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Trace Gas Flux Measurements at the Landscape Scale Using Boundary-Layer Budgets [and Discussion]

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Trace gas flux measurements at the landscape scale using boundary-layer budgets

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Methane effluxes from wetland areas of Scotland were estimated by using the boundary-layer budget method by collecting air samples with an aircraft upwind and downwind of an area of extensive peatland. Nocturnal local area methane fluxes were also estimated at a peat bog site, Loch More, located at 58° 24′ N 03° 36′ W, using the concentration build up under the nocturnal inversion and from profiles of methane concentration using a tethered balloon.

The mean daytime flux for the Loch More case studies in 1993 was found to be 128 ± 57 μmol m^{-2} h^{-1} for the NE region of Scotland, comparable to but generally larger than those obtained for the same region one year earlier. The fluxes are smaller than those obtained in Caithness by the same technique. In 1993 the nocturnal fluxes were found to be 38 ± 7 μmol m^{-2} h^{-1}, significantly smaller than those found during 1992. The daytime fluxes measured by the aircraft were generally larger than fluxes measured by micrometeorological techniques at the same time. These differences can be explained in terms of the significant heterogeneity in surface fluxes that exist on scales of a few hundred metres or less and the possibility of additional sources other than peatland in this region.

1. Introduction

Methane is an important chemically and radiatively active trace gas in the atmosphere being the second most important greenhouse gas. At present methane contributes approximately 15% to enhanced radiative forcing and concentrations have more than doubled since pre-industrial times. Over the past 40 years measurements have shown that methane concentrations are still increasing although the rate has decreased from about 20 ppbv per year in 1970 to as low as 10 ppbv per year in 1989 (Steel *et al.* 1992). Since methane is a much more effective radiative gas than CO_2, it is important to understand its sources and sinks, and how they are linked with climatological feedback processes. At present many of

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these sources are known only semi-quantitatively although the major contributions come from natural wetlands, rice paddies and ruminants.

In the Northern Hemisphere methane is produced biogenically from anoxic soils in natural wetland areas such as peat bogs and rice paddies (Cicerone & Oremland 1988), where the average methane concentration of 1760 ppbv compares with 1680 ppbv in the Southern Hemisphere (Aselman & Crutzen 1989). Natural wetlands contribute significantly to the global methane emission with Northern hemisphere peatlands being the most important contributor, up to 60% (Mathews & Fung 1987). Many experiments have now been conducted over such peatland areas to quantify the magnitude of this source and they have used a variety of techniques, e.g. enclosure techniques (Crill *et al.* 1988), and eddy correlation (Verma *et al.* 1992). One of the main sources of uncertainty with enclosure techniques in particular is the large heterogeneity in source strengths on the microscale which makes estimating area averaged fluxes difficult. The large heterogeneity and relative importance of different wetland types to methane budgets has been highlighted by, for example, Yavitt & Fahey (1994), who suggest that high methane production from flooded forests contribute a significant amount to the methane budget in the Northern Hemisphere despite covering a relatively small area.

In this paper we describe measurements of methane fluxes from a large area of peat bog in northern Scotland on the landscape scale using the atmospheric boundary layer as a natural box. Different methods are used for daytime and night-time measurements. In addition we use aircraft measurements to obtain an estimate of the methane flux on the regional scale.

2. Techniques for field and landscape flux measurements

(*a*) *Boundary-layer budget studies by aircraft*

The boundary-layer budget technique (Betts *et al.* 1992; Harriss *et al.* 1992), using two instrumented aircraft from the UMIST Atmospheric Physics Group and the Meteorological Office Research Flight, was used to estimate methane effluxes from northern Scotland. The experimental details are described in Gallagher *et al.* (1994). The aircraft measurements provided the total methane budget over an area of approximately 10^3–10^4 km^2.

(*b*) *Measurement of methane efflux during nocturnal periods*

Two different techniques were used to estimate the methane efflux on scales ranging from a few hundred square metres to several square kilometres. These were (1) the nocturnal box model method and (2) balloon profile methods.

(i) *Nocturnal box model method*

For the first technique we assume that the increase in methane concentration at a fixed height and as a function of time can be described by the following simple zero-order relation:

$$C(t_0) + F\Delta t/z_i = C(t), \tag{2.1}$$

where F is a flux assumed to be a horizontally homogeneous constant source term, $C(t)$ is the methane concentration at some time t, $C(t_0)$ is the concentration at a specified start time, Δt is the time interval $(t - t)$, and z_i is the depth of the nocturnal boundary layer (NBL). This simplistic approach assumes that the height of the NBL remains constant during the observation period and that the stable boundary layer is well mixed. If this is the case then the concentration increase with time is constant and given by

$$\frac{\partial C(t)}{\partial t} = \frac{F}{z_i}, \tag{2.2}$$

where the flux $F = C(t_0)V$ is constant with time and V is the methane exchange velocity. The height of the NBL was estimated from Doppler Sodar measurements using a range of criteria. These included defining the NBL depth either at the height where the turbulent kinetic energy fell to small values, *ca.* 0.05 of that measured at the surface, or where a maximum in the wind profile or back-scattered signal was encountered or where the sodar return disappeared.

The assumption that the nocturnal boundary layer was well mixed may be investigated by comparing the total heat balance of the layer (radiative cooling, soil heat flux, sensible and latent heat flux to or from the surface) with the rate of change of temperature in the layer. It was generally found that this was a good assumption for the case studies presented in this paper. Further evidence for this may be obtained from the tethered balloon profiles described below.

(ii) *Balloon profile method*

The methane efflux can be estimated by integrating successive concentration profiles measured below the nocturnal inversion and then assuming simple diffusion to compute the flux knowing the increased methane burden in the NBL. A tethered radiosonde balloon was winched vertically up and down through the nocturnal boundary layer. It transmitted measurements of temperature, pressure and relative humidity to the surface. The balloon also carried aloft a tube attached at its lower end to a methane sensor to measure the methane concentration profile.

3. Results

We have selected results from three experiments conducted to date in the peat wetlands of Caithness & Sutherland in northern Scotland. The measurements were made over a period of three years and concentrated at two sites, Strathy Bog located at 58° 27′ N 04° 06′ W where measurements were conducted in 1992 and Loch More located at 58° 24′ N 03° 36′ W where measurements were made in 1993 and again in 1994. During these experiments different micrometeorological techniques were used to measure methane fluxes on local scales (flux profile, eddy correlation and relaxed eddy accumulation). An account of these measurements and the various techniques may be found in Fowler *et al.* (1994*a*) and Beverland *et al.* (1994).

(*a*) *Strathy Bog site experiment 1992*

The measurements made during this campaign have been described in detail by Gallagher *et al.* (1993) and so will only be summarized briefly here. Ground-based micrometeorological measurements made during this campaign were by the flux gradient technique (Fowler *et al.* 1993), and were largely confined to nocturnal periods. The measurements were made over a part of the peat bog which was very dry and the resulting fluxes were significantly smaller than those obtained by the

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

NBL box technique using the Sodar at the same site. Typical fluxes measured by the flux gradient technique ranged from 7 to 52 μmol m^{-2} h^{-1}, which compared with the NBL technique range of 9–178 μmol m^{-2} h^{-1} with mean nocturnal fluxes 101$\pm$63 μmol m^{-2} h^{-1} from the NBL technique. The discrepancy of a factor of 2 to 3 between the techniques was probably as a result of the heterogeneity in methane source strengths and effective measurement fetch for the different techniques. The micrometeorological measurements were made over a height range of 0.1–2.0 m. Applying the simple 'footprint' models of Schuepp *et al.* (1990) and Horst & Weil (1993) it can be estimated that most of the flux (*ca.* 80%) measured by this system would correspond to a source within 60–100 m (depending on stability) of the mast whereas the NBL technique provides an area average over typically 1 km^2 to several square kilometres.

During the daytime fluxes were made by aircraft at the landscape scale. Aircraft fluxes for the NE region covering this site were 79$\pm$37 μmol m^{-2} h^{-1}. In the area to the west of our measurement site much larger fluxes of 205$\pm$115 μmol m^{-2} h^{-1} were obtained from the aircraft measurements, probably reflecting the much wetter peatlands with many small lakes in this region.

It was interesting that the nocturnal fluxes were comparable to the daytime fluxes and about twice as large as one might expect based on previous studies. This was probably as a result of the unusually high nocturnal temperatures encountered during this period. Air temperature measured at a height of 1 m over the peat bog did not fall below 8.0 °C throughout the measurement period while soil temperatures measured at several locations at a depth of 10 cm ranged from 12.0 to 14.5 °C.

If one considers the aircraft fluxes for the SW and W regions where most of the large expanse of peatland is situated then the fluxes were very large, 205 $\pm$ 115 μmol m^{-2} h^{-1}, about twice as large as for the NE region and twice as large as the nocturnal fluxes. It was encouraging that the daytime fluxes measured using the aircraft budget technique in this region were typically twice as large as those measured during the nocturnal period by a very different technique. This type of day–night range has been reported for the Hudson Bay Lowlands area by Fan *et al.* (1992) and Edwards *et al.* (1994). The implications of these results is discussed further below.

(*b*) *Loch More site experiment 93*

In this experiment the Sodar was again used to measure fluxes by the NBL technique, together with some balloon borne measurements, while aircraft made measurements of the daytime fluxes. Methane measurements were also measured by the eddy correlation and relaxed eddy accumulation micrometeorological techniques (Fowler *et al.* 1993; Beverland *et al.* 1993). The site, as described by Fowler *et al.* (1994) was again quite dry. Rainfall, however, increased significantly towards the end of the measurement period and increased the methane emitting area.

(i) *Balloon profiles and nocturnal box model results*

A simple balloon borne system was tested during this experiment to estimate the methane efflux by integrating between successive concentration profiles in the NBL. A sample line was attached between the ITE tunable diode laser (TDL) system and a simple balloon sonde. The sonde data were later correlated with the time series of methane measured by the ITE TDL.

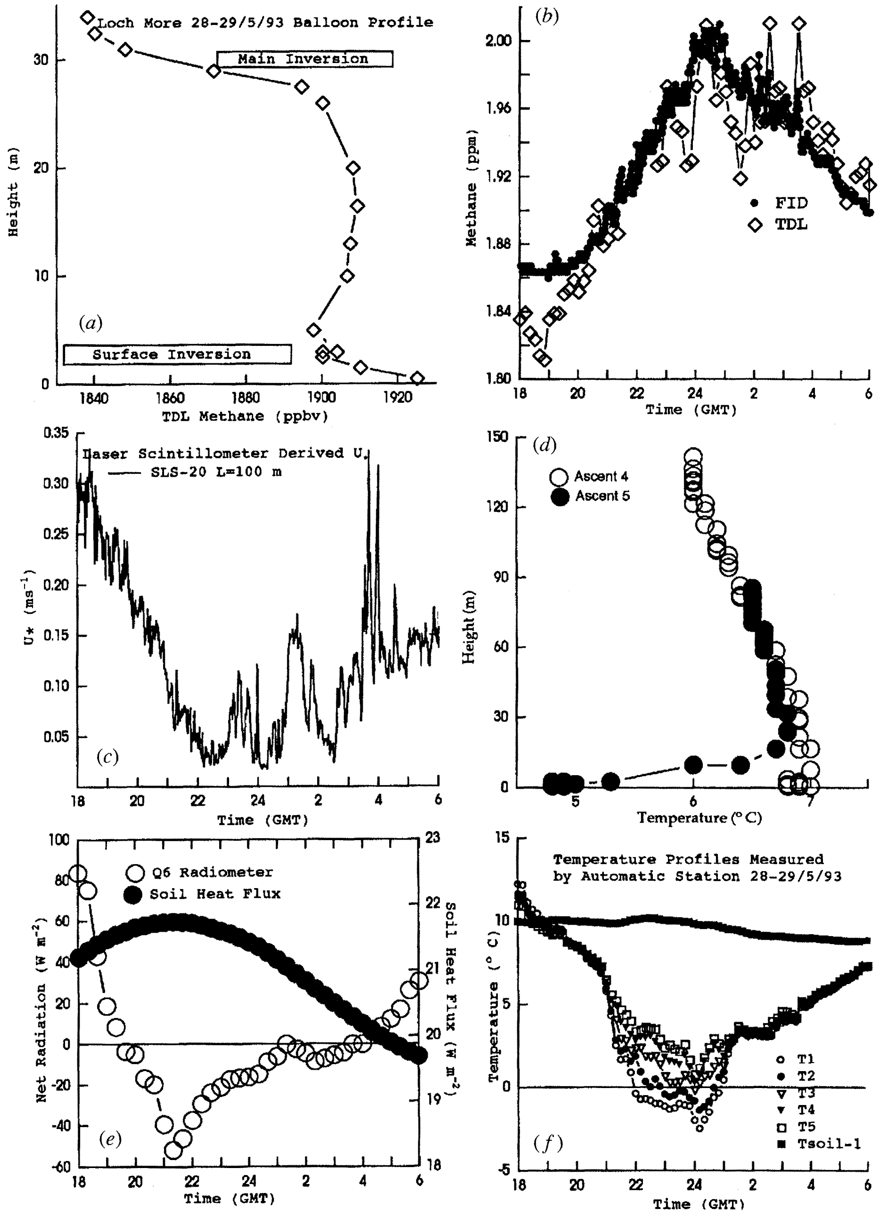

Figure 1. The measurements made during the nocturnal boundary-layer study of 28–29 May 1993.

On all but one occasion, the inversion height, as determined from the Doppler Sodar, exceeded the altitude limit of this simple system. However, on this one occasion good results were obtained which has encouraged us to develop a larger system for future experiments.

Figure 1*a* shows a methane concentration profile for the case study of the 28 May 1993. The main feature is the very sharp drop in concentration at 25 m, of between 50 and 75 ppbv, over a distance of approximately 5 m. The magnitude of the decrease locates the top of the NBL which effectively isolates the near surface

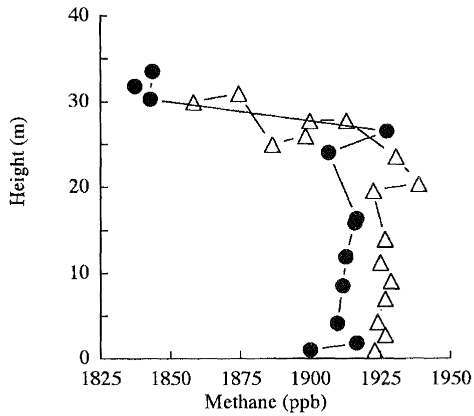

Figure 2. Balloon ascents used to determine methane flux.

layer from the air above. Below this inversion the layer is still quite well mixed with a methane concentration which only changes slightly with height, with the largest gradient very close to the surface. Nearly steady-state conditions could be applied during the period of the balloon flight. The methane concentration measured at 5 m by the TDL system and an FID instrument at 2.5 m (figure 1*b*) shows a steady increase in concentration with time as expected based on the assumptions used in the simple box model approach mentioned above. The TDL measurements show larger variations in concentration, the larger of these were anticorrelated with wind speed. Turbulence levels dropped to very low values, $u_* < 0.05\ \mathrm{m\,s^{-1}}$ between 22:00 and 23:00 GMT (figure 1*c*).

Temperature profiles at 20:30 show a slightly stable layer up to 30 m with a more stable layer extending up to 120 m and a near neutral layer above (figure 1*d*). By 21:00 the surface layer had begun cooling rapidly, with net radiation reaching a minimum of *ca.* 55 $\mathrm{W\,m^{-2}}$ at 21:30, and soil heat fluxes peaking at 60 $\mathrm{W\,m^{-2}}$ (figure 1*e*) The maximum rate of cooling occurred between these times. Figure 1*d* clearly shows the near surface stable layer becoming much more stable with a temperature difference of about 2.0 °C across the layer as a whole, which is about 25 m deep. Turbulence levels had decreased linearly during this period reaching a minimum just after 22:00. Subsequently the rate of cooling decreased as the net radiation increased again. Air temperatures as measured on a mast at heights between 0.1 and 2.8 m (figure 1*f*) continued to fall reaching a minimum just after midnight. Soil temperatures measured at 20 cm (figure 1*f*) decreased from 10.0 °C at 18:00 to 8.9 °C at 06:00 the following morning. During the period of maximum cooling when air temperatures and net radiation dropped rapidly the soil temperature showed a slight increase resulting in a subsequent increase in air temperature against the decreasing trend.

Profile measurements of methane were made between 21:30 and 22:40. Figure 2 shows two profiles which were obtained at 21:30 and 22:00 near the end of the period of maximum cooling, when the surface layer remained relatively constant in extent. There appeared to be no decrease in concentration within the lowest 15 m of the surface although detailed examination of data below 1 m did reveal a large gradient as expected. The two profiles indicated a reasonably well mixed 20 m layer with subsequent profiles being characteristic of a diffusion plume mixing process. Using the approximation that turbulence levels at 5 m are characteristic of the lowest 20 m of this layer as a whole and integrating subsequent profiles of methane concentration we calculated the area averaged flux required to reproduce the observed increase in methane concentration at all levels up to 20 m. There is some error due to the assumptions of steady state since the profile is not a true one due to the finite time taken to sample subsequent heights. The advection and storage error this introduces can be calculated after Fowler & Duzyer (1989), however, for this case study these were small. The fluxes obtained, 34–45 μmol m^{-2} h^{-1} agreed to within 20% of the fluxes obtained before and after the balloon ascent by eddy correlation.

Nocturnal fluxes, measured by the NBL box technique for this experiment as a whole were 56 ± 43 μmol m^{-2} h^{-1} which were about a factor of 2 smaller than those from the previous experiment in 1992. Methane fluxes have been shown to depend on the combined effect of peat temperature and water table depth since a decline in water table depth increases the depth of the aerated zone where oxidation can take place, reducing the methane efflux (Dise 1991). Several workers have reported significant anticorrelations between water table depth and methane flux (Shurpali *et al.* 1993; Yavitt *et al.* 1990; Whalen & Reeburgh 1992). The larger nocturnal fluxes observed during the Strathy Bog experiment were probably due to the site being significantly wetter and warmer. The peat temperatures, measured at a depth of 10 cm, during this Loch More experiment were typically 10 °C compared to an average of 14 °C during the Strathy Bog experiment in 1992.

Typical methane production-temperature response experiments conducted on cores from these sites (Fowler *et al.* 1994*a*) suggest a temperature sensitivity of production rates of 4.9 μmol m^{-2} h^{-1} $°C^{-1}$, this would explain 50% of the variation between the two periods.

(ii) *Daytime fluxes*

Daytime fluxes of methane were obtained from measurements made by the aircraft flying in race track patterns giving measurements of methane concentration within and above the boundary layer upstream, over and downstream of the Loch More site and at several altitudes within the boundary layer. These measurements of methane concentration were compared with predictions of a boundary-layer mixing model to deduce the mean surface flux of methane using exactly the same procedure as described in Gallagher *et al.* (1994). Figure 3 shows the vertical profile of methane measurements made above the site during a typical case study and compared to the predictions of the mixing model. The error bars on the data represent variations in methane concentration from several samples obtained at the same altitude in the same area.

The fluxes deduced from the aircraft data for the entire 1993 experiment were 128 ± 57 μmol m^{-2} h^{-1}, slightly larger than those found for the NE region during the Strathy Bog experiment in 1992. Daytime fluxes of methane measured by the eddy correlation technique typically peaked around 60 μmol m^{-2} h^{-1}, at 14:00 GMT after the solar maximum, and were about a factor of 2 smaller than the aircraft fluxes which were measured at about the same time. Mean fluxes for the entire data set, averaged over a day, were around 15–20 μmol m^{-2} h^{-1} (Fowler *et al.* 1993). The difference between the aircraft and micrometeorological data was thought to have been due to siting the micrometeorological towers on a part of the peat bog which was unusually dry.

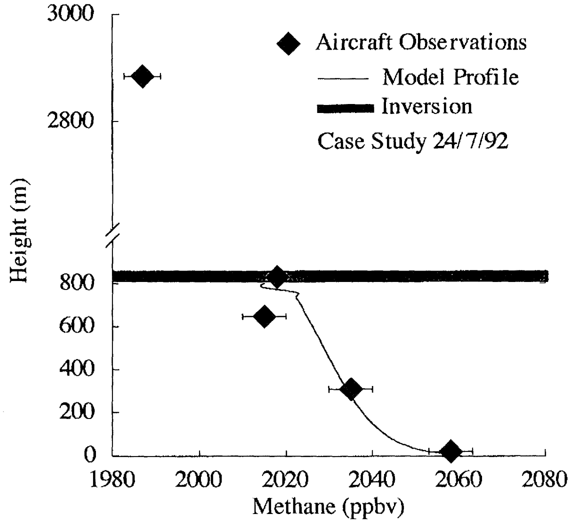

Figure 3. Comparison of observed and calculated methane profiles in the daytime boundary layer.

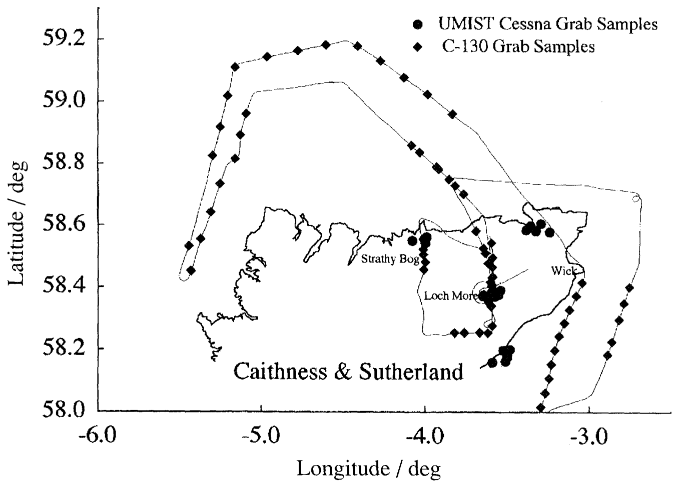

Figure 4. Flight plan for the regional scale study.

(iii) *Use of aircraft to estimate regional scale fluxes*

During the 1993 experiment the Meteorological Office Hercules aircraft made a flight around Northern Scotland at the same time as the UMIST Cessna aircraft was making measurements close to the main site. The flight tracks are shown in figure 4. The object of the flight was to attempt to measure regional scale fluxes from the area. The results are shown in figure 5. A large increase in methane concentration in the boundary layer is observed from around 1850 ppbv upwind of the area to as high as 1950 ppbv near the surface just off the north coast. This was downwind of the peat bogs. Analysing this data using our mixing model as described above, a maximum flux of 270 μmol m^{-2} h^{-1} was detected similar to the flux obtained over the very wet western area in the 1992 experiment.

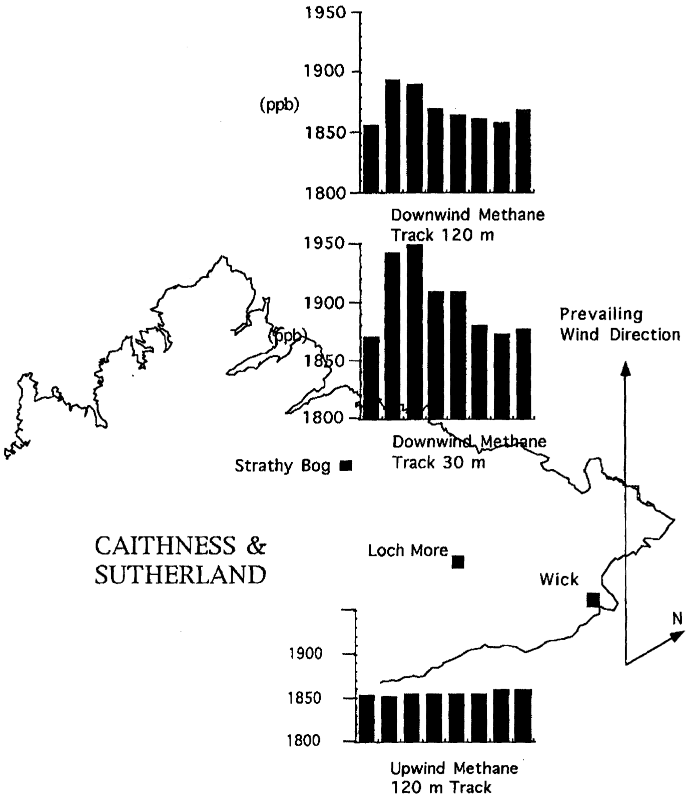

Figure 5. Methane concentrations from the regional scale study.

(c) Loch More experiment 94

These experiments were performed mostly by micrometeorological techniques over the wetter areas of the peatland near Loch More in an attempt to see if some of the discrepancies with the aircraft measurements could be resolved. Areas of the bog where the water table was close to the surface with anoxic conditions dominating were chosen which were expected to lead to higher daytime fluxes. Methane fluxes were found to be highly sector dependent with a near linear relation between effective pool area and methane emission flux (Fowler *et al.* 1994). Measurements made close to areas with many small pools showed fluxes of the order typically 50 μmol m^{-2} h^{-1} or more emission during the day but could reach 100–150 μmol m^{-2} h^{-1}. Daytime fluxes were typically about 1.5 times larger than nocturnal fluxes.

These fluxes are still smaller than those determined for the northeast region as a whole by aircraft budget measurements during the 93 experiment and which ranged from 79 to 144 μmol m^{-2} h^{-1}.

4. Discussion

It should be emphasized that the aircraft budget fluxes were obtained over relatively short periods during mid-afternoons which favoured maximum methane emission rates and over large fetches ranging from 20 to 60 km. The aircraft budget flux measurements will, by their very nature, be biased towards higher values than those which should be used to represent the actual regional mean flux. Fan *et al.* (1992) reported excellent agreement between aircraft eddy flux methane measurements and tower based methane flux measurements over short time periods but noted that the tower mean fluxes during the period of the aircraft flights was 2 times larger than the day mean flux from the tower. The more recent results from the Loch More 94 experiment at a site where a significant fraction of the surface consisted of small lakes and pool areas suggest that heterogeneity in methane fluxes at this site is a significant problem to scaling fluxes up to the regional scale. As found by Harriss & Sebacher (1981) and Bartlett *et al.* (1992) at such sites, variances of a factor of 10 or more occur even for similar vegetation type.

The mean flux for the region was calculated to be 205 ± 115 μmol m^{-2} h^{-1}. Methane source strengths for the northwestern region of Scotland were a factor of 2 to 3 times larger than the north eastern region. A very similar result was obtained for the regional scale fluxes in 1993.

Night-time fluxes reported during the Strathy Bog Experiment averaged 101 ± 63 μmol m^{-2} h^{-1} and were comparable to but generally larger than those measured using the flux gradient technique at the same site (Fowler *et al.* 1993). The values for the Loch More site in 1993 were 38 ± 7 μmol m^{-2} h^{-1}.

Strathy Bog was thus both warmer and wetter than Loch More, although the water table depth was larger at the start of the experiment but by the end the site was covered with standing pools due to the large rainfall amounts during the final period of the experiment.

Nocturnal temperatures at the Strathy site were significantly higher than at the Loch More site, with minimum air temperatures at 1m above the ground never falling below 7.5 °C. This is in qualitative agreement with the observation that the nocturnal fluxes at the Strathy Bog site were significantly larger than at Loch More whereas the daytime fluxes for the northeast region are not significantly different, although the Loch More site showed the larger daytime fluxes in general.

5. Conclusions

The main results of the three campaigns are summarized in table 1. Landscape scale fluxes of trace gases have been estimated.

1. At night a typical flux of around 50 μmol m^{-2} h^{-1} was detected.

2. By day the aircraft measurements gave values of typically 100 μmol m^{-2} h^{-1}. These were generally larger than the surface based micrometeorological measurements by a factor of 2. This was probably due to a bias in favour of dryer regions for these latter measurements. The measurements made in 1994 over wetter regions tended to support this suggestion.

3. Very large fluxes of around 200 μmol m^{-2} h^{-1} have been detected on a regional scale and on a landscape scale over western part of northern Scotland. This again is probably due to the very wet nature of this region.

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Table 1. *Flux summary for all experiments*

(Units are µmol m^{-2} h^{-1}. EC, eddy correlation; REA, relaxed eddy accumulation; NBL, nocturnal boundary layer box model.)

1992	gradient	REA	NBL	aircraft
daytime				79 ± 37 (NE)
				205 ± 115 (SW–NE)
night-time	7–52		101 ± 63	

1993	EC	REA	NBL	aircraft
daytime				128 ± 57 (NE)
				270 (Scotland)
night-time		15 ± 20	38 ± 7	
peak	60	50+		
mean	15 ± 15	22.7		

1994	EC	REA	NBL
daytime	40 ± 62	37 ± 35	
night-time	30 ± 50	21 ± 15	49 ± 28[a]
peak	170	150	
mean	39 ± 44	28 ± 26	

[a]Balloon profile.

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Discussion

J. L. Monteith (*Institute of Terrestrial Ecology, Penicuik, U.K.*). I was rather surprised to see profiles in which methane concentration was almost independent of height within the nocturnal boundary layer. This seems to imply either that the flux became very small at night, or, more likely, that the boundary layer was well stirred. Were the profiles obtained on cloudy or windy nights when the temperature inversion was very weak? If so, what do the profiles look like on cloudless, still nights?

T. W. Choularton. An example of the well mixed methane profile is presented in figure 2. This profile was obtained in light wind conditions (typically $1\ \mathrm{m\,s^{-1}}$) with a clear sky so a well developed nocturnal boundary layer was present. It can be seen that the methane concentration in the nocturnal layer was substanitally in excess of that above the layer. The nocturnal methane flux from the surface during these experiments was calculated to be about $40\ \mu\mathrm{mol\,m^{-2}\,h^{-1}}$. The data suggest that despite the stable stratification in the nocturnal layer it is sufficiently stirred for the layer to be well mixed with this methane flux.

R. J. Harding (*Institute of Hydrology, Wallingford, U.K.*). Atmospheric convergence or divergence is likely to have a significant influence on boundary-layer budgets. Is there an estimate of the convergence over northern Scotland during the flights?

T. W. Choularton. The synoptic scale convergence or divergence was always less than $10^{-5}\ \mathrm{s^{-1}}$. The boundary-layer budget studies using aircraft were performed over a distance scale of typically 50 km. Under these circumstances the influence of this parameter on the concentration changes measured will be less than 10%. This will introduce a similar error in the flux measurements. This is much less than our estimated error in any individual flux measurement (50%) and the day to day variation. The days selected for the budget calculations were marked by an absence of deep convection or major frontal systems.

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