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An Intercomparison of Two Tunable Diode Laser Spectrometers Used for Eddy Correlation Measurements of Methane Flux in a Prairie Wetland

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ABSTRACT

An intercomparison was made between two tunable diode laser spectrometers used to measure methane fluxes by the eddy correlation technique at a prairie wetland site. The spectrometers were built by Unisearch Associates Inc. of Concord, Ontario, Canada, and Campbell Scientific Inc. of Logan, Utah, and were models EMS-50 and TGA-100, respectively. The fluxes were found to agree very well with each other in the range of 0 to 42 mg m^{-2} h^{-1}.

The TGA-100 was observed to exhibit offset drifts. Most of the time, when the offset was only slowly changing (as compared to the eddy correlation averaging time), these drifts did not affect the calculated fluxes. There were times, however, when the offset changed fast enough to have a noticeable affect on the fluxes.

The EMS-50 also exhibits some drifting of the measured concentration. It was, however, much slower and of smaller amplitude than the drift seen in the TGA-100.

1. Introduction

In the summer of 1993, the Center for Laser Analytical Studies of Trace Gas Dynamics (CLAS) conducted a field experiment in a midlatitude, prairie wetland near Valentine, Nebraska, to measure methane and carbon dioxide fluxes using the eddy correlation method (e.g., Businger 1986). In this method, the flux of a scalar quantity (e.g., methane, carbon dioxide, water vapor, heat, etc.) is determined by calculating the covariance of the fluctuations of the concentration of the scalar with the fluctuations of the vertical component of the wind velocity

$$\text{Flux}, \qquad F = \overline{w'c'}, \tag{1}$$

where w' is the deviation from the mean value of the vertical wind velocity w, c' is the fluctuation from the mean value of the scalar concentration c, and the overbar denotes a time average. It should also be noted that this method yields a flux that is a spatial average of an area (or footprint) that is a function of the height of the sensors above the mean canopy level. The methane fluxes were calculated using concentration values obtained from a pair of tunable diode laser spectrometers (TDLS). One goal of this experiment was to intercompare the field performance of these two closed path TDLSs (Campbell Scientific Co. model TGA-100 and Unisearch Associates Inc. model EMS-50). While the underlying principles of infrared absorption spectroscopy apply to both machines, the optical implementation and the signal processing techniques used in the two instruments are quite different.

TDLSs have been used for the measurement of at-

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mospheric trace gases for over a decade (Hastie et al. 1983; Thurtell et al. 1991; Verma et al. 1992; Shurpali et al. 1993; Edwards et al. 1994); however, to our knowledge, there has not been a direct intercomparison of two different TDLSs used to measure fluxes by the eddy covariance technique. By comparing eddy flux measurements from these two machines (which represent the most commonly used closed path types), some of the relative merits of the two designs for eddy correlation flux measurements have been demonstrated.

2. Site description

The experiment was carried out in late July and early August at a wetland site in the Nebraska Sandhills near Valentine (42°52′N, 100°33′W, elev 788 m). The site was located in the Ballards Marsh Wetlands and was situated on the north shore of a large marsh. The marsh surface was totally submerged with a standing water depth of about 0.3–0.6 m. The water had a pH of 7.17 and a conductivity of 460 μmho $(\text{cm})^{-1}$. The dominant plant species were *Phragmites australis* (2–3 m tall) and *Scirpus acutus* (1–2 m tall). There were several small areas of open water that constituted less than 20% of the total marsh area. Under the water there was a heavy mat of litter that was about 0.1–0.3 m thick. Underlying the litter was sediment consisting of plant roots and rhizomes, sand, clay, and mineral soil.

The eddy correlation tower was located at the southern end of a 65-m boardwalk that was floated on Styrofoam pontoons and anchored to the north shore of the marsh. At the end of this boardwalk, a stable platform was bolted to steel pipes fixed into the mineral soil. A 5-m-tall scaffold tower was erected on top of this platform and guyed into the mineral soil for stability.

3. Eddy correlation instruments

All three components of the wind velocity were measured with an Applied Technologies Inc. model SWS-211/3Sx 3D sonic anemometer, water vapor concentrations were measured with a Campbell Scientific model KH20 krypton hygrometer, temperatures were measured with fine-wire thermocouples, and carbon dioxide concentrations were measured with a Li-Cor model 6262 IRGA (infrared gas analyzer). The eddy correlation instruments or their sample collection tubes were mounted on a bar attached to a rotatable plate that could be pointed directly into the wind to provide an adequate (0.5–2 km) fetch between 45° and 315° (northeast-northwest). The plate was bolted to the end of a retractable sled mounted on the southern end of the scaffold tower. When the sled was extended to its operating position, the instrument bar and the instruments were located about 3 m above the mean canopy top and about 5 m above the water surface. Air samples were delivered to the TDLSs and the Li-Cor IRGA through stainless steel and Teflon tubing. The intake tubes were arranged in a cluster around the 3D sonic anemometer such that there was no more than a 20-cm separation between any sensor inlet and the anemometer. Stainless steel tubing (0.775 cm i.d.) was used for the long straight runs along and down the tower. Sections where flexure, motion, or sharp bends were required were made of Teflon-lined, UV-resistant bevaline (0.610 cm i.d.). In addition, there was approximately 2 m of a copper tube (0.787 cm i.d.) inside the TGA-100 case to damp out temperature fluctuations in the gas sample. The total tube lengths (from sample collection point to absorption cell) were 16.6 m for the TGA-100 instrument and 14.0 m for the EMS-50 instrument. Both sample delivery systems were leak checked by plugging the inlet and noting the ultimate vacuum that could be maintained in the tube.

The TDLSs were placed on separate platforms, fixed into the mineral soil, near the main tower. The EMS-50 was housed in an insulated and air conditioned 1.9 m $\times$ 1.1 m $\times$ 1 m hut. The TGA-100 was housed in a 2.0 m $\times$ 0.5 m $\times$ 0.5 m fiberglass box. This box was insulated and weatherproof but was not actively cooled or heated. A plywood sun screen (covered with reflective Mylar) was built over the TGA-100 box to minimize instrument heating, and the tall vegetation also helped screen the instrument from wind and the sun. Data signals from all of the instruments except the TGA-100 were sent through a pair of multiconductor, shielded cables to a data acquisition system housed in a semitrailer, outfitted as a field laboratory, at the end of the boardwalk on the northern shore. The TGA-100 data were sent back to the trailer through a fiber optic link to a controlling microcomputer in the laboratory. This microcomputer generated an analog signal that was input to the data acquisition system mentioned above.

The data acquisition system consisted of an IBM-compatible microcomputer with 16, 16-bit analog input channels. Each signal was preconditioned before conversion. This circuit consisted of an isolation amplifier and an 8-pole Butterworth low-pass filter (12.5-Hz half power point). The signals were digitized at 25 Hz and recorded on a write-once-read-many (WORM) optical drive. The data collection program also calculated and reported on-line concentration averages, standard deviations, covariances, fluxes, and other statistics using approximate calibration and time delay information. These statistics were calculated for 30-min averaging periods where 30 min was deemed sufficient to include most of the low-frequency spectral information contributing to the fluxes. Electrical power was supplied to the site from a local feeder line.

4. Instrument description

The two TDLSs employed in this study were commercially built, and both used closed path absorption cells. At the time of the intercomparison, neither of the TDLSs were fitted with gas driers. Instead, a correction factor (for water vapor-induced changes in air density)

was calculated and applied to the raw fluxes. Some of the physical specifications of the instruments are presented in the appendix.

a. Unisearch Associates Inc. model EMS-50

The older of the two instruments was custom built in 1990 by Unisearch Associates Inc. of Concord, Ontario, Canada (Sigrist 1994), and is shown schematically in Fig. 1. It consists of a small Stirling cycle cooler used to maintain the laser diodes at their operating temperature (near 95 K), a multiple-pass absorption chamber, miscellaneous focusing and beam steering optics, and associated detectors, amplifiers, and data collection electronics. The lasers employed in this instrument are double heterostructure devices produced at the Fraunhofer Institute of Freiburg, Germany, in 1988. Mercury-cadmium-telluride (HgCdTe) photodetectors are used in the photo-resistive mode to detect the laser light. These detectors are thermoelectrically cooled by two stage Peltier stacks and temperature controlled at −40°C for improved noise and sensitivity. The absorption cell is a modified form of the White-Horn-Pimental type (Horn and Pimental 1971) and has a total pathlength of 53 m. This is achieved in an aluminum cell with a base path of 68 cm by reflecting the beam back and forth 78 times. This long pathlength increases the total amount of absorption to a more easily measured level. The temperature and pressure of the gas in the cell are actively controlled and thus remain constant during the measurement period. The laser current (and thus its wavelength) is modulated (small amplitude) at 25 kHz, and the outputs of the detector preamplifiers are sent to a pair of lock-in amplifiers that are referenced to twice the modulation frequency (50 kHz) for second harmonic detection (Reid and Labrie 1981). This results in outputs that are approximations to the second derivatives of the true absorption signals. This technique removes any linear background signal from the laser and has a higher detection sensitivity. In operation, the laser wavelength is locked to the center of the absorption feature by using the zero crossing point of a first harmonic signal (which is an approximation of the first derivative of the absorption profile) as the input to an error amplifier that applies small corrections to the laser current. This signal is generated by a third lock-in amplifier, referenced directly to the modulation frequency (25 kHz). The ''line lock'' signal and a concentration reference signal are obtained from a fraction of the laser beam that passes through a sealed reference cell containing 0.25% methane in nitrogen at a pressure of about 80 mbar (60 Torr). This also serves as a standard absorption level against which the sample signal is compared and provides a method of ensuring that an absorption feature is indeed due to methane. The peak levels of these absorption signals are averaged, ratioed, corrected for fluctuations in the laser output power (as determined directly from the preamplifiers, by using a mechanical chopper to define the ''zero light'' condition), and converted to a concentration (mixing ratio) by multiplying by a previously determined ''calibration'' factor (which will be discussed later).

b. Campbell Scientific Inc. model TGA-100

The newer instrument was built by Campbell Scientific Inc. of Logan, Utah, and was delivered in April 1993. It is shown schematically in Fig. 2. The laser used in this spectrometer is also a double heterostructure device, made by Laser Photonics of Andover, Massachusetts, in 1992. Like the EMS-50, the TGA-100 uses HgCdTe photoresistive detectors to detect the laser light. These detectors (like the EMS-50 detectors) are thermoelectrically cooled (three stage Peltier stacks) and temperature controlled. The detectors chosen for the TGA-100 have a longer wavelength cutoff than the EMS-50 ones and can be operated at temperatures as low as −60°C. The TGA-100 uses a much shorter absorption path than the EMS-50. The absorption cell consists of a single pass through a 1.5-m-long tube. Instead of relying on a long pathlength to achieve a large absorption signal, the TGA-100 uses a short pathlength and sophisticated digital noise reduction techniques to recover its much weaker absorption signal (Thurtell et al. 1991). The philosophy for this approach is that each reflection in a traditional, long path White cell decreases the signal-to-noise ratio of the instrument. By decreasing the number of sample cell reflections, even though the total absorption is smaller than in a multipass cell, the signal-to-noise ratio is comparable to that of a long path White cell. In the TGA-100 design, the detector signals are directly digitized, and the technique of derivative spectroscopy (as used in the EMS-50) is not employed. This simplifies the electronic design but transfers the task of noise reduction and signal recovery to software.

The laser current is ramped in such a way that the TDL wavelength sweeps (linearly) through the absorption feature. This wavelength sweep allows the data collection software to use the integrated absorption intensity instead of just the peak absorption level (as in our EMS-50) to calculate the concentration. The advantage of this is a reduction of the instrument's sensitivity to absorption line broadening effects. The actual laser current waveform is more than a simple ramp. It is, instead, a ramp preceded by two brief dc current levels. These levels are set by the user and are used to determine the zero absorption signals from the detectors and to linearize the laser wavelength during the ramp phase.

The TGA-100 uses a short reference cell as a ''standard'' against which the sample signal is compared. In contrast to the EMS-50, this is an ''open'' or ''flowing gas'' reference cell that requires a source of reference gas (about 2% CH_4 in dry nitrogen) to flow through the cell at a rate of 30–50 sccm (standard cubic centimeter

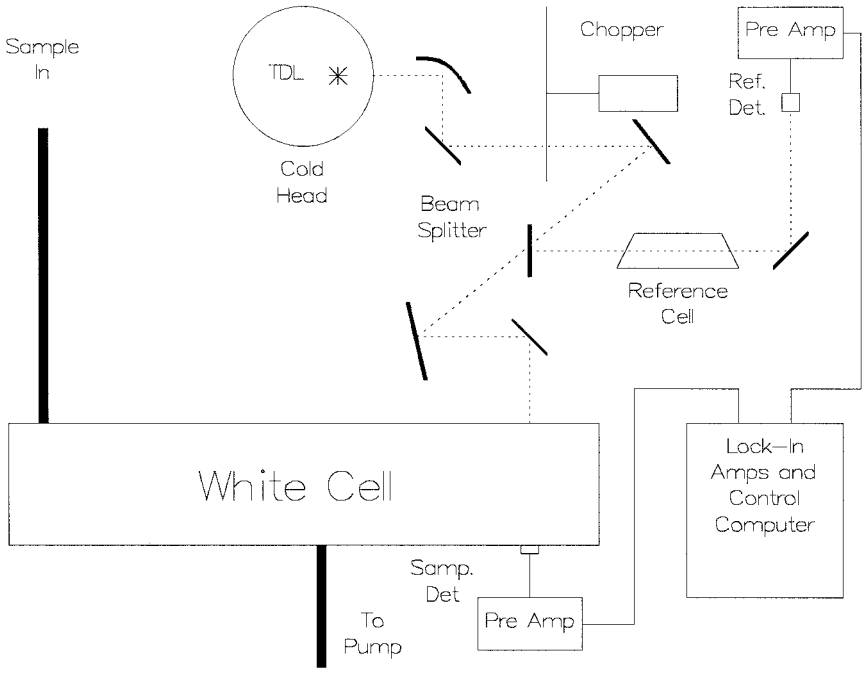

FIG. 1. Schematic view of the Unisearch Associates Inc. EMS-50 TDLS.

per minute). The reference cell outlet is connected to the vacuum pump line in parallel with the absorption cell outlet. In this design, the absolute pressures of the reference cell and the sample cell fluctuate together, so their ratio remains unchanged. In theory, this eliminates the need to control the sample cell pressure actively.

The laser is cooled in a liquid nitrogen dewar and is temperature controlled at its operating point between 84 and 110 K. While this strategy is more logistically complicated than a Stirling cycle cooler for field use, it has the advantage that there are no mechanical vibrations that could couple to the laser and appear in the absorption signals, it is cheaper, and the site power requirements are slightly reduced.

Both instruments operate at flow rates and pressures as recommended by the manufacturers [25 slm (standard

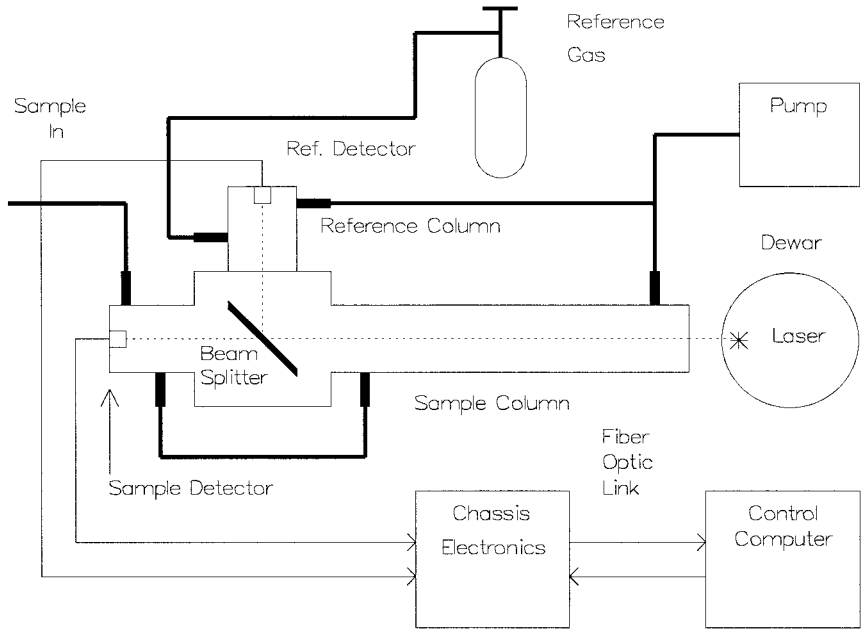

FIG. 2. Schematic view of the Campbell Scientific Inc. TGA-100 TDLS.

liters per min) and 5.3 kPa for the EMS-50, and 18 slm and 7.5 kPa for the TGA-100]. These give sample cell residence times of about 110 ms for the EMS-50 and about 60 ms for the TGA-100. This (and the averaging time) effectively determines the upper-frequency response of the instrument. Both lasers are operated at temperatures and currents that give output frequencies near 3018 cm^{-1}. The exact operating conditions for each laser are chosen to give the lowest noise level when measuring a standard gas.

5. Calibration and setup procedures

a. EMS-50

To calibrate the EMS-50, the zero was first set by tuning the laser away from all absorption features and adjusting the lock-in outputs to read exactly zero. This effectively forced the instrument to correctly read zero for no methane in the sample cell. It should be noted that the second harmonic absorption profiles exhibited a very flat background that was assumed to be the level corresponding to zero absorption. Laboratory tests have shown that this procedure correctly determines the zero point to within about 10 ppbv. Once the zero point was established, the laser was tuned to a strong methane absorption feature and line locked to the peak. A calibration gas mixture of 1.6 ppmv methane in dry air (Matheson Gas Products, either certified standard grade or primary standard grade) was passed through the White cell, and a calibration subroutine was executed. While the calibration gas was flowing, the calibration subroutine compared the optical absorption in the White cell to that in the reference cell and calculated a calibration coefficient from this ratio and the known concentration of the calibration gas. Since the pressure and temperature of the gas in the White cell were the same during calibrations as they were during data collection, this calibration coefficient could then be used to calculate the methane concentration of an unknown gas in the cell. The calibration gas was usually run at a flow rate between 3 and 5 slm and at a pressure of 5.3 kPa. Previous experiments had shown that the calibration was not dependent on flow rate to the levels that we were able to measure. This technique effectively provided two points (zero concentration and approximately 2 ppmv) from which the gain of the instrument was calculated.

b. TGA-100

By design, the TGAS software (supplied with the TGA-100 by Campbell Scientific) used the known concentration of the reference gas as a calibration factor when calculating the concentrations of unknown samples. In operation, the user was to first enter the concentration of the reference gas as a parameter to the TGAS control program. After this step, the user was to adjust four different laser current parameters that controlled 1) the dc current level, 2) the ramp ''span,'' 3) the ''over-current'' pulse level (used to keep the laser warm), and 4) the low current pulse (used to determine the zero absorption level). The user's manual supplied with the TGA-100 quantitatively described how to set up the dc level, the span level, and the low current level. The over-current setting procedure was described as ''. . . adjust the Hi current (overdrive) parameter to obtain the waveshape which most closely approximates a ramp at the beginning and end points with an absorption line in the middle.'' We found that we could achieve this condition over a broad range of parameter settings. Further, we found that the measured concentration (after accounting for the measured offset, and while using a high-grade, calibrated reference gas) varied by several hundred parts per billion volume between the extremes of this range. In addition, because of the expense of obtaining a high-quality reference gas (in addition to the high-quality calibration gas needed for the EMS-50), we felt it necessary to develop a suitable procedure to quantitatively set the laser current parameters and to recalibrate less expensive uncalibrated tanks of the reference gas mixture.

A two-point setup procedure that was (in principle) similar to the calibration procedure used with the EMS-50 instrument was developed. In this procedure, the TGA-100 was first set up according to the user's manual as carefully as possible. The approximate reference gas concentration was then entered into the TGAS program. Next, the zero level was established. This was done by flowing pure nitrogen gas through the sample cell. In theory, there should be no absorption signal from this condition; however, because of optical interference fringes, a residual signal could often be observed. To the eye (and to the control software), this ''fringe'' signal appeared as a small absorption (or emission) peak. This zero (or offset) signal was often of the order of several hundred parts per billion volume. Once the offset level was established, a span gas (Matheson Gas Products primary standard grade, 1.6 ppmv) was introduced into the sample cell, and the concentration level was again noted. The measured zero level was then subtracted from this span level, and the difference was compared to the known concentration of methane in the span gas. Then, the ratio of these two values (the known concentration and the difference) was used to adjust either the concentration parameter of the reference gas or the high current parameter. If the ratio was very far from 1, the reference concentration was adjusted. If the ratio was close to 1, only the laser high current parameter was adjusted. The process was iterated until consistent results were obtained. It was found that the final reference concentration used was usually within 1% or 2% of the value marked by the gas vendor, and that most adjustments were made to the high current parameter. The tanks of reference gas were mixed locally and their cost was 1/5 to 1/6 of the analyzed,

primary standard grade gas. The above procedure was used when a new tank of calibration gas was installed. In more routine, daily operation, the reference concentration was not adjusted, only the high current parameter.

Since this technique was developed in the field, its validity was later verified in the laboratory. A test was run where the TGA-100 was set up with this method, and then four gases with different methane concentrations (0, 1490, 1620, and 1880 ppbv) were measured. This test showed that the instrument consistently and correctly measured the four different concentrations to within the accuracy of their analysis.

6. Experiment

The intercomparison data were gathered between 4 and 7 August 1993. Because the TGA-100 was delivered only 2 months prior to this time, a tested operating procedure for the TGA-100 was not in place and had to be developed in the field. Further, the EMS-50 was committed to be part of a study in central Saskatchewan and had to be moved early in August. For these reasons, the intercomparison period was relatively short.

To characterize the instruments individually, the fixed methane concentration of compressed air (from a tank) was measured for 5-min periods, and the rms deviation from the mean concentration was calculated. We called this value the noise level of the instrument. This was done in the laboratory and in the field at each calibration. This value was used as an indicator of the general ''health'' of the instrument.

A similar measurement was done in the field to estimate the error in flux values. Dry air (from a tank) was fed into a TDLS (instead of ambient air) while the regular data collection program operated. Fluxes were calculated from this dataset and, since there should be no correlated fluctuations in the methane concentrations, the rms average of these residual fluxes was considered to be the ''flux noise'' or the ''minimum detectable flux'' of the combined system (TDLS and anemometer).

Both instruments were set to average for 0.1 s, so the high-frequency cutoff points should be similar for both TDLSs. The instruments were set up and calibrated in the morning (usually at about 0800 LT). The calibrations were checked at 1200 and again at 1800 LT. Since calibrations could not be made often enough to correct offset drifts, this schedule was chosen to maximize the number of flux measurements.

Since the actual TDLS sensors were separated from the sampling point (where the wind velocities were measured), there was a time lag or delay between the TDLS signals and the anemometer signals. The delay times were functions of the sample tube lengths, tube diameters, and the flow rates. While the delay times could be calculated from these quantities, we found that measured values were more accurate.

Delay times were measured in two different ways. First, they were mechanically measured by releasing a burst of nitrogen from an electrically operated solenoid valve near the tube inlet and measuring the time from the solenoid pulse to the first response from the TDLS as observed on an oscilloscope. In the second method, delay times were measured statistically by computing the ''circular correlation coefficient'' between the methane concentration fluctuations and the vertical wind speed fluctuations and using a technique described by Chahuneau et al. (1989). The delay times obtained from both methods agreed well; however, those obtained from the ''circular correlation'' calculation were assumed to be more precise.

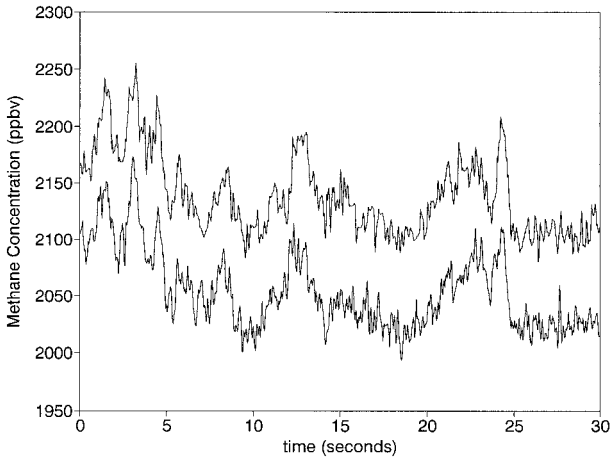

FIG. 3. Atmospheric methane concentration levels as measured by both TDLSs on 5 August 1993 at about 1430 LT. The top trace is from the TGA-100 and the bottom trace is from the EMS-50.

The data were reprocessed off-line where corrections were applied for tube attenuation (Moore 1986; Suyker and Verma 1993), sensor separation, true delay time, and the true calibration of the instruments. Since in-line driers were not used with either TDLS, additional corrections (Webb et al. 1980) were applied for density changes in the gas sample due to water vapor.

After removing half-hour periods that were known to contain invalid data (unacceptable wind velocity or direction, malfunctioning instruments, etc.), there were 59 acceptable half-hours of simultaneous flux measurements.

7. Results

Noise level measurements (as described earlier) showed that both instruments performed virtually identically. The noise levels measured in the laboratory were 5 to 10 ppbv for both TDLSs, while noise levels measured in the field ranged from 5 to 20 ppbv for both TDLSs.

Figure 3 shows the instantaneous concentrations obtained from both instruments for a typical period of about 30 s. The data are from the middle of the day on 5 August 1993 and have been time shifted to account

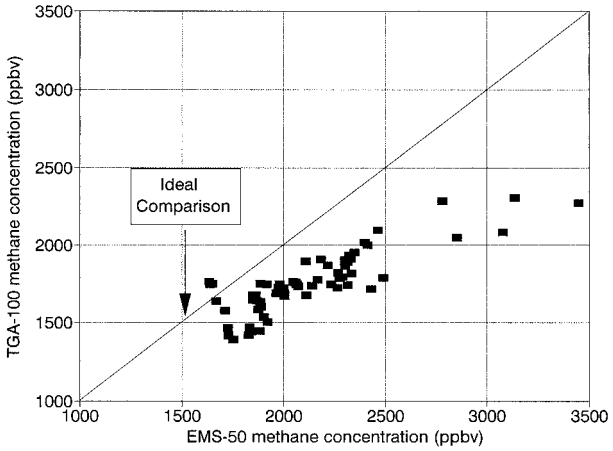

FIG. 4. TGA-100 mean methane concentrations plotted against the EMS-50 mean concentrations for the intercomparison period (3–7 August 1993).

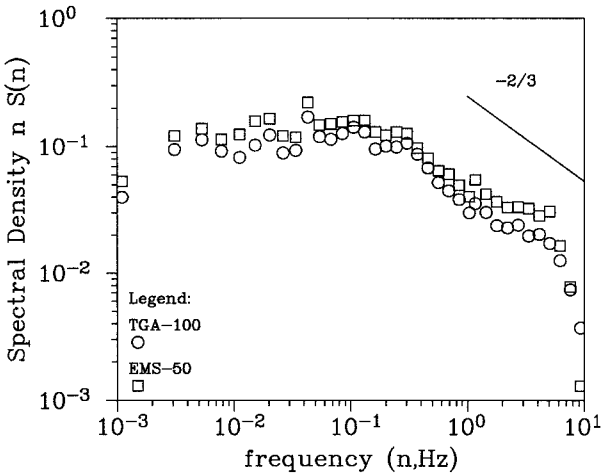

FIG. 6. Power spectra of both TDLSs for the half-hour between 1200 and 1230 LT on 5 August 1993. Open circles are from the TGA-100, and open squares are from the EMS-50. The straight line indicates a slope of −2/3.

for the different sample delay times of the two instruments.

Mean concentration data are plotted in Fig. 4. While these data do not fall on the ideal 1:1 line, they do generally fall on a line parallel to it, but with considerable scatter. That is to say they have a slope of about 1.

Measurements of the minimum detectable flux showed that both instruments had flux noise levels between 0.2 and 0.6 mg m^{-2} h^{-1}. Flux data are shown in Fig. 5. These data fall very close to the ideal line, with little scatter. A linear regression fit to the data of Fig. 5 gives a slope of 1.003 and an intercept of 0.842 mg m^{-2} h^{-1}, with an R^2 coefficient of 0.944. The flux values observed span a large range from almost 0 mg m^{-2} h^{-1} to almost 42 mg m^{-2} h^{-1}.

Figure 6 shows typical frequency spectra of the methane concentrations as measured by the EMS-50 and the TGA-100, and Fig. 7 shows the corresponding cospectra with vertical wind velocity. These data are representative of the entire intercomparison period, and it should be noted that there were no significant differences in the spectra and cospectra between the two instruments. As can be seen in the cospectra, both instruments seem to have frequency cutoffs of about 3 Hz.

8. Discussion

Figure 3 shows that while there was an offset of about 100 ppbv between them, the two instruments appeared to track each other quite well. Also, the magnitudes of the fluctuations were approximately the same, indicating that there was no significant gain difference between the two instruments. This also confirms that the two TDLSs were indeed sampling the same air parcel.

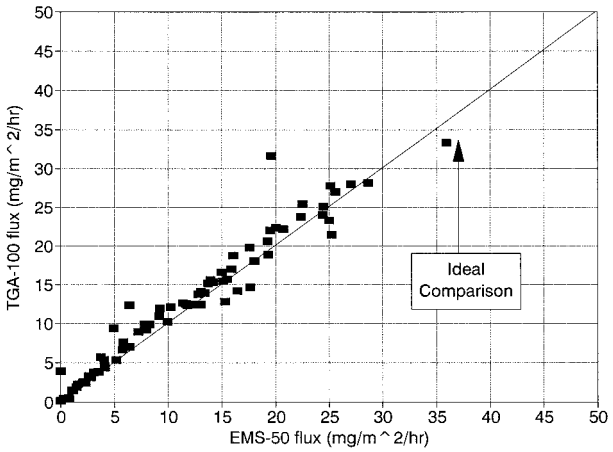

FIG. 5. TGA-100 methane fluxes plotted against the EMS-50 fluxes for the intercomparison period (3–7 August 1993).

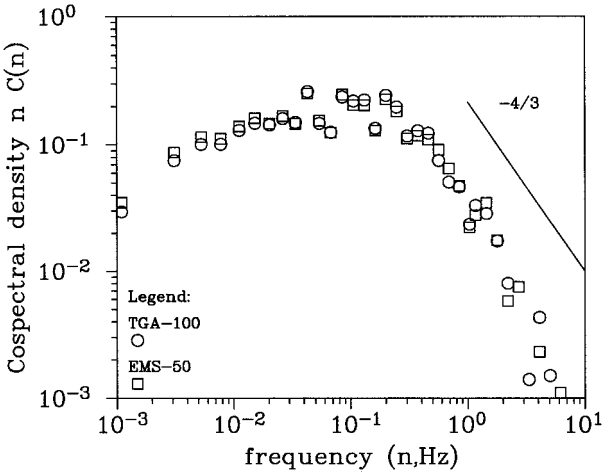

FIG. 7. Cospectra of both TDLSs with the vertical wind velocity for the half-hour between 1200 and 1230 LT on 5 August 1993. Open circles are from the TGA-100, and open squares are from the EMS-50. The straight line indicates a slope of −4/3.

The concentration data of Fig. 4 show considerable scatter and deviation from the ideal 1:1 line. We believe that this is mainly due to offset differences between the two instruments. This view is supported by the flux data shown in Fig. 5. Since fluxes calculated from the measured concentrations are not sensitive to *constant* offsets, the excellent agreement in the fluxes suggests that the discrepancy in concentrations is indeed due to relatively constant offset differences between the two instruments.

The offset between the two instruments is believed to be mainly in the TGA-100 and is probably due to optical interference fringes (Hernandez 1986). The origin of these fringes (known as Fabry–Perot fringes) is the constructive and destructive interference of coherent light, which is reflected from parallel surfaces in the optical path. There are many surfaces that could contribute to this effect (lenses, the beam splitter, detector windows, etc.), and it is impossible to identify a single interferometer ''cavity.'' The only definite conclusion is that the cavity formed has a very low contrast and finesse, as would be expected for uncoated optical surfaces. This results in a fringe amplitude that is comparable to the sample absorption line strength in the TGA-100. The movement of the fringes (and thus the changes in the offset) is the result of the changing cavity length caused by temperature changes. Since the transmission function of a Fabry–Perot cavity is a periodic function of the cavity length, this effect appears as a roughly sinusoidal modulation of the concentration for a linear temperature change. The period of this modulation is dependent on the rate of temperature change of the sensitive optical element or elements, and usually ranges from minutes to hours. A somewhat more serious situation was observed when the instrument temperature did not vary linearly.

The EMS-50 also has a multitude of optical surfaces that could form interferometer cavities, and indeed there is no reason to think that this effect is not present in that instrument too. The effect, however, is much less pronounced in the EMS-50. Because of the long, folded pathlength, the total absorption is much greater than in the short pathlength TGA-100. The fringe strength, on the other hand, will not increase unless both of the optical surfaces are between the White cell mirrors, causing multiple passes through the cavity. Multiple passes through the cavity would increase the finesse and the contrast, resulting in a larger fringe signal (Hernandez 1986). Fringes generated before the White cell will suffer the same reflection losses at the mirrors as the rest of the laser signal. Multipassing is apparently not the case in the EMS-50 sample cell. The few times that fringes were observed in the EMS-50, their relative amplitudes (compared to the absorption signals) were very small, indicating no multipassing. We have calculated that the total absorption in the EMS-50 is between 25 and 30 times that of the TGA-100. Assuming also that the total signal contributed by the interference fringes

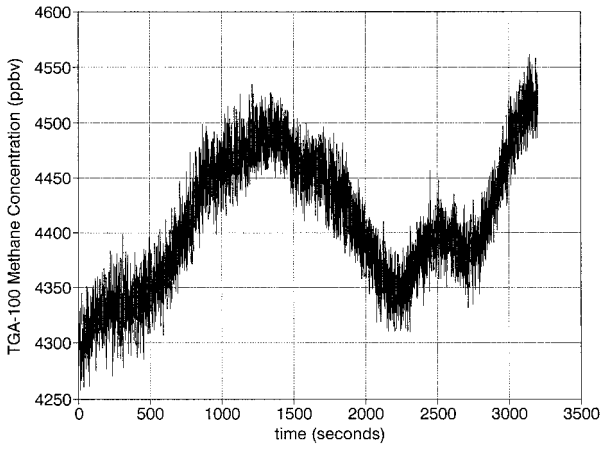

FIG. 8. Methane concentrations obtained from the TGA-100 on 14 September 1993 at about 1230 LT, showing the effects of optical interference fringes. The measurements were made on gas from a tank of compressed air.

(similar contrasts) is the same in both instruments, the EMS-50 fringe effect will only be 3%–4% of that observed in the TGA-100 (or on the order of 1 to 10 ppbv). This level is small enough to be largely masked by other noise sources in the EMS-50.

We note that while interference fringes will produce an offset error in any instrument of this nature, this offset may or may not affect flux values. The fluxes will not be affected by a *constant* offset, but a changing or drifting offset could have an effect. Since the eddy covariance method relies on the temporal correlation between vertical wind fluctuations and trace gas density fluctuations, and on averaging out any ''accidental'' correlations, it is tempting to assume that any contribution to the flux from drifting fringe offsets will average to zero. This will, however, not always be the case. Assuming a sinusoidal fringe offset, these error terms will only average to zero if there are many periods during the eddy correlation averaging period. When the fringes are moving slowly (periods between 10 and 100 min), there may only be one or two accidental correlations, and the statistical canceling will be incomplete. More importantly, the changing fringe offset will produce a bias in the calculated mean concentration value. When this corrupted mean is used to calculate density fluctuations, significant errors can result. In extreme cases, the errors can be orders of magnitude larger than the actual fluxes.

During the intercomparison period, we were fortunate that no serious episodes of fringing were observed. Later in the field season (after the EMS-50 had been moved to another project), serious fringing was observed on several occasions. One instance is illustrated in Fig. 8. The data were obtained on 14 September 1993 at about 1230 LT. At that time the TGA-100 was measuring gas from a compressed air tank. It is obvious to the eye that subtracting a simple mean value from the instantaneous

concentrations would produce very large errors in the resulting concentration fluctuations.

While more sophisticated nonlinear detrending methods might be used to remove the effects of the drifting fringes, these all require major assumptions to be made about the size and shape of the fringe contributions. This practice could lead to the accidental elimination of real flux components, especially in the low-frequency region, and introduce a systematic bias to the calculated fluxes. Because of this, we used a simple moving average method to calculate the mean values.

While the EMS-50 does not suffer significantly from interference fringes, it is susceptible to another thermal problem. As stated previously, the sample cell of the EMS-50 is a modified White–Horn–Pimental resonator (Horn and Pimental 1971). In this design, the incoming light is reflected back and forth between two mirrors. These mirrors are curved to refocus the light on each pass through the cell. The mirror separation is critical to the precise refocusing of the beam. If the mirrors are slightly too far apart or too close together, the focus will be off for each pass. The focus error in the output beam spot that this causes is cumulative, and since there are 78 passes through the cell, a small error will be magnified greatly (Yariv 1985). As the mirror separation drifts away from its optimum condition, the spot size of the final image on the detector element will change. This spot size change results in a changing signal, as seen by the data collection program. These mirror separation changes are caused by the thermal expansion and contraction of the cell body in response to ambient temperature changes. This appears in the data as a fast or slow drift of the measured concentration, the speed of which depends on the rate of temperature change.

To minimize this effect, the EMS-50 sample cell is equipped with heaters and a temperature controller that allow the temperature to be held constant to within about 0.02°C. The temperature is usually set between 28° and 31°C. This represents the maximum usable temperature range for the sample cell. The ambient temperature, however, could often exceed this level, and to compensate for this, the EMS-50 shelter was also temperature controlled. Problems with temperature stability were encountered, however, when the doors to the shelter were opened for extended periods. This caused ''focus drifting'' of the EMS-50, which would normally stabilize within 2 h. To minimize this effect, the doors of the shelter were opened only when absolutely necessary (usually only during calibration periods), and even then, they were only cracked open as much as necessary to manipulate the instrument controls. This procedure minimized the focus drift error to between 1% and 3% of the ambient concentration. This situation could be improved greatly if more remote control were incorporated into the EMS-50.

The similarity of the frequency responses of the instruments (as shown in Figs. 6 and 7) is no surprise. The only factors contributing to this are the sample cell volume, the flow rates, the working pressures of the sample cells, and the averaging times. These factors were all chosen to give similar performances in both instruments.

9. Conclusions

Through this intercomparison, we have determined that when both of these TDLSs are operating at their optimum, they yield very similar performances. This is evidenced by the similar noise levels and minimum detectable fluxes, and the good agreement found between simultaneous flux measurements.

Our biggest concern is with the interference fringes that appeared intermittently in the TGA-100. While these could be effectively dealt with by fast, intermittent switching to pure nitrogen gas in a gradient flux system, this approach is not feasible for an eddy correlation system. The drifting fringes therefore pose a problem for eddy correlation flux measurements. We believe that more effective thermal management of the TGA-100 enclosure (by active cooling and heating) would help by either eliminating the fringes or slowing their drift down to the point where they fall outside the eddy flux frequency regime.

Similarly, there is some concern about the focus drift of the EMS-50. We believe that a greater degree of remote control over this instrument would minimize the problem. By leaving the EMS-50 enclosure closed and undisturbed during routine operation procedures, this problem would be ignorable.

Both instruments do have several desirable features. The EMS-50, with its closed cycle Stirling cooler, has the potential to run unattended (with an automated calibration system) for weeks at a time. This is very desirable for year-round studies in remote locations. The TGA-100, on the other hand, needs operator attention every 8 to 12 h to fill the liquid nitrogen dewar. It is, however, more portable and can be moved to remote locations much more easily than the EMS-50. The smaller volume of its absorption cell (less pumping capacity required) combined with the liquid nitrogen cooling give the TGA-100 a definite advantage in power requirements.

In summary, both of these instruments have demonstrated similar performance in an eddy covariance system and can both be operated successfully for this purpose at remote field sites.

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APPENDIX
TDLS Specifications.

EMS-50 TDLS specifications

Sizes and weights		
Main unit	84 × 76 × 84 cm	~77 kg
Power supply	43 × 51 × 25 cm	~23 kg
UPS (Sola)	23 × 41 × 48 cm	~39 kg
Vacuum pumps (×2)	48 × 28 × 31 cm	~21 kg
Stirling cooler (in main unit)		~4 kg
Power consumption		
Main unit	120 V @ 2 A 1 phase (from UPS)	
Vacuum pumps (× 2)	120 V @ 10 A 1 phase (each)	
Stirling cooler	120 V @ 1 A 1 phase (from UPS)	
Capacities		
Total pumping speed (2 vacuum pumps)	30 slm	
White cell volume	1.4 L	
White cell total path length	53 m	
Reference cell path length	8 cm	
Reference cell gas	0.25% CH_4 in N_2	
Reference cell pressure	~8.0 kPa	
Minimum laser temperature	~80 K–90 K	
Maximum laser current	1000 mA	
CH_4 concentration noise (0.1 s)	5–10 ppbv	
Minimum detectable CH_4 flux	0.2–0.6 mg m^{-2} h^{-1}	
Minimum averaging time	60 ms	
Maximum averaging time	30 000 ms	
Typical operating conditions		
White cell pressure	5.3 kPa	
Sample flow rate	25 slm	
White cell time constant	110 ms	
Averaging time	100 ms	

TGA-100 TDLS specifications

Sizes and weights		
Main unit	211 × 48 × 5 cm	~75 kg
Busch pump	48 × 28 × 31 cm	~21 kg
Power consumption		
Main unit	12 V @ 5 A dc (from a storage battery)	
Busch pump	120 V @ 10 A 1 phase	
Capacities		
Total pumping speed	18.5 slm	
Sample cell volume	0.4 L	
Sample cell total length	160 cm	
Reference cell volume	0.02 L	
Reference cell length	5 cm	
Reference cell gas	2.0% CH_4 in N_2	
Dewar LN_2 capacity	1.5 L	
LN_2 use rate	3–5 L day^{-1}	
Minimum laser temperature	82 K	
Maximum laser current	1000 mA	
CH_4 concentration noise (0.1 s)	5–20 ppbv	
Minimum detectable flux	0.2–0.6 mg m^{-2} h^{-1}	
Typical operating conditions		
Sample cell pressure	7.5 kPa	
Sample flow rate	~18 slm	
Sample cell time constant	60 ms	
Averaging time	100 ms	
Reference gas flow rate	30–1000 sccm	

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