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Eddy correlation measurements of methane fluxes using a tunable diode laser at the Kinosheo Lake tower site during the Northern Wetlands Study (NOWES)

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Abstract. As part of the Canadian Northern Wetlands Study (NOWES) measurements of methane flux were made at the Kinosheo Lake tower site for a 1-month period during the 1990 summer intensive. The measurements were made with a diode-laser-based methane sensor using the eddy correlation technique. Measurements of the methane fluxes were made at two levels, 5 or 18 m. Approximately 900 half-hour average methane flux measurements were obtained. Weak temporal and diurnal trends were observed in the data. Fluxes averaged over the study period showed an overall methane emission of 16 mg CH_4 m^{-2} d^{-1} with a daytime average of 20 mg CH_4 m^{-2} d^{-1} and a nighttime average of 9 mg CH_4 m^{-2} d^{-1}. The effect of emission footprint was evident in the data. A strong relationship between the daily average methane flux and wet bog temperature at 20-cm depth was observed.

1. Introduction

The Northern Wetlands Study (NOWES) is a multiyear program with the overall objective to assess the importance of northern wetlands as sources of biogenic gases to the atmosphere under current and future climate scenarios. The study and its objectives are described in detail by *Roulet et al.* [this issue].

The mean atmospheric concentration of methane has increased twofold over the past several hundred years and is estimated to be currently increasing at a rate of approximately 0.8% yr^{-1} [*Cicerone*, 1988; *Crutzen*, 1991]. Recently published estimates of the global input indicates that northern wetlands are one of the larger natural sources of atmospheric methane [*Aselmann and Crutzen*, 1989; *Fung et al.*, 1991]. These estimates have primarily been derived from measurements of small isolated wetlands using discrete enclosure methods. Little is known of the methane emissions from the large expansive wetlands of northern North America, Europe, and Asia.

The quantification of the methane emission from these areas is a challenging task. Enclosures can be used to characterize the flux from individual wetland types at discrete points in time throughout the year [*Moore et al.*, this issue], and these measurements can be spatially extrapolated using some form of ecosystem characterization by remote sensing [*Roulet et al.*, this issue]. However, the spatial average from the method is inferred through modeling and therefore is not an actual measured spatial average. In addition, this method can only resolve very coarse changes in gas flux which translates to measureable concentration changes that are only appreciable over time periods of weeks and therefore can only examine the relationship between the environmental controls on trace gas exchange on the time scale of seasons.

There is a need for high time resolution flux measurements that integrate over an area larger than that of a chamber but an area smaller than that measured by an aircraft [*Ritter et al.*, this issue] so the dynamic response of individual or several ecosystems can be examined. Micrometeorological approaches to flux measurement are ideal to examine the mesoscale exchange of trace gases. For methane these approaches have been limited by the precision and response time of available methane sensor technologies. For example, gas chromatography (GC) requires at least 30-s integration time and therefore is not appropriate for eddy correlation. As well, GCs do not have sufficient precision or accuracy to resolve gradients of a few parts per billion by volume (ppbv) which are typical over the bogs that are reported here. There have been eddy correlation studies that have used a flame ionization detector (FID) but a FID measure total hydrocarbon and not just methane. In some situations the non-methane hydrocarbons can make up a significant component of the hydrocarbon flux [*Klinger et al.*, this issue]. Hence for the tower methane flux measurements an eddy correlation approach was used and methane was measured using a new methane sensor based on tunable diode laser (TDL) spectroscopy, developed at the University of Guelph.

The objective of this paper is to discuss the component of NOWES which involved tower-based measurements of methane exchange. These measurements were taken over a raised bog near Kinosheo Lake on the Hudson Bay lowlands (HBL) during the 1990 summer intensive (i.e., June 25 to July 28). The laser system is described followed by the presentation of the flux measurements.

2. Site Description

The site was located approximately 100 km northwest of Moosonee, Ontario, at latitude 51°33′N and longitude 81°49.5′W. The tower was located 400 m to the west of the Kinosheo Lake shoreline. The area between the shoreline and the tower was primarily covered with black spruce. The canopy height decreased from the shoreline from approximately 15 m to 1 and 3 m within 100 m of the tower. The remainder of the fetch surrounding the tower was a raised open shrub-rich/graminoid bog, intermixed with open sphag-

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num bog, and small bog and fen pools, basal date approximately 4000 years [*Klinger*, this issue]. The dominant vegetation types were mosses and sedges (*Scripus caspitosus, Carex limosa, Eriophorum vaginatum, Sphagnum capillifolium, Drosera rotundifolia*) and shrubs (*Picea mariana, Larix laricina, Vaccinium vitisidaea*). In addition, there were large areas of permanent pond, and stunted black spruce, 1 to 3 m tall, occurred at random and were widely spaced. Further details on these wetlands can be found in the work of *Moore et al.* [this issue].

Climatological data on this wetlands region can be found in the work of *Mortsch* [1991]. The average ambient temperature at the Kinosheo Lake site over the June 25 to July 28 period was 17.2°C and 30.4 mm of precipitation was received. Ice was detected near 50-cm depth under the hummocks until mid-July [*den Hartog et al.*, this issue].

Climatic conditions during the 1990 summer season were not atypical for temperature and the snow-free period based on 1932 to 1989 means. Precipitation was abnormal during the month of June when it was twice the long-term average. During all other months the precipitation was within one standard deviation of the long-term average.

3. Methods

Full details on the Kinosheo Lake tower-based measurement program can be found in the work of *den Hartog et al.* [this issue]. A lightweight scaffold tower constructed of fibreglass and aluminum members was erected at the site. The tower height of 20 m facilitated the measurement of methane fluxes at two levels, 5 or 18 m. The methane sensor was located at midtower to allow movement of the sample tube to either measurement height. Flux measurement at the top level maximized the spatially integrated area of emission (i.e., footprint) for aircraft intercomparison. Measurements made at the lower level allowed examination of smaller fetch footprints.

The low shadow density of the tower and instrument booms minimized blocking effects. An air conditioned instrument tent was located 30 m to the east of the tower; 80 m to the east of the tower was the propane-powered generator. Ideal fetch conditions (i.e., uniform roughness) for flux measurement were from 210° through to 30° relative to true north. Measurements made in other wind sectors were eliminated from the data set as they were either influenced by significant roughness element changes or tower-blocking effects.

Eddy Correlation Technique

Micrometeorological techniques used for the measurement of trace gas exchange between the atmosphere and the Earth offer several advantages. Eddy correlation when standard micrometeorological criteria are met [*Hicks et al.*, 1989] will provide absolute evaluations of vertical fluxes in natural environments without making assumptions associated with diffusivities or the nature of the surface cover. In addition, the exchange rate measured represents a spatially integrated flux and the technique is unobtrusive, therefore not disturbing the environment under study. The micrometeorological technique of eddy correlation has been applied to measurement of the fluxes of heat, momentum, and water vapor for over 30 years [*Chamberlain*, 1961]. The technique requires that sensitive and fast response instrumentation be colocated above a surface to detect fluctuations of the vertical wind velocity w' and of the gas concentration c'. The covariance calculated over a time period will represent the flux, that is,

$$F_c = \overline{w'c'}$$

Extensive use of the technique for trace gas flux measurement has been limited by the availability of suitable chemical sensor technologies and data acquisition and real-time data processing systems. Recent developments in computer-based data systems (i.e., high-speed, high-resolution analog to digital conversion systems) and sensitive, fast response TDL-based gas monitors [*Thurtell et al.*, 1991] have facilitated the continuous measurement of the flux of some of the trace atmospheric gases (e.g., nitrous oxide and methane).

Further details of the eddy correlation technique and its application to the measurement of trace gas exchange can be found in the work of *Neumann and den Hartog* [1985]. Data reduction techniques (e.g., coordinate rotation, density correction, wind direction, wind speed data selection, etc.), as employed by *Tanner and Thurtell* [1969], *Webb et al.* [1980], and *Hicks et al.* [1989], were used in the analysis of the data presented here. In addition, a correction for storage effects based on the half-hour variation in the mean methane concentration and the measurement height was applied to the data. This correction accounts for the divergence of the flux between the surface and the measurement height. Flux divergence can arise when horizontal uniformity and steady state conditions are not completely met [*Businger*, 1986].

The eddy correlation instrumentation used for methane flux measurement was a three-dimensional omnidirectional sonic anemometer (model dat 310, Kaijo Denki Company, Tokyo, Japan), a Lyman alpha hygrometer (A.I.R. Incorporated, Boulder, Colorado), and the TDL methane sensor. All eddy correlation data were acquired at a rate of 20 Hz, using a 16-bit A/D converter (Scientific Solution Labmaster) and stored on hard disk (Dell Computer Corporation 386) and removable hard disk drives (Mega Drive Corporation). The analog signals were passed through a low-pass Bessel filter with a cutoff of 10 Hz prior to digitization. All data, including supporting measurements, were archived on optical worm drives (Corel Systems). The supporting micrometeorological measurements and the micrometeorological characteristics of the site are described by [*den Hartog et al.*, this issue].

TDL Methane Sensor

The technique of tunable diode laser absorption spectroscopy has been applied to the detection of trace gases in the atmosphere for over a decade [*Reid et al.*, 1978*a*, *b*; *Schiff et al.*, 1983, 1987]. The value of the technique lies in the fact that it couples the advantage of high sensitivity, of the order of a few parts per billion by volume or better, with highly selective detection of any one of a wide range of trace gas species such as CH_4, N_2O, CO_2, NH_3, CO, and NO_2.

TDL absorption spectroscopy has also been successfully applied to the measurement of the fluxes of trace atmospheric gases [*Edwards et al.*, 1982, 1984, 1985, 1987, 1988; *Ogram et al.*, 1988; *Thurtell et al.*, 1991; *Verma et al.*, 1992]. Data recovery were low during the earlier experiments because the TDL instrument design used was not resilient when operated in field environments. For the NOWES the TDL field system was completely redesigned to be compact, portable, and sufficiently robust to operate continuously out of doors unaffected by varying weather conditions [*Thurtell et al.*, 1991]. This involved simplifying the optical design, engineering the chassis to render the instrument insensitive to vibration, designing an integrated digital signal-processing and laser control system, and controlling the laser-operating environment.

The TDL technique is based on infrared absorption spectroscopy, whereby the extent of absorption depends on path length, line strength, and absorber concentration. Individual absorption lines are easily and unambiguously resolved at low pressure (i.e., typical emission line width 10^{-4} cm^{-1}), even in wavelength regions where H_2O and CO_2 absorb strongly.

A schematic of the TDL gas monitor developed for the NOWES tower-based measurements is shown in Figure 1. The diode laser is mounted in an LN_2 cooled dewar (Laser Photonics, Analytics Division). A heater mounted inside the dewar facilitates precise control of the laser temperature in the 78°–110°K region. The center frequency of the laser emission is controlled by the cold head temperature. Each diode is temperature tunable over about 100 cm^{-1}.

Ultimate detection sensitivity of a TDL sensor will vary with strength and clarity of the absorption feature. The LN_2 temperature diode used for the methane sensor described here was operated in the IR spectral region between 3000 and 3025 cm^{-1}. This region corresponds to the C-H antisymmetric streching band and therefore there are strong methane lines present.

The laser beam is collimated into the 2-cm-diameter, 1.4-m-long sample cell. The beam is then split and focused onto the 1 mm × 1 mm elements of a sample and a reference detector. Both sample and reference detectors are Peltier-cooled H_g C_d T_e IR detectors (EG&G Judson). The 2.5-cm reference gas cell contains a known concentration of the absorber gas.

Dynamic response is obtained by allowing the atmospheric samples to flow through the sample cell at low pressure (i.e., 35 mbar). The small cell volume (approximately 0.4 L) allows the use of a relatively compact rotary vacuum pump to achieve exchange times of less than 1 s. For the eddy correlation system a pump with a pumping capacity of 5 L s^{-1}, at the pump, was used to achieve system data acquisition rates of the order of 10 Hz.

A 11.3-m sample inlet tube (i.e., polyethylene-lined polyvinyl chloride (PVC)) composed of contiguous sections (i.e., 7-m section of 6.35-mm ID plus a 4.3-m section of 4.76-mm-ID tube) of increasing diameter was designed and experimentally verified for plug flow. A calculated sample delay time of 0.25 s for flow through the tube was also verified experimentally. The sample temperature was equilibrated to the sample cell temperature through a 1.5-m long, 7.5-mm ID-soft copper coil.

For in-field use the laser chassis is housed in an aluminum enclosure 2 m × 0.5 m × 0.5 m. The enclosure provides protection from the elements and allows the control of the laser's ambient environment.

The laser system's operating electronics are integrated with a 286 AT bus PC. PC integration allowed for software control of the digital signal processing, laser function, and the real-time display of the laser-operating characteristics. Data logging was carried out external to the host computer.

The NOWES CH_4 instrument had a total system noise of 5 ppbv rms with an effective bandwidth of 0 to 1.6 Hz. For a covariance calculated over a 30-min period, a resolution of 0.1 ppbv is achieved. The maximum sampling rate is 6.6 Hz and data point averaging time is 0.15 s. These instrument specifications were sufficient for the measurement of methane flux at the 0.1 $\mu g\ m^{-2}\ s^{-1}$ level using the eddy flux technique.

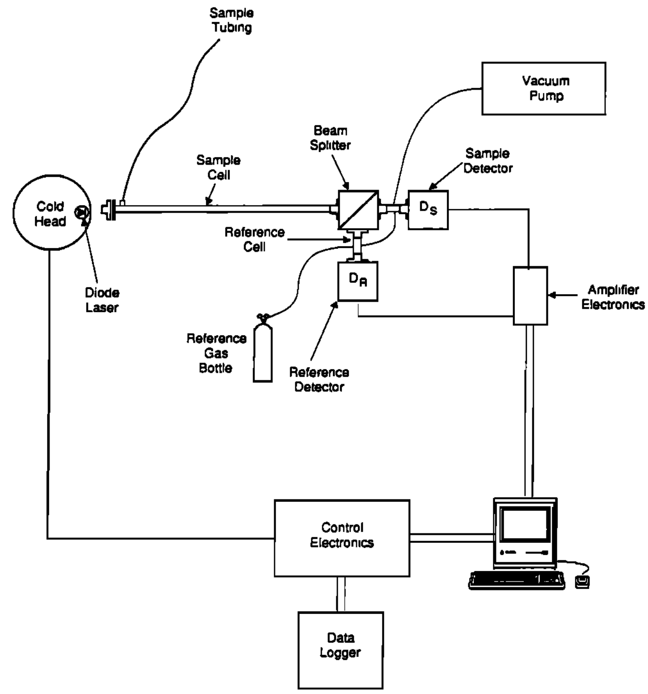

Figure 1. Tunable diode laser methane sensor schematic.

4. Results and Discussion

The TDL methane sensor facilitated the continuous measurement of the methane exchange between the atmosphere and the bog at the Kinosheo Lake tower site during the summer 1990 intensive study. Data retrieval were disturbed only during times when laser calibration and LN_2 filling (i.e., 10 min) were carried out or during power failure and severe weather events. Approximately 900 half-hour average methane flux measurements were obtained.

Data were selected out of the larger set based on defined wind-direction windows for acceptable data and when wind speeds were less than 1 m s^{-1} resulting in a reduced data set for analysis of 538 half-hour average methane flux measurements. These were sufficient data to construct diurnal and temporal trends and a statistically reliable average flux rate.

Average Emission

The overall average emission, using all data (i.e., 538 half-hour averages), was calculated to be 0.18 $\mu g\ CH_4\ m^{-2}\ s^{-1}$ with a standard deviation of 0.26 (16 mg $CH_4\ m^{-2}\ d^{-1}$). The data were normally distributed and the standard error on the mean is approximately 6%.

The average emission measured by the tower-based method compares well with the spatial average of 16 mg $m^{-2}\ d^{-1}$ [*Roulet et al.*, this issue] for the same wetland determined using enclosure data for the period July 11 to 26, 1990. The spatial average constructed from the enclosure data is a weighted average taking into account the frequency distri-

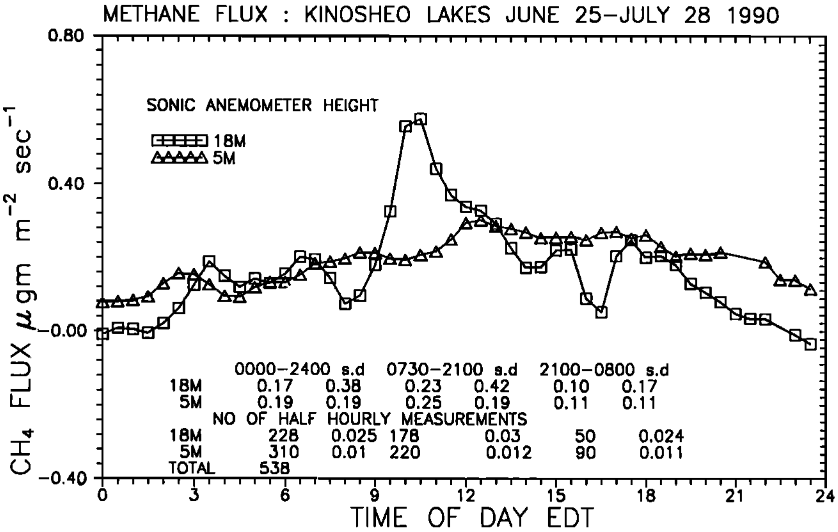

Figure 2. Diurnal trend constructed for the 5-m and 18-m measurement heights from the half-hour average flux measurements obtained from June 25 to July 28, 1990.

bution of the microtopographical elements derived from high-resolution color air photography taken at the same site where the micrometeorological measurements were made. However, the extrapolation modeling technique used to construct the overall average from the enclosure data represented a wetland area larger than the tower sample. The tower data also compares well with the spatial average of 22 $\mp$ 16 mg m^{-2} d^{-1} derived from the aircraft measurements [*Ritter et al.*, this issue] for the Kinosheo area during July.

The fluxes measured at the tower site are approximately 4 to 10 times smaller than expected, based on the work of *Crill et al.* [1988] and *Moore et al.* [1990], but are comparable to the methane emission from low boreal bogs, as measured by *Roulet et al.* [this issue]. They are also 10 times smaller than those measured by *Verma et al.* [1992] using a TDL methane sensor and eddy correlation over the bogs in north central Minnesota that *Crill et al.* [1988] sampled. These differences in the measured fluxes may be due to effects of temperature and nutrient limitations [*Dise*, 1991].

The average emission rate measured at Kinosheo is lower than was predicted [*Schiff and Barrie*, 1988] for wetlands in this region. Overall, the entire Hudson Bay lowland (HBL) turned out to be 5 to 15 times lower than was expected [*Roulet et al.*, this issue] based on the global wetland estimates of *Mathews and Fung* [1987], *Aselmann and Crutzen* [1989], and *Fung et al.* [1991]. However, the tower-based method, the enclosures, and the aircraft all confirm the low flux rates. The significance of these lower emission rates and the role of the HBL in terms of the global methane budget are discussed by *Roulet et al.* [1992].

Diurnal Trend Analysis

The diurnal trend was constructed by selecting half-hour bin intervals over 24 hours and averaging all points in each of the 48 bins. Bin populations were not entirely uniform, possibly introducing some bias. To compensate, smoothing techniques were applied to the diurnal trend after it was constructed. This involved applying a running three point weighted average to the data with the center weight set to 0.5. Figure 2 presents the results of the diurnal trend data analysis. Trends are presented for measurements taken at the 5-m and 18-m levels separately. The standard deviation, number of half-hour average data points, and the standard error of estimate on the mean are tabulated in Figure 2.

On average, significant differences between the upper and the lower measurement height fluxes are not observed. A weak diurnal trend is observed in the data with a daytime maximum and an early morning minimum. The average daytime (i.e., 0730 to 2100 EDT (eastern daylight time)) flux was calculated as 0.24 μg m^{-2} s^{-1} $\mp$ 0.3 (20 mg m^{-2} d^{-1}) and the average nighttime (i.e., 2100 to 0800) as 0.11 μg m^{-2} s^{-1} $\mp$ 0.14 (9 mg m^{-2} d^{-1}). The emission of methane from the surface is controlled by many parameters and processes controlling its production and emission. These include meteorological (i.e., emission footprint, turbulent processes, etc.), ecosystem structure, temperature, and *p*H [*Andreae and Schimel*, 1989]. Many of these parameters, for example, temperature at depth in the peat, would display only small variation over the diurnal period. The observed weak diurnal trend is likely related to small changes in these controlling processes over the same period.

The biogenic production of methane results in significant storage of methane in the upper layers of the bog. Early into the experiment this was evidenced during tower anchor work. Figure 3 shows a 10-min time series of methane concentration measured at the 5-m tower level. During this period, digging to relocate the tower anchor resulted in significant methane emission. Peaks of methane concentration of approximately 0.55 parts per million by volume (ppmv) above ambient levels were observed by the sensor upwind. This example, albeit extreme, points out that other phenomena that disturb the bog surface layer, such as pressure fluctuations due to turbulence and rain, can lead to methane release to the atmosphere.

The top data shown in Figure 2 were characterized by larger standard deviations than those at the 5-m level and showed a significant excursion from the general trend at 1030. Both these features observed in the data are explained by the differences in the fetch seen by the two measurement heights. In general, fetches seen by the 5-m measurement

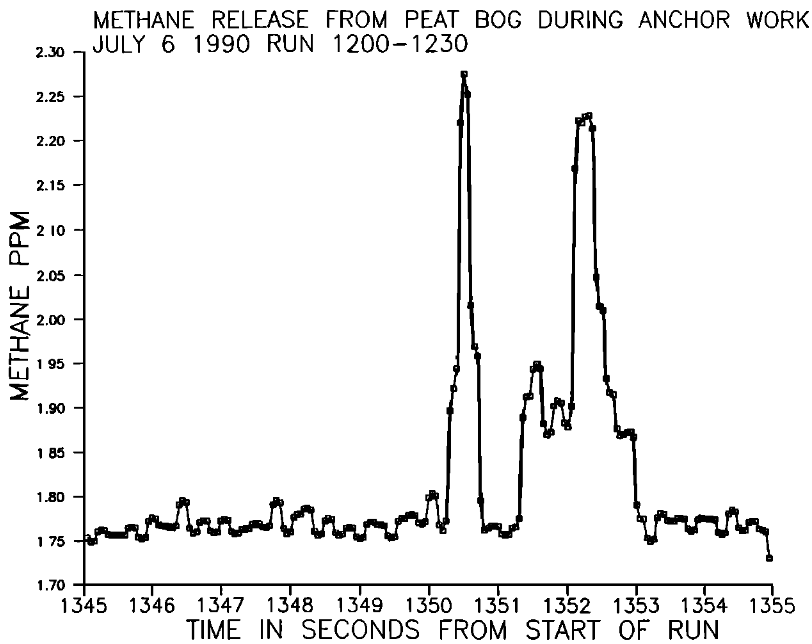

Figure 3. A methane release from the peat bog during tower anchor work.

height, as shown by LANDSAT-TM imagery and air photography of the site, are more uniform with wind direction and contain drier elements. The fetches seen by the 18-m height are more heterogeneous and contain a larger fraction of wet area. The shallow water depressions and small ponds had a 50% greater flux than many of the vegetated portions of the Kinosheo peatlands, as measured by *Moore et al.* [this issue] using enclosures and as estimated by *Hamilton et al.* [this issue] using measured surface water concentrations and a simple transfer model. The peak seen at 1030 in the top data is a result of several measurements in this time bin that by coincidence the wind direction was aligned with high emitting areas.

To further investigate these phenomena, the fetch was divided into wind direction intervals of 10° and a sector average flux calculated using all data. An estimate of the maximum downwind extent of the emission footprint was made using the data of *Leclerc and Thurtell* [1990]. The fraction of each wind sector out to 2 km that was wet area was then estimated using the LANDSAT-TM imagery data available for the site [*Roulet et al.*, this issue]. This is a course estimate as the LANDSAT-TM used has a pixel size of 30 × 30 m and many of the water bodies are smaller than this and therefore will not be resolved. The percentage wet area per sector derived by this method and the sector average flux were plotted and are shown in Figure 4. A strong relationship is seen between the two parameters.

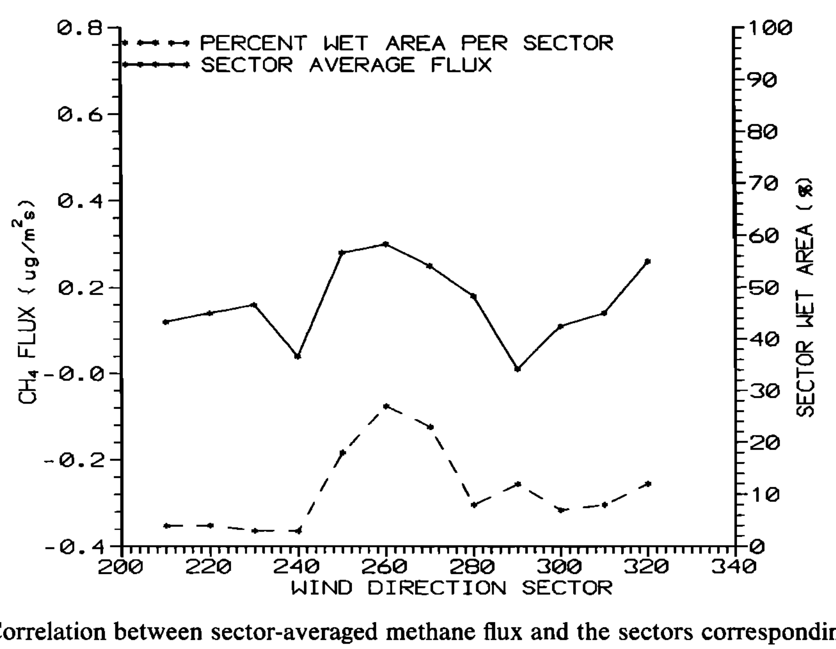

Figure 4. Correlation between sector-averaged methane flux and the sectors corresponding percentage wet area.

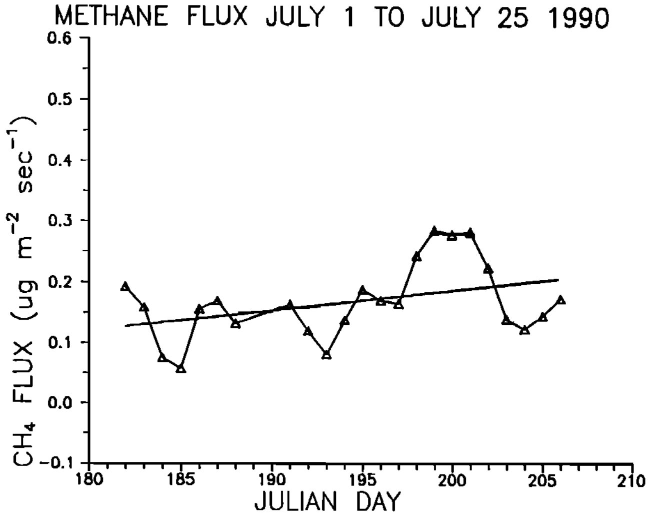

Figure 5. Seasonal trend constructed from the daily average methane flux calculated for the measurement period, June 25 to July 28, 1990.

Temporal Trend in Fluxes During the Experimental Period

The temporal trend was determined by calculating the daily average methane flux using all data. Where there were insufficient data, a daily average was not calculated. To account for slight variation in the number of points available to construct the daily average, the temporal trend was digitally smoothed by the same method applied to the diurnal trend described above. Figure 5 shows the observed trend. A best fit linear regression line was applied to the data and is also shown in Figure 5. The slope of the regression (i.e., 0.0033) indicates a slight increase in the emission over the month of July.

The production and emission of methane from wetlands has been shown to be strongly related to temperature [*Baker-Blocker et al.*, 1977; *Crill et al.*, 1988; *Moore and Knowles*, 1987; *Moore et al.*, 1988; *Roulet et al.*, 1992]. Over the tower study period, continuous measurements of bog temperature at several depths in both dry and wet areas were taken. These data are reported by *den Hartog et al.* [this issue]. To evaluate the effect of temperature on emission, the seasonal trends were correlated with bog temperature measurements. The best correlation was found between the seasonal trend and the bog temperature measured in the wet area at 20-cm depth or lower. This correlation is illustrated by Figure 6, where the 20-cm bog temperature (wet) is superimposed onto the seasonal trend. The undulations observed in the temperature data reflect the diurnal variation of temperature. The data clearly show that wet bog temperature at depth is a controlling parameter for the methane emission over the measurement period at Kinosheo Lake.

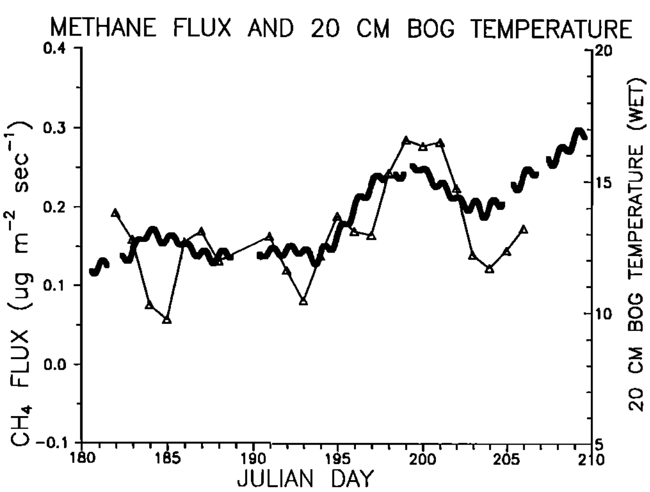

Figure 6. The correlation between the seasonal variation of the methane flux and the corresponding daily average bog temperature measured at 20-cm depth (i.e., dark trace) in a wet area.

5. Concluding Remarks

A sensitive fast response, fieldworthy methane sensor, based on tunable diode laser absorption spectroscopy, has been successfully developed for the measurement of the exchange of methane between a surface and the atmosphere using a tower-based eddy correlation technique. It has been proven to be suitable for the reliable continuous measurement of the methane flux. This represents a significant advancement in TDL techniques for the detection of the fluxes of trace gases.

There were sufficient methane flux data obtained to meet the experiment objectives of providing information on diurnal and temporal trends and to allow intercomparison with enclosure and aircraft measurements. The extensive data base of methane flux and supporting measurements collected provides considerable opportunity for further analysis.

Fluxes averaged over the study period showed an overall methane emission of 16 mg m^{-2} d^{-1} with a daytime average of 20 mg m^{-2} d^{-1} and a nighttime average of 9 mg m^{-2} d^{-1}. The effect of emission footprint was evident in the data. A strong relationship between the daily average methane flux and the bog temperature (wet) at 20-cm depth was observed.

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