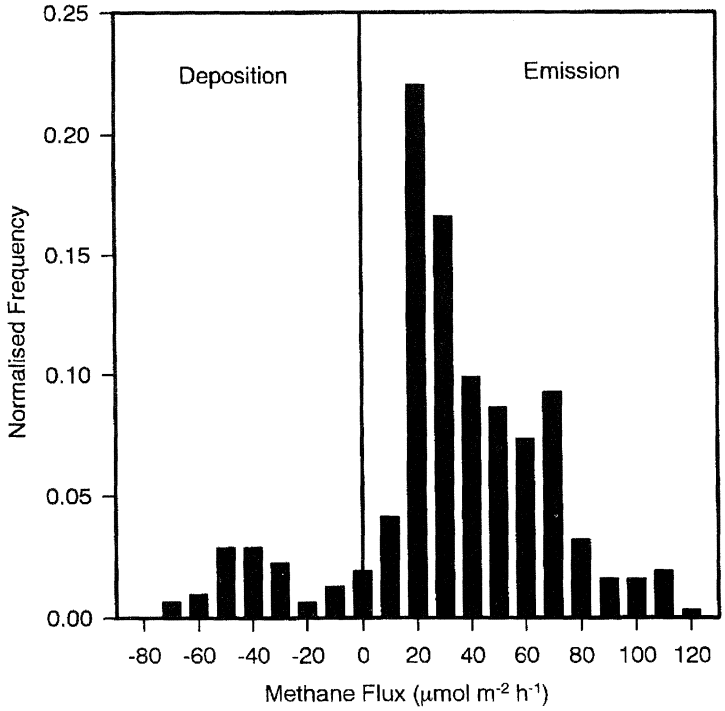


Figure 6. Frequency distribution of CH_4 fluxes by relaxed eddy accumulation over peatland at Loch More, Caithness May–June 1993.

8. Footprints of trace gas flux measurements

The clear relationship between properties within upwind fetch and measured fluxes is a key step in the interpretation of these measurements. Techniques to quantify the spatial contributions within the fetch to measured fluxes have recently been developed by Schuepp *et al.* (1990), and by Leclerc & Thurtell (1990). The approach of Schuepp *et al.* in providing a two-dimensional analytical solution of the diffusion equation, yields a cumulative normalized contribution to the flux (F_n) at heights from an upwind fetch x_L from the measurement mast given by

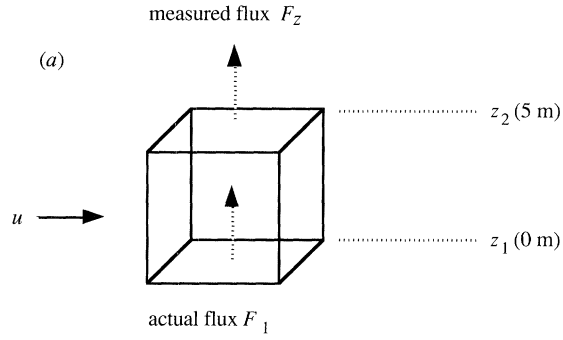
$$F_n(x_L) = \int_0^{x_L} \left[\frac{U(z-d)}{u_* k x^2}\right] \exp\left[\frac{-U(z-d)}{k u_* x}\right] \mathrm{d}x \tag{8.1}$$

$$= \exp\left[\frac{-U(z-d)}{k u_* x_L}\right]. \tag{8.2}$$

For non-neutral conditions equation (8.2) becomes

$$F_n = \exp\left[\frac{-U(z-d)\phi_M}{k u_* x_L}\right], \tag{8.3}$$

where U is horizontal wind velocity ($\mathrm{m\,s^{-1}}$), z the measurement height (m), d the zero plane displacement (m), ϕ_M a dimensionless stability coefficient and u_* the friction velocity ($\mathrm{m\,s^{-1}}$). Taking typical values for U, z, d, ϕ_M and u_* from measurements at the CH_4 flux tower (figure 8) the scale of the footprint for flux measurements at Loch More can be quantified for a range of atmospheric



Storage:

$$\Delta F_s = \int_{z_1}^{z_2} \frac{d\chi}{dt}\, dz = \frac{d\chi}{dt}(z_2 - z_1),$$

where $\Delta F_s = 1.8\ \mu\text{mol m}^{-2}\ \text{h}^{-1}$ for 10 ppb h^{-1}.
Advection:

$$\Delta F_\chi = \int_{z_1}^{z_2} U \frac{d\chi}{dx}\, dz = \frac{d\chi}{dx}(z_2 - z_1),$$

where $\Delta F_\chi = 3.1\ \mu\text{mol CH}_4\ \text{m}^{-2}\ \text{h}^{-1}$ per ppb km.

Figure 7. The magnitudes of non-stationarity errors due to advection and storage using typical conditions from the Loch More CH_4 measurements May–June 1993.

stabilities (figure 10). The data obtained in neutral and unstable conditions (*ca.* 90% of the data) are dominated by sources within 300 m of the tower, with typically 75% of the flux from this section of the fetch. In stable conditions, the footprint providing 75% of the flux extends to more than 1km from the tower. At this site, the fetch at distances greater than 400 m west of the tower becomes drier again, which complicates the sector analysis in strongly stable conditions.

Footprint analysis is a particularly important development in micrometeorological methods now that instruments are able to provide fluxes for a broad range of trace gases. It is necessary to link observed fluxes quantitatively with key physical and biological variables in the footprint.

9. Temperature

The clear sector dependence of mean fluxes in the data from Loch More (figure 8) shows the degree to which it is necessary to characterize the surface within the footprint before interpretation and analysis of the effects of environmental variables on emission rates. The large increase in emission with temperature observed from enclosure studies (Bartlett *et al.* 1992; Svensson 1984) has been confirmed in landscape scale measurements by Fan *et al.* (1993) and Edwards *et al.* (1994), although both groups found that responses to temperature were weaker than those observed by Bartlett *et al.* and were approximately linear at about 4 $\mu\text{mol m}^{-2}\ \text{h}^{-1}\ ^\circ\text{C}^{-1}$. Combining all data from Loch More would, of course, confound any relationship which might exist for a particular wetland sector. The data have therefore been analysed by sector to show the temperature response. An example for the 30° sector centred on 260° (figure 11) shows a doubling in emission

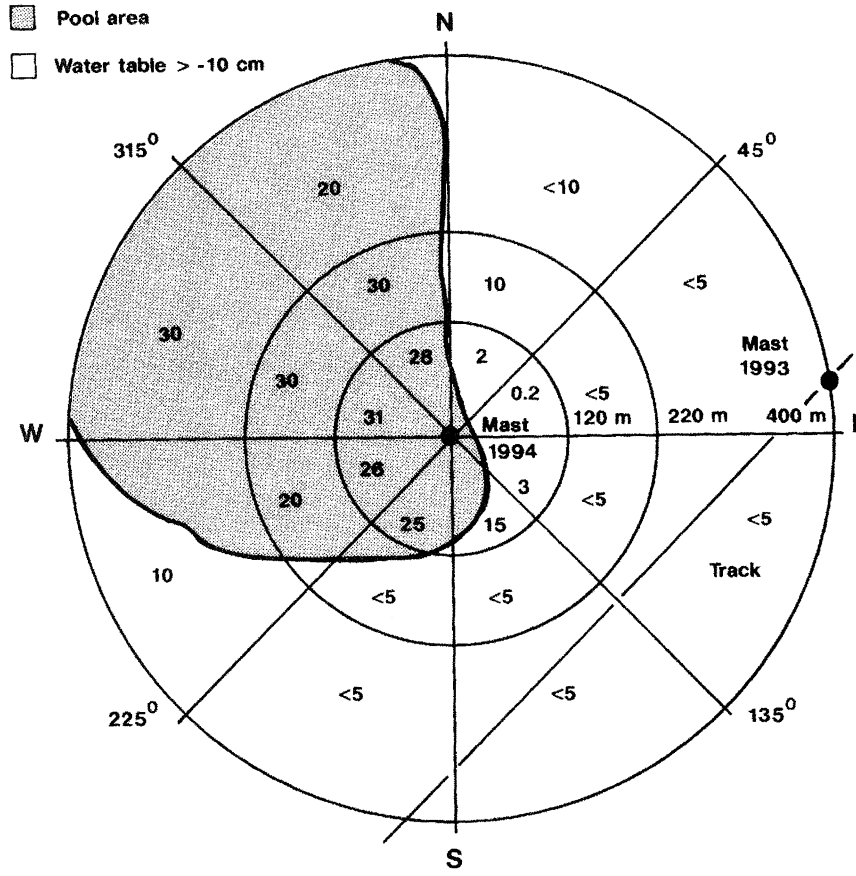


Figure 8. Location of CH_4 measurement tower, Loch More 1993 and 1994. The shaded area represents the area of pools, the values representing the % open water within each sector.

from 20 μmol CH_4 m^{-2} h^{-1} over the temperature range 7–12 °C. The approximately linear increase in this temperature range is close to the value obtained from controlled environment studies of wetland monoliths for pool and lawn of 6 μmol CH_4 m^{-2} h^{-1}. Similarly, clear day-night variations in mean CH_4 emission rates may be seen in the same data set. Taking the westerly sector 180–360° and comparing mean fluxes over the period with global radiation $St < 20$ W m^{-2} with data for the period with $St > 100$ W m^{-2} showed 30% (table 1) larger fluxes during the day. Mean temperature differences between the two periods of 2.5 °C were equivalent to 4 μmol m^{-2} h^{-1} °C^{-1} indicating that the bulk of the day-night differences in emission flux is simply an effect of temperature.

10. Extrapolation to global scales

The analysis of wetland methane emission by Matthews & Fung (1987), Fung *et al.* (1991) and by Asselmann & Crutzen (1989), provide similar estimates of the areas of wetlands and their likely source strength for CH_4 of 111 Tg CH_4 a^{-1} and 80 Tg CH_4 a^{-1} respectively. A detailed analysis of the issues and a revised estimate lie outside the scope of this paper, but it is clear from the work over Alaskan

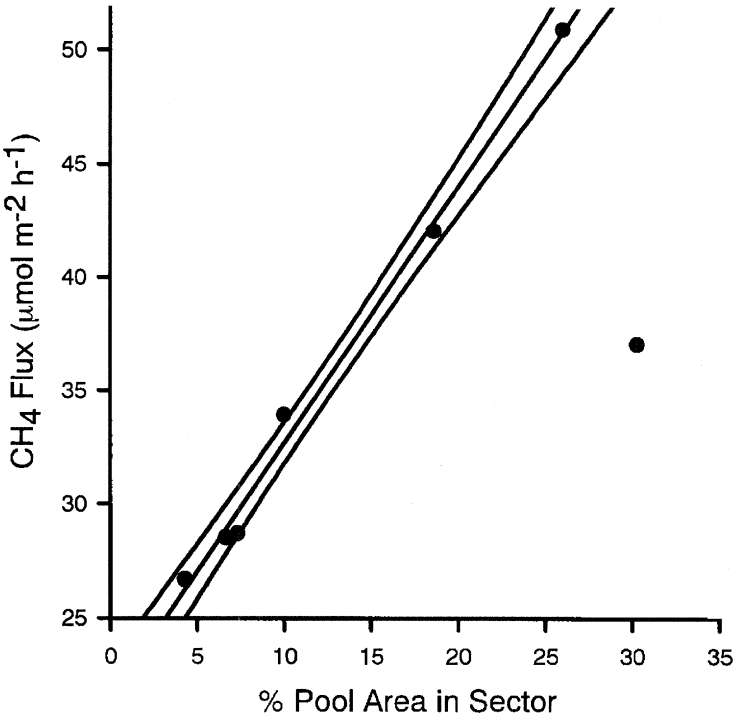


Figure 9. The relationship between the percentage of pool area and CH_4 flux within the fetch at Loch More, May–June 1994. 95% confidence interval shown.

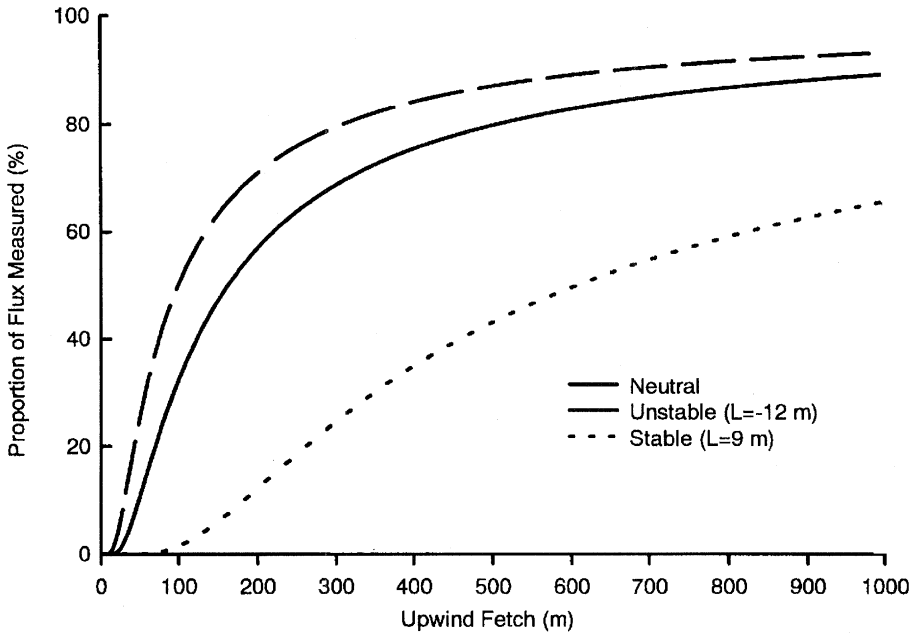


Figure 10. Examples of flux footprints calculated for the site at Loch More, May–June 1994.

Table 1. *Comparison of daytime and night-time fluxes – Loch More 1994*

	day	night
CH_4 flux (μmol m^{-2} h^{-1})	40.3	30.2
temperature (°C)	10.9	8.3

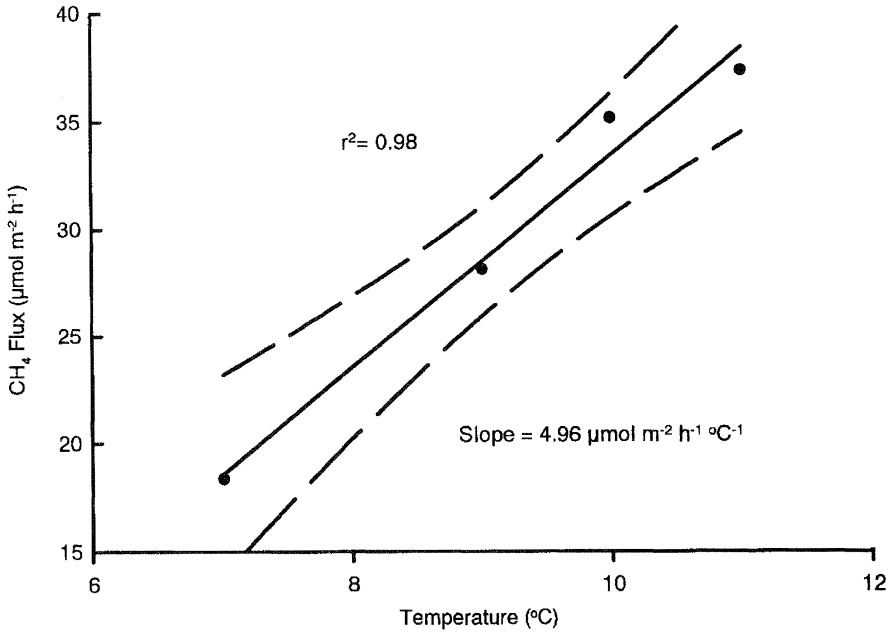


Figure 11. The temperature dependence of CH_4 emission from the 30° sector centred on 260° at Loch More containing approximately 20% open water during May and June 1994. 95% confidence interval shown.

tundra (Fan *et al.* 1992) over the Hudson Bay lowlands (Edwards *et al.* 1994) and from the work reported here for Scottish wetlands, that summer mean CH_4 fluxes are very similar for all three sites, ranging from 11 to 40 μmol CH_4 m^{-2} h^{-1} (table 2). These values are smaller than the mean flux from these regions adopted by Matthews & Fung (1987) by a factor in the range 4–7.

Taking the area of high latitude wetlands as 3×10^{12} m^2 and annual emissions of 25 Tg as typical of the estimates reported by Matthews (1993), and using global climate data, the methane emitting season may be shown to be approximately four months with average annual emission rates of 9.3 g m^{-2} and 180 μmol CH_4 m^{-2} h^{-1}. There are several possible causes for the difference, including the small number of sites at which measurements have been made. However, it is instructive to examine the global data bases for peatland and wetland land classes.

The Olsen global land-use database identifies several ecosystems which contain high latitude peat wetlands, including taiga, tundra and wooded tundra. These areas are extensive and collectively represent 20.8×10^{12} m^2, whereas the area taken for northern wetlands by Matthews & Fung is 2.7×10^{12} m^2. Other authors

Table 2. *Northern wetland* CH_4 *emission*
(Mean flux in μmol m^{-2} h^{-1}.)

location	surface	time	mean flux	reference
Caithness, U.K.	dry blanket bog	May/June 1993	14.7	this work
	wet blanket bog	May/June 1994	38.6	
Alaska, U.S.A.	wet meadow tundra	July/Aug 1988	29	Fan *et al.* 1992
	dry tundra	July/Aug 1988	11	Fan *et al.* 1992
Hudson Bay, Canada	raised bog	July 1990	40.5	Edwards *et al.* 1994

including Walen & Reeburgh (1988), Ritter *et al.* (1992) and Seebacher *et al.* (1986), all suggest larger values, between 4.5 and 9×10^{12} m^2. It seems that the difference between the measured landscape scale field measurements and those derived from inverse modelling are as likely to be due to uncertainties in the areal extent of high latitude wetlands as errors in the measurements.

11. Response of high latitude wetlands to temperature

The clear dependence of methane emission on the temperature of the surface layer of the wetland provides an indication of the likely sensitivity of methane emission to the surface temperature changes expected at high latitudes during the next half century (IPCC 1992). The methane fluxes from field measurements may be used to assign an appropriate landscape high latitude wetland flux at the mean temperature of the measurements. The temperature response in emission (figure 11) of 4.5 μmol CH_4 m^{-2} h^{-1} $°C^{-1}$ may be used with monitored wetland climate data to estimate the annual emission. This provides an annual cycle in emission for the Scottish wetlands which have an unusually long 'emitting season' relative to that over more continental peatland. If the temperature is assumed to be uniformly increased by 4 °C without changing the water table, then the annual emission becomes 6.4 g m^{-2}, an increase of 77%.

Such exercises are very speculative and contain naive assumptions, in particular that the seasonal changes in temperature in a warmer climate are uniform and that there is no influence on the hydrology of wetlands. However, the pronounced positive response to temperature of methane emission is a particular feature of wetlands and significant responses in CH_4 emissions are to be expected. It is of interest, therefore, to apply the above arguments to the wetland area of the high northern latitudes. The computed flux for current climate using the Matthews & Fung wetland area of 2.7×10^{12} m^2 is 6.1 Tg CH_4 annually, and increases to 10.7 Tg CH_4 with a 4 °C warming which is a factor of 4 to 6 smaller than most global estimates for this source.

This work forms a part of the Terrestrial Initiative in Global Environmental Research (TIGER) of the UK Natural Environment Research Council.

References

Asselmann, I. & Crutzen, P. 1989 Global distribution of natural freshwater wetlands and rice paddies, their net primary productivity, seasonality and possible methane emissions. *J. atmos. Chem.* **8**, 307–358.

Bartlett, K. B., Crill, P. M., Sass, R. L., Harriss, R. C. & Dise, N. B. 1992 Methane emissions from tundra environments in the Yukon–Kuskokwim delta. *J. geophys. Res.* **97**(D15), 16645–16660.

Businger, J. A. & Oncly, S. P. 1990 Flux measurement with conditional sampling. *J. atmos. Ocean. Technol.* **7**, 349–352.

Beverland, I. J., O'Neill, D. H., Scott, S. L. & Moncrieff, J. B. 1995 Design, construction and operation of flux measurement systems using the conditional sampling technique. *Atmos. Environ.* (Submitted.)

Bouwman, A. E. 1990 *Soils and the greenhouse effect.* Wiley.

Desjardins, R. L. 1977 Description and evaluation of a sensible heat flux detector. *Boundary-layer Meteorol.* **11**, 147–154.

Desjardins, R. L., MacPherson, J. I., Schuepp, P. H. & Hayhoe, H. N. 1994 Airborne flux measurements of CO_2, sensible and latent heat over the Hudson Bay lowland. *J. geophys. Res.* **99**(D1), 1551–1561.

Duyzer, J. H., Verhagen, H. L. M., Weststrate, J. M., Bosveld, F. C. & Vermetten, A. W. M. 1994 The dry deposition of ammonia on to a Douglas fir forest in the Netherlands. *Atmos. Environ.* **28**(7), 1241–1253.

Dyer, A. J. & Hicks, B. B. 1970 Flux gradient relationships in the constant flux layer. *Q. Jl R. meteor. Soc.* **96**, 715–721.

Edwards. G. C., Neumann, H. H., denHartog, G., Thurtell, G. W. & Kidd, G. 1994 Eddy correlation measurements of methane gluxes using a tunable diode laser at the Kinosheo Lake tower site during the Northern Wetlands Study (NOWES). *J. geophys. Res.* **99**(D1), 1511–1517.

Ehhalt, D. H. 1974 The atmospheric cycle of methane. *Tellus* **26**, 58–70.

Fan, S. M., *et al.* 1992 Micrometeorological measurements of CH_4 and CO_2 exchange between the atmosphere and subarctic tundra. *J. geophys. Res.* **97**(D15), 16627–16643.

Fowler, D. & Duyzer, J. 1989 Micrometeorological techniques for the measurement of trace gas exchange. In *Exchange of trace gases between terrestrial ecosystems and the atmosphere* (ed. M. O. Andreae & D. S. Schimel), pp. 189–207. Wiley.

Fung, I., John J., Lerner, J., Matthews, E., Prather, M., Steele, L. P. & Fraser, P. J. 1991 Three-dimensional model synthesis of the global methane cycle. *J. geophys. Res.* **96**, 13033–13065.

Hargreaves, K. J., Skiba, U., Fowler, D., Arah, J., Wienhold, F. G., Klemedtsson, L. & Galle, B. 1994 Measurement of nitrous oxide emission from fertilized grassland using micrometeorological techniques. *J. geophys. Res.* **99**(D8), 16569–16574.

Harriss, R. C., Gorham, E., Sebacher, D. I., Bartlett, K. B. & Flebb, P. A. 1985 Methane flux from northern peatlands. *Nature, Lond.* **315**, 652–653.

IPCC 1992 *Climate change.* Supplementary report to the IPCC Scientific Assessment (ed. J. T. Houghton, B. A. Callander & S. K. Varney). Cambridge University Press.

Khalil, M. A. K. 1993 *Atmospheric methane: sources, sinks and role in global change.* Springer.

Leclerc, M. Y. & Thurtell, G. W. 1990 Footprint prediction of scalar flux using a markovian analysis. *Boundary-layer Meteorol.* **53**, 247–258.

Matthews, E. 1993 Wetlands. In *Atmospheric methane: sources, sinks and role in global change* (ed. M. A. K. Khalil), pp. 314–361. Springer.

Matthews, E. & Fung, I. 1987 Methane emission from natural wetlands: global distribution, area and environmental characteristics of sources. *Global Biogeochem. Cycles* **1**, 61–86.

McMillen, R. T. 1986 *NOAA Tech. Memo.* ERL ARL-147.

Migeotte, M. V. 1948 Methane in the Earth's atmosphere. *J. Astrophys.* **107**, 400–403.

Moncrieff, J. B., Massheder, J. R., DeBruin, H., Elbers, J., Friborg, T., Huesunkveld, B., Kabat, P., Scott, S., Soegaard, H. & Verhoef, A. 1995 Surface fluxes of momentum, sensible heat flux, water vapour and carbon dioxide. *J. Hydrology.* (Submitted.)

Monteith, J. L. & Unsworth, M. H. 1990 *Principles of environmental physics.* London: Edward Arnold.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Morrissey, L. A. & Livingston, G. P. 1992 Methane emissions from Alaska Arctic tundra. An assessment of local spatial variability. *J. geophys. Res.* **97**(D15), 16661–16670.

Rasmussen, R. A. & Khalil, M. A. K. 1981 Atmospheric methane: trends and seasonal cycles. *J. geophys. Res.* **86**, 826–829, 832.

Raupach, M. R. 1989 Stand overstorey processes. *Phil. Trans. R. Soc. Lond.* B **324**, 175–190.

Raynaud, D., Chappellaz, J., Barnola, J. M., Korotkevich, Y. S. & Lorius, C. 1988 Climatic and CH_4 cycle implications of glacial–interglacial CH_4 change in the Vostok ice core. *Nature, Lond.* **333**, 655–657.

Ritter, J. A., Barrick, J. D. W., Sachse, G. W., Gregory, G. L., Woerner, M. A., Watson, C. E., Hill, G. F. & Collins, J. E. 1992 Airborne flux measurements of trace spaces in an arctic boundary layer. *J. geophys. Res.* **97**, 16601–16625.

Scheupp, P. H., Leclerc, M. Y., MacPherson, J. I. & Desjardins, R. L. 1990 Footprint prediction of scalar fluxes from analytical solutions of the diffusion equation. *Boundary-layer Meteorol.* **50**, 355–376.

Sebacher, D. I., Harriss, R. C., Bartlett, K. B., Sebacher, S. M. & Grice, S. S. 1986 Atmospheric methane sources: Alaskan tundra bogs, an alpine fen, and a subarctic boreal marsh. *Tellus* B **38**, 1–10.

Smith, K. A., Scott, A., Galle, B. & Klemedtsson, L. 1994 Micrometeorological and chamber methods for measurement of nitrous oxide fluxes between soils and the atmosphere: overview and conclusions. *J. geophys. Res.* **99**, 16 585–16 592.

Stauffer, B., Lochbronner, E., Oeschger, H. & Schwander, J. 1988 Methane concentrations in the glacial atmosphere was only half that of the preindustrial Holocene. *Nature, Lond.* **332**, 812–814.

Svensson, B. H. 1984 Different temperature optima for methane formation, when enrichments from acid peat are supplemented with acetate or hydrogen. *Appl. environ. Microbiol.* **48**, 389–394.

Svensson, B. H. & Rosswall, T. 1984 *In situ* methane production from acid peat in plant communities with different moisture regimes in a subarctic mine. *Oikos* **43**, 341–350.

Thom, A. S. 1975 Momentum mass and heat exchange. In *Vegetation and the atmosphere* (ed. J. L. Monteith), pp. 57–109. Academic Press.

Thom, A. S., Stewart, J. B., Oliver, H. R. & Gash, J. H. C. 1975 Comparison of aerodynamic and energy budget estimates of fluxes over a pine forest. *Q. Jl R. meteor. Soc.* **101**, 93–105.

Webb, E. K., Pearman, G. I. & Leuning, R. 1980 Correction of flux measurements for chemistry effects due to heat and water vapour transfer. *Q. Jl R. meteor. Soc.* **106**, 85–100.

Whalen, S. C. & Reeburgh, W. S. 1988 A methane flux time series for tundra environments. *Global Biogeochem. Cycles.* **2**, 399–409.

Woodward, F. I. & Sheehy, J. E. 1983 *Principles and measurements in environmental biology*, pp. 129–133. London: Butterworths.

Discussion

P. Ineson (*Merlewood Research Station, Grange-over-Sands, Cumbria, U.K.*). From the data presented Dr Fowler concludes that methane emissions can be very closely related to soil temperature, and he subsequently used the relationship to predict the impact of predicted global warming on methane fluxes. It is likley that temperature and photosynthetically active radiation will be correlated in this study, since day and night observations were made. Can he comment on potential links between solar radiation and methane emissions?

D. Fowler. The temperature response observed in the field using micrometeorological methods averaged over an area of the upwind fetch containing approximately 20% open water was approximately 5 μmol CH_4 m^{-2} h^{-1} $°C^{-1}$ within the

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

temperature range 7 to 12 °C. This may be compared with the average difference between CH_4 during the night and day of about 10 μmol m^{-2} h^{-1} with temperature differences of 2.6 °C. So, while it is possible that photosynthetically active radiation influences methane emission, probably through substrate supply to the root zone from current photosynthesis, the night–day differences in CH_4 emission may be explained entirely as a temperature effect.

J. Garland (*AEA Technology, Culham, U.K.*). Was Dr Fowler able to compare fluxes measured by different micrometeorological methods, and how well did the results compare? Does the heterogeneity of the fetch cause uncertainty regarding the applicability of any of the micrometeorological methods?

D. Fowler. The eddy covariance and relaxed eddy accumulation methods were compared for both CO_2 and CH_4 fluxes. For CH_4, the eddy covariance method provided lower noise in the 30-minute average fluxes and a lower detection threshold, but these may largely be attributed to the use of the TDL for CH_4 measurements using eddy covariance, while the relaxed eddy accumulation method relied on FID gas chromatography.

For the CO_2 flux measurements, the long-term (daily) fluxes using the two methods, both of which used identical CO_2 analyzers were within approximately 10%, which for micrometeorology represents good agreement.

On the question of effects of heterogeneity in the fetch on the measurements, the short-term data show evidence of flux divergence, which may be a consequence of spatial variability in the emission source strength. With more detailed analysis of these data and application of footprint analyses, it may be possible to quantify these effects directly. An alternative, and more rigorous approach, would be to include transects of chamber measurements in the upwind fetch to obtain independent estimates of spatial variability and therefore quantify these effects. This question identifies one of the key areas of uncertainty in the current uses of micrometeorology for trace gas fluxes, and one of the main research opportunities.

C. Johnson (*Hadley Centre for Climate Prediction and Research, Bracknell, U.K.*). Given that we may expect temperature and moisture to be anti-correlated over the longer time scales, does Dr Fowler expect that an increase in temperature would lead to an increase or decrease in methane fluxes?

D. Fowler. The temperature response of CH_4 emission from wetlands is a marked effect and would lead to a doubling of CH_4 emission for a short-term temperature increase of about 3 °C, but the long-term effects are unknown. Furthermore, the effect of the water table on CH_4 emission is equally dramatic, with average emission fluxes from the relatively 'dry' blanket bog 50% smaller than those from the areas with open water. The net long-term effect on CH_4 emission, of increases in mean temperature, will most probably therefore be determined by changes in the hydrology of these wetland areas. But the overall source strength remains very uncertain and must be a priority area for further research.