

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

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Evaluating the performance of commonly used gas analysers for methane eddy covariance flux measurements: the InGOS inter-comparison field experiment

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

The performance of eight fast-response methane (CH_4) gas analysers suitable for eddy covariance flux measurements were tested at a grassland site near the Cabauw tall tower (Netherlands) during June 2012. The instruments were positioned close to each other in order to minimize the effect of varying turbulent conditions. The moderate CH_4 fluxes observed at the location, of the order of $25 \text{ nmol m}^{-2} \text{ s}^{-1}$, provided a suitable signal for testing the instruments' performance.

Generally, all analysers tested were able to quantify the concentration fluctuations at the frequency range relevant for turbulent exchange and were able to deliver high-quality data. The tested cavity ring-down spectrometer (CRDS) instruments from Picarro, models G2311-f and G1301-f, were superior to other CH_4 analysers with respect to instrumental noise. As an open-path instrument susceptible to the effects of rain, the LI-COR LI-7700 achieved lower data coverage and also required larger density corrections; however, the system is especially useful for remote sites that are restricted in power availability. In this study the open-path LI-7700 results were compromised due to a data acquisition problem in our data-logging setup. Some of the older closed-path analysers tested do not measure H_2O vapour concentrations alongside CH_4 (i.e. FMA1 and DLT-100 by Los Gatos Research) and this complicates data processing since the required corrections for dilution and spectroscopic interactions have to be based on external information. To overcome this issue, we used H_2O mole fractions measured by other gas analysers, adjusted them with different methods and then applied them to correct the CH_4 fluxes. Following this procedure we estimated a bias on the order of $0.1 \text{ g (CH}_4\text{) m}^{-2}$ (8% of the measured mean flux) in the processed and corrected CH_4 fluxes on a monthly scale due to missing H_2O concentration measurements. Finally, cumulative CH_4 fluxes over 14 days from three closed-path gas analysers, G2311-f (Picarro Inc.), FGGA (Los Gatos Research) and FMA2 (Los Gatos Research), which were measuring H_2O vapour concentrations in addition to CH_4 , agreed within 3% ($355\text{--}367 \text{ mg (CH}_4\text{) m}^{-2}$) and were not clearly different from each other, whereas the other

BGD

11, 797–852, 2014

Evaluating the performance of EC CH_4 analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



instruments derived total fluxes which showed small but distinct differences ($\pm 10\%$, 330–399 mg (CH₄) m⁻²).

1 Introduction

Methane (CH₄) is the third most important greenhouse gas, after water (H₂O) and carbon dioxide (CO₂). Due to its high global warming potential of 25 (at the 100 yr horizon) changes in its abundance have an effect on the on-going climate change. The total global methane source is thought to be relatively well-quantified, but the relative contributions of each source and changes in individual sources are not (Forster et al., 2007). The gradual increase in atmospheric methane concentration observed over the last century decreased in the past decades and came to a standstill around 2000 for several years before starting to increase again since 2007 (Dlugokencky et al., 2011). Explanations for the temporary standstill are inconsistent with each other, highlighting the need for improved bottom-up inventories to understand better the most important factors controlling the changes in atmospheric methane (Heimann, 2011). Several mechanisms have been proposed to explain the temporary decline in the growth rate, such as (a) decreases in anthropogenic emissions (e.g. Dlugokencky et al., 1994), (b) decreases in anthropogenic emissions before 1999, and decreases in wetland emissions after that (Bousquet et al., 2006) or (d) changes in the removal rate by OH (e.g. Monteil et al., 2011). A network of eddy covariance (EC) towers can provide direct and continuous monitoring of ecosystem scale surface fluxes that can be upscaled to landscape and continental scales using process-based models. This combined approach can help unravelling the relative contribution of different sources and sinks on global methane budget.

Over the last 20 yr, thanks to advancements in laser absorption spectroscopy (LAS), several methane gas analysers suitable for eddy covariance measurements have been developed. Field applications of the first generation of methane EC analysers were limited due to the need for cooling lasers and/or detectors to cryogenic temperatures,

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

[Title Page](#)

[Abstract](#)

[Introduction](#)

[Conclusions](#)

[References](#)

[Tables](#)

[Figures](#)

[⏪](#)

[⏩](#)

[◀](#)

[▶](#)

[Back](#)

[Close](#)

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)



and they also had relatively high flux detection limits (e.g. Fowler et al., 1995). This made long-term measurements challenging and time consuming and inhibited measurements at remote locations which are often important methane sources, such as remote wetlands in Siberia and North America. After development of gas analysers that can use peltier cooling systems, measurements became possible at these remote locations, although availability of mains power remained an issue. Recently, a new generation of commercial fast-response CH₄ sensors has become available which offers a much improved signal/noise ratio and is even easier to use due to stable operation. Consequently there are large efforts to create continental scale networks of measurement stations (e.g. Integrated Carbon Observation System (ICOS) and Integrated non-CO₂ Greenhouse gas Observing System (InGOS)) allowing for CH₄ fluxes to be measured continuously. In this context, it is important to characterise the currently available CH₄ analysers and assess their performance. A few inter-comparison studies exist (Detto et al., 2011; Peltola et al., 2013; Tuzson et al., 2010), but none of them included such a wide range of models currently available on the market as this study.

This study reports results from an inter-comparison experiment held at a Dutch grassland site in June 2012 as part of the InGOS (Integrated non-CO₂ Greenhouse gas Observing System) EU-FP7 project. The objective of this study was to evaluate the field performance of the instruments tested, to determine the accuracy and precision of the measured fluxes and to assess the relative merits of different instrumental designs (e.g. open vs. closed path gas analysers). This presentation of the results addresses the following topics in detail: (i) precision of CH₄ measurements from each instrument; (ii) an analysis of the impact of corrections due to density fluctuations (WPL) and spectroscopic effects on the fluxes; (iii) an evaluation of the errors arising when external H₂O signal is used for the WPL and spectroscopic corrections of the CH₄ flux and (iv) the consistency in total accumulated CH₄ emissions during a 14 day period. To this effect, eight methane gas analysers, including models by Picarro Inc., Los Gatos Research, LI-COR Biogeosciences and Aerodyne Research, were set up to measure side-by-side for 22 days.

2 Experimental setup

2.1 Site

The gas analyser inter-comparison experiment was performed at the Cabauw Experimental Site for Atmospheric Research (CESAR) (51°58'12.00" N, 4°55'34.48" E, -0.7 m a.s.l.). The Cabauw tall tower combines a comprehensive set of observations to profile many aspects of the atmospheric column (www.cesar-observatory.nl). A broad range of atmospheric and ecological measurements are conducted on a continuous basis. The site is located in an agricultural landscape, with methane emissions originating from ruminants and other agricultural activities, but also from the peaty soil and the drainage ditches between the surrounding fields. The soil consists of a 0.5 to 1 m deep clay layer on top of a peat layer which is several meters deep. The water table level is on average 0.5 m below the surface. For a more thorough site description, see e.g. Beljaars and Bosveld (1997) and van Ulden and Wieringa (1996).

During the campaign, the vegetation surrounding the measurement tower was low (0.05–0.2 m) although a taller maize field was located 70 m away in SW–W direction from the measurement mast (see Fig. 1). The landscape is relatively flat and level, making the area ideal for micrometeorological flux measurements. The eddy covariance measurement tower was approximately 87 m away from the nearest building, the Cabauw tall tower (height 213 m), located to the NE. The fetch in the main wind direction (W–SW) was free from any large obstacles for several hundreds of metres.

Meteorological conditions during the campaign were recorded next to the measurement mast by a weather station operated by the Royal Netherlands Meteorological Institute (KNMI). Air temperature during the campaign ranged between 4.3 and 22.0 °C with an average of 14.9 °C. Rain was recorded on 19 out of 22 measurement days, 21 June being the rainiest day with a total of 14.1 mm of rain. Cumulative rainfall during the whole campaign was 63.1 mm, which is close to the climatological mean for the site for this period. Relative humidity ranged between 43 % and 100 %, with an average of 78 %. The campaign was thus held in relatively cool and moist conditions.

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2.2 Measurement system

Eddy covariance flux measurements were conducted between 6 and 27 June 2012 at a 6.5 m high tower. Two sonic anemometers were used (both USA-1, METEK, Germany) to acquire fast measurements of wind velocity components and sonic temperature. Similar instruments were used in an effort to minimize the potential systematic bias on CH₄ fluxes caused by the use of different types of anemometers. The anemometers were placed on the same tower at the same height with approximately one meter horizontal distance in NW–SE direction between them (see Fig. 1).

All data were logged at a frequency of 20 Hz by a central computer located in a trailer that also housed the closed-path analysers. The trailer was approximately 26 m away from the measurement tower in the NE direction. The trailer was air conditioned and the indoor temperature was kept around 22 °C. Data logging software capable of handling and saving all 56 variables at 20 Hz was written in LabView for this campaign (National Instruments, USA).

The gas analysers were divided between the two anemometers which will from now on be referred to as METEK1 and METEK2. Two open-path analysers (Models LI-7500 and LI-7700, LI-COR Biogeosciences, USA) were situated on both sides of METEK1, displaced in NW–SE direction by approximately 0.3 m. The LI-7500 measured CO₂ and H₂O mole densities, while the LI-7700 measured CH₄. Two separate LI-7700 analysers were used sequentially and the switch between instruments was done at 19th of June. The LI-7700 is a low-power, lightweight methane gas analyser with a 0.8 m long open measurement cell. Its operation principle is based on laser absorption spectroscopy, more precisely wavelength modulation spectroscopy (McDermitt et al., 2010). The LI-7700 reports CH₄ molar density (mmol m⁻³) while all the other CH₄ analysers report mole fraction (ppm). Unfortunately the data were recorded with three decimals (e.g. 0.088 mmol m⁻³) and thus the resolution of the LI-7700 data was limited to 0.001 mmol m⁻³ (about 20 ppb); for the other instruments a resolution of 0.001 ppm was used. Because of this oversight the CH₄ fluxes from the LI-7700 instrument were

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



more attenuated and noisier than normal (cf. Fig. 5h below). This data logging problem should be kept in mind for the interpretation of the results of this study.

In addition to the two open-path devices, three closed-path gas analysers (G2311-f, Picarro Inc., USA; FGGA, Los Gatos Research, USA; DLT-100, Los Gatos Research, USA) were sampling from METEK1 (the sampling setup of all gas analysers is described in Table 1). The G2311-f is a new gas analyser by Picarro Inc. and is based on cavity ringdown spectroscopy (CRDS). The analyser is able to make simultaneous fast measurements of CO₂, CH₄ and H₂O mole fractions. The whole sampling line connected to the G2311-f was heated in order to prevent condensation of water on the tube walls. The FGGA (“Fast Greenhouse Gas Analyser”) developed by Los Gatos Research is based on off-axis integrated cavity output spectroscopy (OA-ICOS) and the instruments provide fast measurements of CO₂, CH₄ and H₂O mole fractions on a continuous basis. The FGGA tested is the standard (rack-mount) version rather than the later introduced enhanced performance version with more accurate temperature control. The third closed-path analyser accompanying METEK1 was a DLT-100, an older benchtop model (production year: 2005) by Los Gatos Research. It is based on the same measurement principle as the FGGA (OA-ICOS); however, it is able to measure and report mole fractions only of CH₄. Until 16th of June the instrument measured reported data at 1 Hz and after that the sampling frequency of this instrument was set to 10 Hz. This was taken into account when spectral corrections were applied (Sect. 4.3.1.).

Five closed-path gas analysers sampled from near METEK2. Four of them measured CH₄ and one only CO₂ and H₂O. These four methane analysers were: a G1301-f (Picarro Inc., USA), a QCL (Aerodyne Research Inc., USA) and two FMAs (Los Gatos Research, USA). Henceforth, the two rackmount FMAs are referred to as FMA1 and FMA2. FMA2 was taken back to the laboratory for cleaning in the middle of the campaign (between 19th and 20th of June) and after the cleaning operation the instrument measured only with 2 Hz until the end of the campaign. G1301-f is an older model by Picarro Inc. The measurement principle is the same as in G2311-f and the instrument is

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



capable of measuring any two out of three gases (CH₄, CO₂ and H₂O) simultaneously. During this campaign the instrument was not able to measure H₂O and thus CH₄ and CO₂ were selected. The QCL is an older (pulsed) quantum cascade laser by Aerodyne Research Inc. The detector was cooled with liquid nitrogen using an automated LN₂ filling system. During this campaign, the QCL measured both CH₄ and N₂O. A Perma Pure drier was connected to the QCL sampling line in order to remove H₂O from the air samples. The Fast Methane Analyser (FMA) is a slightly older model by Los Gatos Research using OA-ICOS. The standard FMA instrument is able to measure and report only CH₄ mole fractions but it can be updated to measure also H₂O. While FMA1 was a standard CH₄ instrument, FMA2 was upgraded to enable the parallel H₂O measurements. The LI-7000 (LI-COR Biogeosciences, USA) was used to measure CO₂ and H₂O from the vicinity of METEK2. These water measurements were needed for the corrections applied to some of the CH₄ fluxes (Sect. 3).

3 Data post-processing

3.1 Introduction

Under the assumptions of turbulence stationarity, horizontally homogeneous surface and turbulence characteristics, the mass balance equation is reduced into a simple form which is the basis of eddy covariance measurements:

$$F_c = \frac{\overline{\rho_d}}{M_d} \overline{w' r'_c} \quad (1)$$

Where F_c is the flux of gas c at the surface, $\overline{\rho_d}$ is mean dry air density, M_d is the molar mass of dry air, w the vertical wind component and r_c is the molar mixing ratio of gas c . Following Reynolds decomposition, overbars denote mean values and primes ($'$) fluctuations around the mean. However, usually gas analysers do not report gas concentration relative to dry air (i.e. dry mixing ratio, r_c), rather they report gas

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



concentration relative to moist air (i.e. mole fraction, χ_c), or in the case of open-path analysers the gas mass density (ρ_c), which is affected by fluctuations in temperature and humidity. In addition, the instruments used to measure gas concentrations and wind components are non-ideal, i.e. they do not make truly instantaneous measurements and therefore act as bandpass filters effectively filtering out certain high and low frequencies in the signal. Both of these effects need to be corrected, in addition to other post-processing steps, before the measured flux represents the flux at the surface. The correction methods used in this study are discussed in greater detail in the following sections.

3.2 General data post-processing steps

Data obtained during the campaign was post-processed with EddyUH software (freely available at http://www.atm.helsinki.fi/Eddy_Covariance/index.php). The post-processing steps followed generally adopted methods by Aubinet et al. (2000) and they were organized as follows:

- First the CH₄ time series were converted with calibration parameters (Table 2) from measured ppm (mmolm^{-3} for LI-7700) to calibrated units. The parameters were obtained with the method described in Sect. 3.3.
- The raw eddy covariance data were despiked by comparing two temporally adjacent CH₄ concentration measurements. If their difference was larger than 3 ppm the following point was considered a spike and was replaced with the value of the previous data point. For LI-7700 also data points were considered as spikes, for which the diagnostic value indicated by the instrument was 32 768 (instrument not ready), 16 384 (no laser signal detected) or 8192 (reference methane signal not locked).

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- For closed-path instruments which measured also H₂O the effect of fluctuating humidity on methane concentration measurements were corrected point-by-point according to Eq. (4) below, with coefficients given in Table 3.
- The coordinate system of the sonic anemometer was rotated using the double rotation method (Rebmann et al., 2012), aligning the x-axis of the anemometer with the mean flow.
- Linear detrending was used to separate the turbulent signal from the measurements, and the covariances were calculated by searching the maximum of the cross-covariance function within certain predefined lag windows. A 30 min averaging period was used for all fluxes.
- Within an iterative loop, the rest of CH₄ fluxes were corrected for the effect of fluctuating humidity and also for fluctuating temperature for open-path LI-7700 (Sect. 3.4). Temperature fluctuations are expected to be fully damped by the time the sample reaches the closed-path instrument.
- Finally, the fluxes were corrected for high and low frequency damping, based on the method described by Aubinet et al. (2000).

The overall correction factor CF used to correct for band-pass filtering was calculated as

$$CF = \frac{\int_0^{\infty} C_{\text{mod}}(f) df}{\int_0^{\infty} TF_{\text{LF}}(f) TF_{\text{HF}}(f) C_{\text{mod}}(f) df} \quad (2)$$

where TF_{LF} is a transfer function describing low frequency dampening, adopted from Rannik and Vesala (1999), TF_{HF} is a transfer function which describes high frequency damping, C_{mod} is a scalar model cospectrum and f is natural frequency. C_{mod} was

ing the instrument which caused a sudden decrease in the cavity ringdown time and consequently a slight change in calibration.

3.4 Corrections for density and spectroscopic effects

Humidity and temperature fluctuations affect gas flux measurements by causing changes in air density (Webb et al., 1980; Massman and Tuovinen, 2006; Ibrom et al., 2007) and, for gas analysers based on laser absorption spectrometry, also by altering the shape of gas absorption line (McDermitt et al., 2010; Neffel et al., 2010; Tuzson et al., 2010). Corrections for these effects are referred to as density and spectroscopic correction, respectively.

Rella (2010) proposed to correct both of these effects for closed-path gas analysers by using a second order polynomial function

$$\chi_c = r_c \left(1 + a\chi_v + b\chi_v^2 \right) \quad (4)$$

where χ_c is the molar fraction of gas c , r_c is the mixing ratio of gas c corrected for any interference from water, χ_v the water vapour molar fraction and a and b are instrument specific coefficients which describe the dependence of χ_c on χ_v . This expression has been demonstrated to be suitable for correcting measurements for dilution and spectroscopic effects in several studies (Chen et al., 2010; Hiller et al., 2012; Nara et al., 2012; Rella et al., 2013), and it is nowadays widely used. It is worth noting that Eq. (4) is reduced to the correction for dilution alone (i.e. density correction for closed-path gas analysers) for $a = -1$ and $b = 0$. This form of correction is easily applicable if the gas analyser measures both, χ_c and χ_v , and in fact is implemented into some of the instruments that can thus report a dry mixing ratio (e.g. G2311-f, which uses coefficients from Chen et al., 2010). However, some of the laser spectrometers used in this study did not measure water and thus external water measurements were needed to correct the gas measurements. When using external water measurements, it may be preferable to apply the corrections to the half-hourly averaged fluxes, instead of correcting the raw data point-by-point, since this way the possible discrepancies between

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the external and internal χ_v are not passed on to the high frequency χ_c time series. By using Reynolds decomposition, averaging and slightly reorganizing the terms, the expression above can be converted into flux form (see Appendix A for derivation):

$$F_c = \frac{\bar{\rho}_a}{M_a} \frac{1 - \bar{\chi}_v}{1 + a\bar{\chi}_v + b\bar{\chi}_v^2} \left(\overline{w' \chi'_c} + \frac{\bar{\chi}_c}{1 - \bar{\chi}_v} \overline{w' \chi'_v} - \frac{a + 2b\bar{\chi}_v - b\bar{\chi}_v^2 + 1}{(1 + a\bar{\chi}_v + b\bar{\chi}_v^2)(1 - \bar{\chi}_v)} \overline{\chi_c w' \chi'_v} \right) \quad (5)$$

$$= \frac{1 - \bar{\chi}_v}{1 + a\bar{\chi}_v + b\bar{\chi}_v^2} (F_c^{\text{RAW}} + F_c^{\text{WPL}} + F_c^{\text{SPECT}})$$

where $\bar{\rho}_a$ is the mean total air density and \bar{M}_a is the mean molar mass of moist air. The first term on the right hand side is the measured flux (F_c^{RAW}), the second term is the effect of air density fluctuations (F_c^{WPL} , H₂O term in WPL correction) and the third term (F_c^{SPECT}) and the multiplier in front of the parentheses arise from the spectroscopic effects that water vapour has on the measurements of gas c .

One should note that Eq. (5) is identical to Eq. (3a) in Ibrom et al. (2007) if we take $a = -1$ and $b = 0$, meaning that the spectroscopic effects are neglected, and only the effect of density fluctuations is corrected. In addition, it should be emphasized that in normal circumstances the two correction methods presented above (Eqs. 4 and 5) will deliver the same result, if and only if the water vapour covariance, $\overline{w' \chi'_v}$, reflects the variation in water vapour concentration in the measurement cell when gas c is measured, a point which has been highlighted for density correction (Ibrom et al., 2007; Massman, 2004). This applies also to spectroscopic correction and to the combined correction of Eq. (5).

The coefficients a and b used in correcting data from different gas analysers are listed in Table 3. For FMA2 and FGGA the correction was done point-by-point applying Eq. (4) with the water vapour concentration measured within the instrument; for some of the instruments with no in-situ measurement of water vapour (i.e.

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



G1301-f and DLT-100), the correction was performed using Eq. (5), and the water vapour covariance, $w' \chi'_v$, was adjusted using the empirical procedure described below (Sect. 3.4.1). For FMA1 it was performed using Eq. (5) and the water vapour covariance from the LI-7000, $w' \chi'_v$, calculated with FMA1 CH₄ lag time because LI-7000 and FMA1 shared the same inlet line. CH₄ signals from the G2311-f and the QCL were free from H₂O interference since the G2311-f applied a similar correction internally during the measurements and the QCL was connected to a drier. Open-path gas analyser LI-7700 methane measurements are not only affected by the water vapour concentration, but also by temperature fluctuations, and these resulting CH₄ fluxes were corrected with the method proposed by McDermitt et al. (2010).

3.4.1 Using external H₂O in correcting CH₄ fluxes

Adsorption/desorption of H₂O molecules on the sampling tube walls and filters (closed-path gas analysers only) cause amplitude and phase shifts compared to an unperturbed, ideal signal (Fratini et al., 2012; Ibrom et al., 2007; Mammarella et al., 2009; Massman and Ibrom, 2008; Nordbo et al., 2013). Thus, H₂O lag times tend to be longer than for other gases in the same sampling tube and attenuations of H₂O fluctuations are enhanced. Adsorption/desorption of H₂O depends on sampling line characteristics, i.e., tube material, flow rate, filter etc. Moreover the mechanisms are enhanced when relative humidity increases and/or dirt accumulates on the wall of the tube. This enhancement is sampling line specific (Ibrom et al., 2007; Mammarella et al., 2009). To date a comprehensive explanation for the amplitude and phase shifts is missing; however Nordbo et al. (2013) showed promising development in this aspect.

A problem arises from the fact that attenuation and phase shift of H₂O are not known in the sampling lines of those gas analysers not capable of measuring H₂O. Thus, correcting CH₄ data for the H₂O effect is difficult. To partly overcome these difficulties following procedure was used to estimate the phase shift between H₂O and other scalars. First, the ratio of H₂O and CO₂ lag times in the LI-7000 sampling line was

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



parameterized as a function of RH:

$$\frac{t_{\text{H}_2\text{O}}}{t_{\text{CO}_2}} = c + d \left(\frac{\text{RH}}{100} \right)^e, \quad (6)$$

where RH is relative humidity in %. Data were grouped in 12 relative humidity classes and the fit parameters c , d and e were obtained by fitting the above expression to median values in the 12 relative humidity classes ($r^2 = 0.9963$ and $\text{RMSE} = 0.0841$), yielding the values $c = 1.081$ (95 % confidence bounds: 1.002–1.159), $d = 6.369$ (5.849–6.888) and $e = 14.06$ (12.43–15.69). Then it was assumed that the ratio has the same relative humidity dependence in all sampling lines and, finally, the water vapour covariance, $\overline{w' \chi'_v}$, used in Eq. (5) was calculated with a lag time of

$$t = t_{\text{H}_2\text{O}}^{\text{ANA2}} - \left(\left(c + d \left(\frac{\text{RH}}{100} \right)^e \right) t_{\text{CH}_4}^{\text{ANA1}} - t_{\text{CH}_4}^{\text{ANA1}} \right), \quad (7)$$

$$t = t_{\text{H}_2\text{O}}^{\text{ANA2}} - \Delta t$$

where $t_{\text{H}_2\text{O}}^{\text{ANA2}}$ is lag time of H₂O measured with for instance LI-7000 and $t_{\text{CH}_4}^{\text{ANA1}}$ is lag time of CH₄ measured with for instance FMA. Δt takes into account the phase shift between H₂O and CH₄ in the sampling line. Now, instead of calculating $\overline{w' \chi'_v}$ with $t_{\text{H}_2\text{O}}^{\text{ANA2}}$, which corresponds to maximizing the covariance, the covariance is calculated with t and then Eq. (5) was applied. No attempt was made to correct for differences in attenuation of H₂O in different sampling lines. Moreover, the assumption that the ratio between H₂O and CO₂ lag times follows the same relative humidity dependence in all sampling lines may not necessarily be valid.

3.5 Random errors in the flux

When evaluating instrument performance, it is vital to know how much noise there is in the signal. In many situations the random error of an eddy covariance flux measurement is dominated by one-point sampling uncertainty (Businger, 1986; Kroon et al.,

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2010), caused by the stochastic nature of turbulence, rather than the performance of the instrument used in the measurement. In this study, the total random uncertainty of a flux estimate is calculated with the method proposed by Finkelstein and Sims (2001). In this method, the error variance of the covariance is estimated based on cross-covariances and auto-covariances and thus the uncertainty estimate contains contributions originating from sampling uncertainty and instrumental noise.

Lenschow et al. (2000) introduced a method to estimate instrumental noise in LiDAR measurements and later Mauder et al. (2013) applied it to eddy covariance data. Due to the fact that instrumental noise is uncorrelated with the turbulent signal, it can be assumed that the noise contributes to the auto-covariance only at lag zero. Thus, the instrumental noise can be estimated as the difference between the observed value of auto-covariance at lag zero (i.e. variance of the time series) and extrapolation of auto-covariance function values to lag zero:

$$\left(\sigma_c^{\text{noise}}\right)^2 = C_{11}(0) - C_{11}(\rho \rightarrow 0), \quad (8)$$

where $C_{11} = C_{11}(\rho)$ is the auto-covariance function, ρ is the lag and $C_{11}(\rho \rightarrow 0)$ is the extrapolation of auto-covariance function to lag zero. $C_{11}(\rho \rightarrow 0)$ was estimated using linear extrapolation and auto-covariance values at lags $4 \leq \rho \leq 11$ were used for the extrapolation. In essence, with this method the observed variance of time series $C_{11}(0)$ is divided into two parts: variance caused by turbulent mixing, $C_{11}(\rho \rightarrow 0)$, and variance caused by instrumental noise $(\sigma_c^{\text{noise}})^2$. Therefore, σ_c^{noise} describes the noise level in the measured time series. Contribution of this noise to uncertainty of the covariance is estimated using error propagation (Eq. 7 in Mauder et al., 2013):

$$\sigma_{w,c}^{\text{noise}} = \sqrt{\frac{\left(\sigma_c^{\text{noise}}\right)^2 \sigma_w^2}{N}} \quad (9)$$

where N is the amount of samples in a time series and σ_w^2 is variance of vertical wind component. Unlike in the original equation given in Mauder et al. (2013), the contribu-

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



tion of instrumental noise in w was neglected since the focus was on comparing the gas analysers. This method is sensitive only to random noise, i.e. white noise, in the signal and it cannot be used to estimate drift or low frequency noise in the signal, since such variations contribute to auto-covariance at multiple lag times, not just at lag zero.

Eddy covariance fluxes are usually estimated by maximizing the cross-covariance between time series w and χ_c . Wienhold et al. (1995) developed a method to assess how well this maximum can be detected from the cross-covariance. In their method the detection limit of a flux is estimated as the standard deviation of the cross-covariance function values far from the maximum value, i.e. the flux. In essence, this method estimates the magnitude of the background variation of the cross-covariance. In this study the detection limits were estimated by using the position of the maximum as the origin of time, calculating the cross-covariance between time series w and χ_c within lag windows -150 s to -50 s and 50 s to 150 s, and finally calculating the standard deviation of the obtained values cross-covariance values.

3.6 Flux data filtering and quality control

The flux data were screened in order to remove clearly erroneous values. Flux data were removed if there were too many spikes during an averaging period (over 3000 spikes), 30 min mean CH_4 concentrations were unrealistic (below 1.7 ppm or above 3.5 ppm) or sonic anemometer data were erroneous. In addition, the open-path LI-7700 data were screened based on the RSSI (Received signal strength indicator) and the diagnostic value provided by the instrument. The diagnostic value describes instrument activities during operation and RSSI represents how clean the mirrors in the open measurement cell are. Periods were omitted, if the 30 min mean value for RSSI was below 15. Periods were also rejected if no laser signal was detected for more than 5% of the time, or if over 3% of time the lower mirror spin motor was on. In addition to these criteria, a few clear outliers were removed from LI-7700 data based on visual inspection. A threshold of 0.08 m s^{-1} for friction velocity (u_*) was used to discard periods with low turbulent mixing. This threshold was used for all methane fluxes. The fluxes

BGD

11, 797–852, 2014

Evaluating the performance of EC CH_4 analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



were distributed into three quality classes based on a flux stationarity test (Foken and Wichura, 1996). If the test yielded smaller values than 0.3, the fluxes were given quality flag 0 (highest quality), fluxes with test values between 0.3 and 1 were given flag 1 (medium quality) and if the test yielded values above 1, then the fluxes were flagged with 2 (low quality).

4 Results

4.1 Data coverage and quality

The poorest data coverage (Fig. 2) was obtained with the G1301-f and DLT-100 instruments (26.7 % and 64.4 %, respectively); however this was caused by data logging problems and does not reflect instrument performance. FGGA and G2311-f achieved the best data coverage, which also had often the high quality flag (Fig. 3).

The largest data filtering effect using screening procedure described in Sect. 3.6 was for the LI-7700, with approximately 43 % of data rejected. This was mainly related to one period (from 9 to 14 June 2012) during which both mirrors became dirty and manual cleaning was not possible. From 18 June onwards the mirrors were cleaned manually every second day. The friction velocity criterion removed 66 30 min CH₄ flux data points (6 % of data) from all time series.

4.2 Random errors and instrumental noise

The statistics of the estimates for instrumental noise levels are shown for each instrument in Fig. 4 for periods when all gas analysers were working. The noise was not estimated for the LI-7700 due to the data logging problem (explained in Sect. 2.2). On average, the two Picarro analysers, G2311-f and G1301-f, had the lowest instrumental noise (0.4 ppb and 1.2 ppb, respectively) during the study period (Fig. 4a and b). It is however questionable whether the method used can adequately separate the turbulent signal from the instrumental noise in the case of the G2311-f for which the overall noise

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

[Title Page](#)

[Abstract](#)

[Introduction](#)

[Conclusions](#)

[References](#)

[Tables](#)

[Figures](#)



[Back](#)

[Close](#)

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)



level was very small. For comparison, the standard deviations of CH₄ concentrations, which contain contributions from both instrumental noise and atmospheric fluctuations, measured by the two Picarro instruments were approximately 4.6 ppb and 4.3 ppb, respectively. Thus the variations in the methane time series from these two analysers were clearly dominated by atmospheric fluctuations, rather than instrumental noise. This is evident in the example fast-response time series shown in Fig. 5. For the two Picarro instruments the turbulent signal can easily be seen in the time series (high concentration during upward motion and close to ambient concentration during downward motion), while for the other instruments the signal in CH₄ concentration is mixed with instrumental noise and is therefore not visible as clearly. All the other gas analysers show much higher instrumental noise levels; FGGA had the smallest instrumental noise of the non-Picarro analysers, while QCL had the highest.

The instrumental noise of the FMA1 was highly variable during the experiment and affected by cell temperature: when the cell temperature of the FMA1 was between 27 °C and 29 °C, the instrumental noise was approximately 13.8 ppb and reached up to 20 ppb, whereas in other situations (cell temperature above 29 °C or below 27 °C) it was on average 6.8 ppb. The reason for this odd temperature dependence is unknown. The noise in the FMA2 data also responded to cell temperature, but not as strongly as for the FMA1 instrument. Instrumental noise from other analysers by Los Gatos Research did not show such strong temperature dependence.

The LGR analysers report also cavity ringdown (CRD) times which describe how long it takes for the laser signal to attenuate in the optical cavity. Roughly speaking the CRD time can be thought to represent cleanliness of the mirrors in the cavity: short CRD time corresponds to dirty mirrors and long CRD time to clean mirrors. For most of the LGR analysers the CRD times decreased significantly during the campaign due to dirt accumulating on the mirrors in the cavity. Only the FGGA had a rather stable CRD time throughout the campaign, around 11.7 μs, while for other LGR analysers the values at the end of the campaign were 7 μs. Surprisingly, the instrumental noise did not significantly depend on the CRD time and periods when it was around 7 μs were

still usable for flux calculations. For example for FMA1 the instrumental noise increased from 6.5 ppb to 7.8 ppb when the CRD time was above 10 μ s or below 8 μ s (only periods when cell temperature was above 29 °C or below 27 °C were considered).

The detection limits, shown in Fig. 4c, were approximately 2 nmol m⁻² s⁻¹, except for the QCL and the FMA2, which had higher values (4 nmol m⁻² s⁻¹). The flux detection limit estimated with this method is mostly determined by the stochastic nature of turbulence during the 30 min periods selected. For the QCL the high noise level increased also the value of the detection limit, but the higher detection limit for FMA2 is not explainable in terms of white noise, since for instance FMA1 had more white noise but lower detection limit when compared to FMA2. Possibly, the FMA2 signal was contaminated also with (structured) noise rather than just white noise, which contributed more to the detection limit than to the instrumental noise.

4.3 Flux corrections

4.3.1 Spectral corrections

Ensemble-averaged methane cospectra are shown in Figs. 6 and 7, together with the corresponding temperature cospectra. In the ideal case the methane cospectra would collapse onto the temperature cospectra and both would follow the model cospectrum, which is also shown for reference. However, all methane cospectra fell below the temperature cospectrum at the high frequency end.

As the flux is the integral of the cospectrum, it is clear that the contribution of the high frequency fluctuations (small eddies) is underestimated and should be corrected. This is done with the method presented in Sect. 3.2; the response times used for the corrections are given in the figures. G2311-f had the slowest response time which was caused by instrument malfunction: although the instrument was set to measure at 10 Hz, it was effectively measuring at approximately 2.3 Hz, and the recorded 10 Hz data were a linear interpolation of the 2.3 Hz data. Thus frequencies exceeding 2.3 Hz were lost. The reason for this malfunction is unknown, but it could have been caused

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



by a memory leak produced by some additional code designed specifically for this campaign and added to the instrument's internal software (Gloria Jacobson, Picarro Inc., personal communication, 2013). Thus, if this is the case, other G2311-f analysers should not be affected.

5 The magnitudes of spectral corrections are presented in Fig. 8 as percentages of the measured raw fluxes. At close to 40 %, the correction was largest for the G2311-f. This is not surprising since it had the slowest response time. For the other methane fluxes the correction ranged from 10 to 30 % of the originally measured flux.

4.3.2 Density and spectroscopic corrections

10 As a test, the CH₄ fluxes of the FGGA were calculated by applying the H₂O corrections point-by-point (Eq. 4) and comparing these values to half-hourly fluxes calculated using block-averaging and corrected using (Eq. 5). Linear fit to the data processed with the two methods has a slope of 1.000 and intercept of $-0.001 \text{ nmol m}^{-2} \text{ s}^{-1}$ and RMSE and correlation coefficient (r) are $0.008 \text{ nmol m}^{-2} \text{ s}^{-1}$ and 1.000, respectively. The excellent agreement between the datasets confirms that the two correction methods are identical and that the choice of correction method should not induce a systematic bias between the instruments, as long as the water vapour flux used in Eq. (5) is calculated with the same lag time as the CH₄ flux.

20 The magnitude of density and spectroscopic corrections are given in Fig. 8. The density correction was on average approximately 10 % during the day and a few percents at night when water vapour fluxes were small. Spectroscopic corrections were much smaller: a few percent during the day and less than a percent at night time. However, the LI-7700 is an exception: the density correction was on average 40 % during the day and -27% at night. In addition, the spectroscopic correction was also larger (daytime 25 14 %, night time -13%). The difference between the LI-7700 and the other gas analysers is caused by the fact that the effect of temperature must be taken into account in the case of the open-path analyser, while temperature fluctuations are smoothed out for closed-path sensors with long inlet lines.

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



To validate the density and spectroscopic corrections applied, the corrected CH₄ fluxes were compared with the fully corrected G2311-f CH₄ flux. This instrument does both corrections automatically during measurement using coefficients reported in Chen et al. (2010) and it has been shown that the automatic correction implemented in Picarro instruments performs well (e.g. Rella et al., 2013). Thus the fully corrected G2311-f CH₄ flux is a good reference for the other instruments and there should not be any residual H₂O effect left in G2311-f fluxes. A linear correlation between the difference in methane flux and the density correction term ($F_{\text{CH}_4}^{\text{WPL}}$, second term on right hand side in Eq. 5) was used to evaluate if the corrections were done properly (Fig. 9 shows an example using FGGA data). If the slope equals zero, the H₂O corrections were done correctly and the differences between CH₄ flux time series does not depend on $F_{\text{CH}_4}^{\text{WPL}}$.

Values for the slope before and after applying the H₂O corrections are given in Table 4. Before applying the H₂O correction, the slopes differ from zero and the difference from zero is statistically significant. This was expected since the fluxes were still affected by density and spectroscopic effects and the difference should be related to $F_{\text{CH}_4}^{\text{WPL}}$. If the slope is small before the H₂O corrections is applied, it can be said that for that setup the effect of H₂O on CH₄ fluxes is small. This presumably can be explained by enhanced phase and amplitude shifts of H₂O signal in that particular setup which diminish the effect of H₂O on CH₄ flux measurements. This is the case for instance for DLT-100 for which the slope was -0.181 before applying any H₂O corrections.

For the QCL no density or spectroscopic corrections were applied, because the gas analyser was connected to a drier. In theory, the differences between G2311-f and QCL methane fluxes should not correlate with $F_{\text{CH}_4}^{\text{WPL}}$, since both should be free from any interference from H₂O. This is supported by the small slope derived for the QCL of -0.035 (Table 4). Thus, it can be said that the drier connected to the QCL worked well.

After applying the H₂O corrections the slopes were generally closer to zero which implies that the corrections modified the CH₄ fluxes into the right direction. For the FGGA, the slope after applying the H₂O corrections was -0.026 ± 0.047 which is

not significantly different from zero; this suggests that the coefficients obtained from Hiller et al. (2012) were applicable to the FGGA used in this study. Only the slopes for DLT-100 and FMA1 remained statistically significantly different from zero, even after correction. DLT-100 fluxes were over-corrected (the slope was positive after H₂O correction) and FMA1 fluxes were under-corrected (the slope was negative after H₂O corrections). Neither of these gas analysers measured H₂O and thus external water vapour measurements were needed to correct their CH₄ fluxes. Assuming that the coefficients obtained from Hiller et al. (2012) are applicable to the DLT-100 and FMA1 used in this study, the non-zero slopes suggest that the empirical method used to parameterize the lag time difference between the H₂O signal and other scalar signals (Sect. 3.4.1) was not fully successful. Moreover, different attenuation and shape of the cross-covariance function may have contributed to the miscalculation of the H₂O correction (see Sect. 4.3.3 and Fig. 10). For the DLT-100 the value of the covariance of w with H₂O, $w'\chi'_v$, used in Eq. (5) appears to have been too high and for the FMA1 too small. In the case of the G1301-f, which did not measure H₂O either, the method seemed to overcorrect the data since the slope was positive (0.259), but the difference from zero was not statistically significant.

The above discussion deals with the systematic error in the H₂O correction. Despite the fact that the correction is highly sensitive to the lag time, attenuation and the shape of the cross-covariance function, one might be tempted to use external H₂O measurements to correct CH₄ fluxes if the H₂O obtained from the CH₄ analyser is noisy. Uncertainties in $w'\chi'_c$ and $w'\chi'_v$ calculated with the data from FGGA were estimated based on Finkelstein and Sims (2001) and this uncertainty is assumed to be the total uncertainty of $w'\chi'_v$. By applying error propagation to Eq. (5) we can estimate how much the noise in $w'\chi'_v$ is affecting the precision of CH₄ fluxes when the density and spectroscopic corrections are done. Relative uncertainty of FGGA CH₄ fluxes is increased from 24.3% to 24.4% after applying the H₂O corrections. If the uncertainty in $w'\chi'_v$ is artificially doubled, then the relative CH₄ flux uncertainty is increased by 0.4%. Thus it can be

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



said that use of noisy water vapour data in the H₂O corrections does not compromise the precision of CH₄ fluxes. Moreover if the CH₄ analyser also measures H₂O these measurements should be used to correct the methane data no matter how noisy the H₂O signal is, rather than external water vapour data.

5 4.3.3 Correcting CH₄ fluxes without concurrent H₂O measurements

Fig. 10 exemplifies the problem of using external H₂O measurements. The cross-correlation between CH₄ (FGGA) and w , and between H₂O (FGGA) and w both peak at different lag times even though the gases are measured with the same sampling line and instrument. The difference is caused by the sorption/desorption of H₂O on the internal walls of the sampling tube and filters. The H₂O cross-covariances shown in these plots are normalized with the values that should be used in Eq. (5) for correcting FGGA CH₄ fluxes (blue dots in the plots). Thus, for instance, if in these situations the covariances between H₂O (FGGA) and w are maximized and then used in the H₂O corrections, the corrections are overestimated by 132 % (left plot in Fig. 10) and 21 % (right plot in Fig. 10). Moreover, the two H₂O cross-covariances (LI-7000 and FGGA) shown in both plots have different degree of attenuation, the FGGA H₂O cross-covariance being more attenuated, and the shape of the cross-covariance is affected by the tube effects. The FGGA H₂O cross-covariance functions are wider and the peaks not as sharp as in the LI-7000 H₂O cross-covariances. In order to successfully correct CH₄ fluxes using H₂O covariance measurements, all three effects (lag time, attenuation and shape of the cross-covariance) induced on H₂O by the sampling line must be properly taken into account. If the H₂O is measured by the same instrument as the CH₄, this is achieved by calculating the water vapour covariance with the same time-lag that is used for calculating the CH₄ flux. If H₂O is measured by an external instrument, the effects need to be estimated.

In order to study how sensitive the H₂O corrections are to the effects of the sampling line on measured H₂O, FGGA CH₄ fluxes were corrected with LI-7000 H₂O measurements and then compared with the correction calculated with the internal H₂O

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



measurements. The LI-7000 H₂O covariance used in Eq. (5) was modified with four different methods (see Fig. 10): the covariance between LI-7000 H₂O and w was maximized and then used in Eq. (5) (no revision), LI-7000 H₂O cross-covariance maximum was adjusted to be the same as FGGA H₂O cross-covariance maximum (Attenuation revised), LI-7000 H₂O lag time was revised to match FGGA H₂O lag time (Lag time revised) and both, attenuation and lag time, are revised (Both revised). A comparison between these four methods and the reference correction calculated using in-situ FGGA H₂O measurements is shown in Fig. 11. With no revision applied to LI-7000 H₂O data (i.e. the water vapour covariance is calculated from the LI-7000 data choosing the time lag that maximizes the covariance for that sensor), the correction is clearly over-estimated, by approximately $2.85 \text{ nmol m}^{-2} \text{ s}^{-1}$ (74 % overestimation), when LE was above 150 W m^{-2} . If the attenuation is revised this error decreases to $1.07 \text{ nmol m}^{-2} \text{ s}^{-1}$ (25 % overestimation), if the lag time is revised the error is $0.24 \text{ nmol m}^{-2} \text{ s}^{-1}$ (5 % overestimation) and if both are revised the correction becomes underestimated by $0.88 \text{ nmol m}^{-2} \text{ s}^{-1}$ (21 % underestimation). By revising both, attenuation and lag time, the results were worse than just by altering the lag time. This stems from the fact that the shape of the H₂O cross-covariance is also altered which was not accounted for here.

The fact that H₂O corrections are highly sensitive to the lag time used in calculating $w' \chi'_v$ result from the shape of cross-covariance between w and χ_v . The cross-covariance between w and χ_v is an exponential function of the time lag between the time series and thus the error in the correction increases exponentially when error in the lag increases.

4.4 Agreement between flux estimates

After corrections have been applied as well as possible, all the instruments agreed relatively well with the median CH₄ flux (Table 5). The slopes of the linear fits with this median flux were between 1.090 (G1301-f) and 0.997 (FGGA) and the intercepts

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



were acceptable, ranging from -6.835 (LI-7700) to $2.997 \text{ nmol m}^{-2} \text{ s}^{-1}$ (DLT-100). The values of the correlation coefficient were close to one, which implies that all flux results are highly correlated. The LI-7700 had the highest value of root mean square error (RMSE); this implies that it had the highest scatter in CH₄ fluxes. This might be at least partly caused by the LI-7700 data logging problem and partly caused by the fact that it is an open-path gas analyser and the open sampling cell was vulnerable to disturbances which might appear as high variation in the fluxes. Of the closed-path instruments FMA2, QCL and G1301-f gave the highest values for RMSE.

Despite the good agreement between the CH₄ fluxes shown in Table 5, there seems to have been a systematic bias between the different instruments which is revealed by the cumulative sums shown in Fig. 12 (left plot). Most of the flux time series gave a value around 360 mg m^{-2} for the cumulative CH₄ emission during the 13 days long period shown in the plot, however three time series deviated most from this value: FMA1 and QCL derived about 330 mg m^{-2} and the DLT-100 399 mg m^{-2} for the cumulative sum.

Cumulative CH₄ emissions from three gas analysers (G2311-f, FGGA and FMA2) agree best. For these three analysers the H₂O corrections were done using internal H₂O measurements and thus it can be assumed that the corrections were done accurately. Two out of the three time series which diverged most from the mean (DLT-100 and FMA1) were corrected with external H₂O measurements, which suggests that the differences in cumulative CH₄ emissions could have been caused by the H₂O corrections. To test this, the H₂O corrections were adjusted so that the slopes in the column on the right in Table 4 became zero, meaning that a time series $-kF_{\text{CH}_4}^{\text{WPL}}$, where k is the slope, was added to the fluxes. After the adjustment the difference between the G2311-f CH₄ flux and other CH₄ fluxes no longer depended on $F_{\text{CH}_4}^{\text{WPL}}$. Cumulative CH₄ emissions after adjusting the H₂O corrections are shown on the right in Fig. 12. The agreement is better than before adjustment and now the values range from 335 mg m^{-2} (QCL) to 367 mg m^{-2} (G2311-f). Thus the differences seen before the adjustment can at least partly be attributed to miscalculated H₂O corrections.

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Monthly values were calculated by computing mean values for the CH₄ fluxes during 12 to 25 June (same time period as in Fig. 12) and then multiplying the mean values with the length of the month June (Fig. 13). Figure 13 shows the same pattern as Fig. 12: the DLT-100 initially gave higher fluxes and the FMA1 gave lower fluxes than all the analysers on average; however, the difference decreased if the H₂O corrections were set to match the G2311-f. For the DLT-100 the difference between fluxes calculated with original and adjusted H₂O correction on a monthly scale was approximately 0.1 g (CH₄) m⁻², which is 8 % of the monthly CH₄ emission observed on average. These results highlight the importance of proper H₂O corrections, especially for calculating long term CH₄ balances. As the absolute value of the H₂O corrections depends on the magnitude of the latent heat flux, the corrections are significant in locations where latent heat fluxes are large and CH₄ fluxes are low.

The slightly smaller cumulative CH₄ emissions observed with QCL, FMA2 and FMA1 than with the other three gas analysers cannot be explained by the use of two anemometers: the anemometer which was accompanying the three analysers mentioned above (METEK2) gave on average 4 % larger value for turbulence intensity (σ_w/U) and 2 % larger value for sensible heat flux than the other anemometer (METEK1). This implies that the fluxes measured with METEK2 should not be underestimated compared to fluxes measured with METEK1.

5 Discussion

All three previously published CH₄ flux inter-comparison studies (Detto et al., 2011; Peltola et al., 2013; Tuzson et al., 2010) showed relatively good correspondence between measured CH₄ fluxes. Detto et al. (2011) compared the performance of LI-7700 against two LGR analysers (FMA and FGGA) and they showed that minimum detectable flux was similar from all instruments and the fluxes from LI-7700 agreed with the closed-path LGR CH₄ fluxes during both low and high flux periods, although there was scatter in the results. They also concluded that using a low capacity pump instead of a high ca-

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

[Title Page](#)

[Abstract](#)

[Introduction](#)

[Conclusions](#)

[References](#)

[Tables](#)

[Figures](#)

[⏪](#)

[⏩](#)

[◀](#)

[▶](#)

[Back](#)

[Close](#)

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)



capacity version for the closed-path LGR analyser will add uncertainty to the fluxes. Tuzson et al. (2010) compared the results from QCL and FMA gas analysers. In that study artificial methane flux was created in the prevailing wind direction and both instruments were able to quantify the artificial flux accurately. Peltola et al. (2013) presented results from methane flux measurements at a boreal fen during summer 2010. In that study four methane gas analysers were deployed (FMA, G1301-f, TGA-100A (Campbell Scientific Inc., USA) and early prototype version of LI-7700). Excellent agreement between FMA and G1301-f was shown, whereas LI-7700 and TGA-100A produced slightly more scattered results. Less than 0.5 g (CH₄) m⁻² difference (4% relative difference) was found in the cumulative sums of the gapfilled CH₄ flux data over 6 months from FMA, G1301-f and TGA-100A. Although all these studies provide valuable knowledge on the relative performance of the instruments tested, they do not include a comprehensive set of CH₄ gas analysers and thus our study with eight methane instruments contains important information for the scientific community.

One of the main aims in running an eddy covariance site and measuring greenhouse gas fluxes is to obtain monthly, annual or even decennial GHG budgets. Calculation of long-term budgets requires high data coverage in order to minimize the need for gap filling. Typically, open-path instruments, such as the LI-7700 used in this study, produce less flux data than closed-path devices due to the vulnerability of the open measurement cell to the elements. Data gaps often occur under certain circumstances, such as rainy periods or episodes with snow. As a consequence, when using open-path instruments, it is difficult to study how the flux levels react to precipitation. This complicates gap-filling during these episodes.

It was shown in Sect. 4.2 that the Picarro instruments G2311-f and G1301-f are superior to the other instruments tested in terms of signal noise. For some LGR analysers, the noise responded to cell temperature and to a lesser extent to cavity ringdown time. (The newer gas analysers by LGR, so called enhanced performance or EP models, control cell temperature more rigorously and this problem may at least partly be solved, however this could not be verified in this study since no enhanced performance

models were used.) As for the QCL the higher noise level is typical for this older pulsed laser system. Newer QCL systems use continuous wave lasers and show significantly lower white noise levels. Unfortunately, such a system was not yet available for this campaign.

5 Noise in the signal makes the data analysis more difficult. For instance, spectral analysis of the data is difficult since parts of the spectra are covered by white noise. Also assessing high frequency attenuation of the signal becomes difficult if the measurements and the calculated cospectra are noisy, nevertheless frequency ranges that contribute the most to the turbulent exchange are well-resolved by all instruments (Figs. 6 and 7).
10 Moreover the noise in concentration data increases the random uncertainty of fluxes. However, the flux uncertainty caused by noise in CH₄ concentration is in the order of 1 nmolm⁻²s⁻¹ (Fig. 4), whereas the fluxes measured during this study were at least ten times larger and thus even the relatively noisy QCL signal did not compromise the use of the measurements. In addition, for these instruments and the magnitude of the
15 fluxes at the site, most of the flux random uncertainty originates from one-point sampling of the flux (Businger, 1986; Kroon et al., 2010) and not from instrumental noise. The sampling error is an inherent property of eddy covariance measurements which cannot be minimized by instrument selection or design.

All CH₄ flux time series were corrected with commonly used methods. The magnitudes of the corrections (Fig. 8) are measurement site and setup specific and should be only thought of as indicative; in any case, they can be compared with each other to find out which analyser needs large corrections. As pointed out by Detto et al. (2011) and Peltola et al. (2013) the density and spectroscopic corrections are much larger for open-path LI-7700 than for the closed-path devices due to the additional contribution
20 from un-attenuated temperature fluctuations. Large corrections are not problematic as long as the sensible and latent heat fluxes used in density and spectroscopic corrections are measured accurately. However, any bias in these fluxes will also bias the CH₄ fluxes, especially where CH₄ fluxes are small. Usually, sensible and latent heat fluxes can exhibit significant diurnal patterns and thus miscalculated density and/or spectro-

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



scopic corrections can cause a false daily cycle in the CH₄ flux which might lead to misinterpretation. This is especially problematic if measurements are done at locations where small CH₄ fluxes are measured with large sensible and latent heat fluxes. In such conditions, the corrections are large relative to the real flux (cf. Smeets et al., 2009). This problem is relevant for all analysers, but especially open-path instruments and those closed-path devices that do not measure H₂O simultaneously. As shown in Sect. 4.3.2, estimating the H₂O effect on the measured CH₄ flux without any knowledge on how the measurement system alters H₂O fluctuations will most likely lead to miscalculated corrections, and may even cause an erroneous daily cycle in CH₄ fluxes and overall bias the CH₄ fluxes. One approach to minimize this problem is the usage of one main sampling line for two analysers: one which measures CH₄ (e.g. FMA) and one which measures H₂O (e.g. LI-7000) and then use the measured H₂O to correct CH₄ data. For FMA1 such a measurement setup was used in this study, nevertheless the H₂O corrections were still underestimated (Table 3). This might have been caused by the use of different filters in FMA1 and LI-7000 sampling lines (Table 1) and their different contribution to H₂O phase and amplitude shifts. Alternatively, one can determine the H₂O effects in the sampling line before it is deployed in the field (cf. Querino et al., 2011), or the air sample may be dried; however, none of the driers completely removes H₂O from the air samples, there is always some residual H₂O left. Thus if possible one should use gas analysers which measure CH₄ and H₂O in-situ or alternatively open-path analysers for which the WPL and spectroscopic corrections can be done using atmospheric sensible and latent fluxes. All the new CH₄ closed-path gas analysers measure also H₂O, and thus this problem is more related to use of slightly older models, such as the FMA.

6 Summary and conclusions

Eight fast-response methane gas analysers suitable for eddy covariance flux measurements were inter-compared at a Dutch agricultural site and the high frequency data was

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



post-processed with up-to-date methods. The measurements were evaluated based on several parameters, such as data coverage and quality, amount of noise in the signal, magnitude and simplicity of different corrections, and the agreement of the fluxes obtained with the different instruments.

5 Mostly the differences in performance result from data coverage and amount of noise in the data. For some of the slightly older instruments the fact that they did not measure H₂O proved to be a challenge during data processing and a significant potential source of bias in the CH₄ fluxes, which amounted to around 0.1 g (CH₄) m⁻² on a monthly scale and can therefore be significant at sites with small or medium CH₄ fluxes. Biased time series are difficult to correct since the bias does not decrease with averaging, as in the case of random noise.

10 Out of the three newest gas analysers (G2311-f, FGGA and LI-7700), the G2311-f was superior to the others with respect to instrumental noise, but noise aside, the two closed-path instruments (G2311-f and FGGA) performed similarly. The performance of LI-7700 was difficult to assess due to data logging issues with that instrument particular to our study. In any case, the analyser needed a lot more maintenance than the other analysers due to repeated contamination of the open measurement cell. However, with clean mirrors, the performance of the LI-7700 was comparable to the other new (closed-path) instruments. Thus the analyser remains a viable option for locations with limited power availability, as long as it can be cleaned regularly which can be demanding at remote locations. Also, the G1301-f performed well during the campaign, although the performance was difficult to assess fully because it was operated only a few days at the end of the campaign. However, the data obtained seemed to agree well with the FGGA and the G2311-f and instrumental noise was low.

25 The main conclusion is that all systems (excluding G1301-f and LI-7700 due to low data coverage) agreed on the cumulative flux pattern over an episode of thirteen days within ±10%. At the moderate/medium range flux levels observed over the Cabauw fields each of these setups can deliver good data. For low flux conditions however, systems with less noise will outcompete the others. Moreover the good agreement

BGD

11, 797–852, 2014

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



between cumulative CH₄ emissions indicates that the selection of an instrument does not strongly bias the measured fluxes in one way or another, as long as the data is processed appropriately.

Appendix A

5 Derivation of Eq. (5)

Equation (4), originally proposed by Rella (2010), can be used to correct measurements of gas *c* for any interference from H₂O. The equation can be modified to be applicable to the averaged fluxes by first using Reynolds decomposition to all three time series, namely molar fraction and mixing ratio of gas *c* (χ_c and r_c , respectively) and molar fraction of H₂O (χ_v):

$$\overline{\chi_c} + \chi'_c = (\overline{r_c} + r'_c) \left(1 + a\chi'_v + a\overline{\chi_v} + b\chi'_v\overline{\chi_v} + b\overline{\chi_v}^2 + b\overline{\chi_v}\chi'_v + b\chi_v'^2 \right) \quad (\text{A1})$$

Now if equation above is multiplied with w' (vertical wind speed fluctuations) and the result is averaged, then

$$\overline{w'\chi'_c} = a\overline{r_c}\overline{w'\chi'_v} + 2b\overline{r_c}\overline{\chi_v w'\chi'_v} + \overline{w'r'_c} + a\overline{\chi_v w'r'_c} + b\overline{\chi_v}^2\overline{w'r'_c}, \quad (\text{A2})$$

15 where terms such as $\overline{w'\chi'_c}$ yield zero by definition and third order terms such as $\overline{w'r'_c\chi'_v}$ are assumed to be negligible. By solving $\overline{w'r'_c}$ from the equation above and using Eq. (4) for mean values, following equation is achieved:

$$\overline{w'r'_c} = \frac{1}{1 + a\overline{\chi_v} + b\overline{\chi_v}^2} \left(\overline{w'\chi'_c} - \frac{a + 2b\overline{\chi_v}}{1 + a\overline{\chi_v} + b\overline{\chi_v}^2} \overline{\chi_c w'\chi'_v} \right). \quad (\text{A3})$$

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Next the second term on the right hand side within the parentheses is separated to density and spectroscopic parts:

$$\overline{w'r'_c} = \frac{1}{1 + a\overline{\chi_v} + b\overline{\chi_v}^2} \left(\overline{w'\chi'_c} + \frac{\overline{\chi_c}}{1 - \overline{\chi_v}} \overline{w'\chi'_v} - \frac{a + 2b\overline{\chi_v} - b\overline{\chi_v}^2 + 1}{(1 + a\overline{\chi_v} + b\overline{\chi_v}^2)(1 - \overline{\chi_v})} \overline{\chi_c} \overline{w'\chi'_v} \right). \quad (\text{A4})$$

Finally, the obtained expression for $\overline{w'r'_c}$ is substituted to Eq. (1), which yields Eq. (5).

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 10 acknowledged for lending the G2311-f instrument and for valuable discussion during the preparation of this study.

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Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Evaluating the performance of EC CH₄ analyzers

O. Peltola et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 1. Characteristics of gas analyser setups.

Gas analyser	Production year	Gas measured	Horizontal sensor separation [cm]	Vertical sensor separation [cm]	Sampling tube length [m]	Inner diameter [mm]	Flow rate [LPM]	Filters	Tube material	Pump
LI-7700 (LI-COR)	2010	CH ₄	30	0	N/A	N/A	N/A	–	–	–
LI-7500	–	CO ₂ , H ₂ O	30	0	N/A	N/A	N/A	–	–	–
G2311-f (Picarro)	2011	CH ₄ , CO ₂ , H ₂ O	5	15	30	8	25	Coarse dust (inlet) + 10 µm particulate filter	PTFE	Side channel blower (Samos SB 0080D, Busch Produktions GmbH, Maulburg, Germany)
FGGA (Los Gatos Res.)	2008	CH ₄ , CO ₂ , H ₂ O	5	15	9 + 21	6 + 9	29	Coarse dust (inlet) + 10 µm particulate filter	PTFE	dry vacuum scroll pump (XDS35i, BOC Edwards, Crawly, UK)
DLT-100 (Los Gatos Res.)	2005	CH ₄	5	25	30	5	50	60 µm Coarse dust (inlet) + 2 µm Swagelok (Swagelok pat no. SS-4FW4-2)	PTFE	dry vacuum scroll pump (XDS35i, BOC Edwards, Crawly, UK)
FMA1 (Los Gatos Res.)	2008	CH ₄	5	15	41	9	32	Coarse dust (inlet) + 2 µm Swagelok (Swagelok pat no. SS-4FW4-2)	PTFE	dry vacuum scroll pump (XDS35i, BOC Edwards, Crawly, UK)
FMA2 (Los Gatos Res.)	2006	CH ₄ , H ₂ O	5	15	42	5	n.d.	Coarse dust (inlet) + Whatman Glass Fibre thimbles, 603G	PTFE	Vacuum scroll pump (Varian TriScroll 300, Palo Alto, California, USA)
G1301-f (Picarro)	2009	CH ₄ , CO ₂	5	15	42	5	n.d.	Coarse dust (inlet) + Whatman Glass Fibre thimbles, 603G	PTFE	Vacuum scroll pump (Varian TriScroll 300, Palo Alto, California, USA)
QCL (Aerodyne Res. Inc.)	2005	CH ₄ , N ₂ O	5	15	41	5	25	Balston DFU Grade BQ	PTFE	Vacuum scroll pump (Varian TriScroll 300, Palo Alto, California, USA)
LI-7000 (LI-COR)	–	CO ₂ , H ₂ O	5	15	41	9	32	Coarse dust (inlet) + 1 µm Gelman filter	PTFE	dry vacuum scroll pump (XDS35i, BOC Edwards, Crawly, UK)

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Table 2. Calibration coefficients for different instruments used in this study. Values for offset are given in ppm for other gas analysers than LI-7700. For LI-7700 the coefficient is given in mmol m⁻³. Values for the coefficient of determination are given in the last column.

Gas analyser	Applied after	Offset	Gain	R^2
FMA1	6 Jun 2012	-0.072	1.055	0.94
FMA2	6 Jun 2012	0.033	1.001	0.94
	20 Jun 2012	-0.143	1.074	0.96
G1301-f	6 Jun 2012	-0.036	1.030	0.97
QCL	6 Jun 2012	-0.155	1.092	0.83
	13 Jun 2012	-0.252	1.227	0.80
	21 Jun 2012	0.114	0.953	0.85
DLT-100	6 Jun 2012	-0.077	1.039	0.93
	18 Jun 2012	-0.225	1.099	0.96
FGGA	6 Jun 2012	0.033	0.971	0.95
G2311-f	6 Jun 2012	0.014	0.983	0.96
LI-7700	6 Jun 2012	0.0047	0.901	0.67

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 3. Spectroscopic coefficients used in Eqs. (4) or (5) for different gas analysers.

Gas analyser	a [(mol mol ⁻¹) ⁻¹]	b [(mol mol ⁻¹) ⁻²]	Source
FMA (Los Gatos Res.)	-1.219	1.678	Hiller et al. (2012)
DLT-100 (Los Gatos Res.)	-1.219	1.678	Hiller et al. (2012)
FGGA (Los Gatos Res.)	-1.189	0.2096	Hiller et al. (2012)
G1301-f (Picarro)	-1.27	0.14522	Rella (2010)

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Table 4. Summary of the values for slope k of a linear fit $\Delta F_{\text{CH}_4} = k F_{\text{CH}_4}^{\text{WPL}}$, where ΔF_{CH_4} is the difference between fully corrected G2311-f methane flux and flux from gas analyser in question, and $F_{\text{CH}_4}^{\text{WPL}}$ is correction term related to density corrections (second term on right hand side in Eq. 5). $F_{\text{CH}_4}^{\text{WPL}}$ was calculated using H₂O measured with LI-7000 and the lag time of H₂O itself, meaning that the used H₂O covariance was maximized for every averaging period. Values and standard errors for the slope k are given before and after H₂O corrections (density and spectroscopic correction). In theory, after H₂O corrections the slope should be zero. Symbol * highlights those values for k which are significantly different from 0 at $P = 95\%$. LI-7700 is not shown, since also temperature, not only H₂O, is affecting LI-7700 CH₄ fluxes.

Gas analyser	Before H ₂ O corrections	After H ₂ O corrections
G2311-f	$-1.266 \pm 0.018^*$	0
FGGA	$-0.657 \pm 0.050^*$	-0.026 ± 0.047
DLT-100	$-0.181 \pm 0.140^*$	$0.846 \pm 0.140^*$
FMA1	$-1.134 \pm 0.178^*$	$-0.200 \pm 0.180^*$
G1301-f	$-0.416 \pm 0.269^*$	0.259 ± 0.272
FMA2	$-0.458 \pm 0.172^*$	0.116 ± 0.177
QCL	-0.035 ± 0.245	-0.035 ± 0.245

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 5. Agreement between CH₄ fluxes obtained from different instruments and mean CH₄ flux from all instruments. 221 CH₄ flux data points when all instruments were working were used in the analysis, except for G1301-f only 139 points were used due to short operating time of the instrument. Only periods when all CH₄ fluxes had the quality flag 0 or 1 and difference between sensible heat fluxes from the two anemometers was smaller than 40 W m⁻² were used.

Gas analyser	Slope	Intercept [nmol m ⁻² s ⁻¹]	RMSE [nmol m ⁻² s ⁻¹]	Correlation coefficient <i>r</i>
FMA1	1.029	-2.076	5.642	0.986
QCL	1.000	-1.652	7.092	0.977
G1301-f	1.090	-2.489	6.386	0.989
FMA2	1.058	-0.303	8.001	0.974
FGGA	0.997	1.015	4.782	0.990
LI-7700	1.014	-6.835	16.554	0.879
G2311-f	1.003	1.128	5.573	0.986
DLT-100	1.055	2.997	5.742	0.987

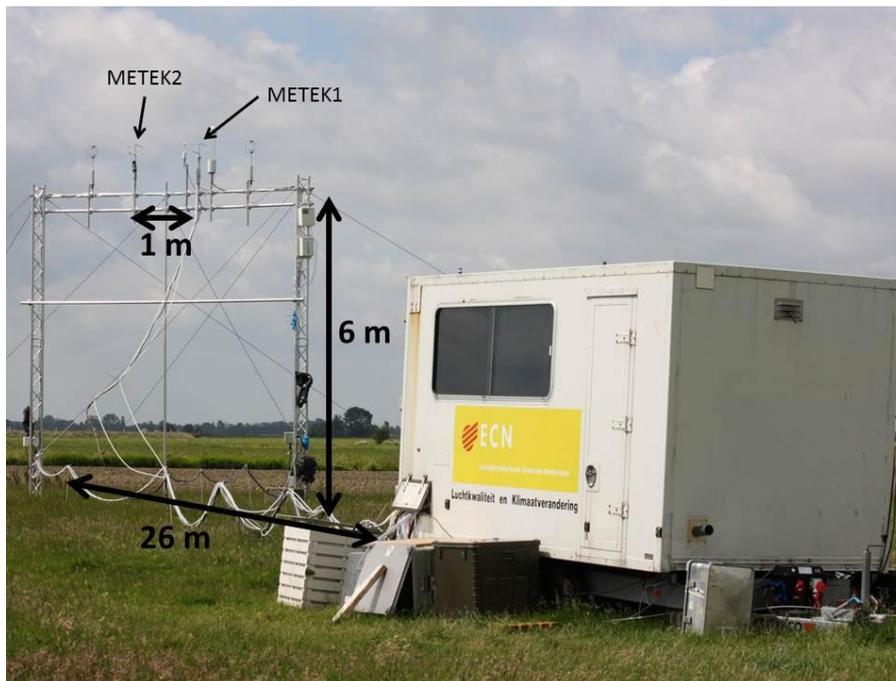


Fig. 1. Picture of the measurement setup. The picture was taken towards the most common wind direction during the campaign. The trailer which housed the closed-path instruments and data logging computer is at the front of the picture.

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



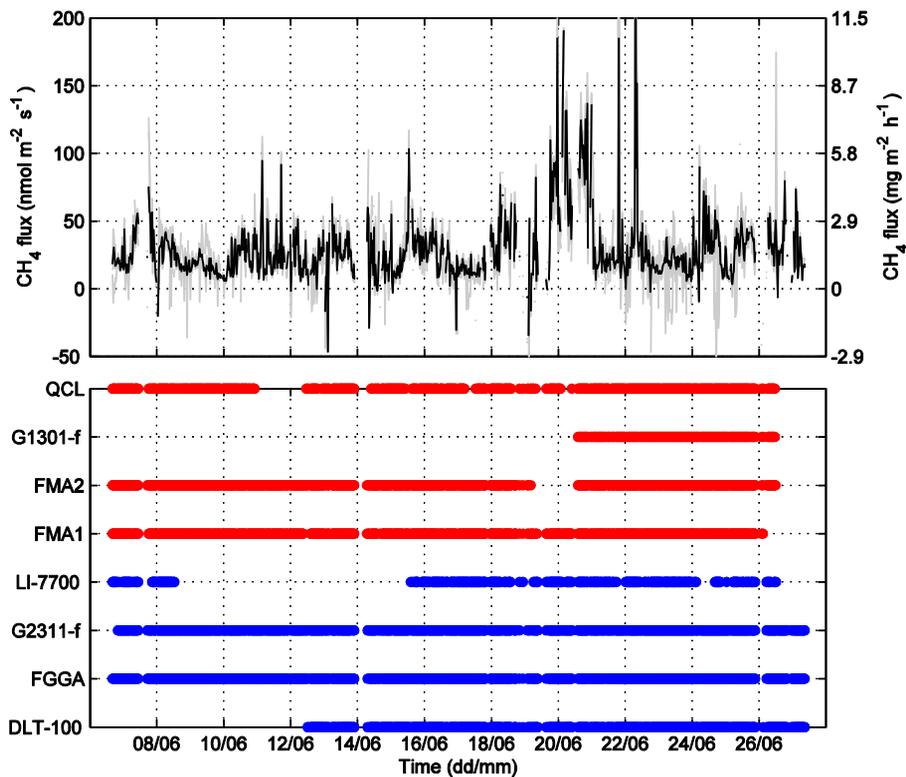


Fig. 2. Upper plot show methane flux time series (grey lines: individual CH_4 flux time series, black line: median CH_4 flux time series) and the bottom plot shows periods when gas analysers were working. Red colour corresponds to fluxes measured with METEK2 and blue to fluxes measured with METEK1.

Evaluating the performance of EC CH_4 analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

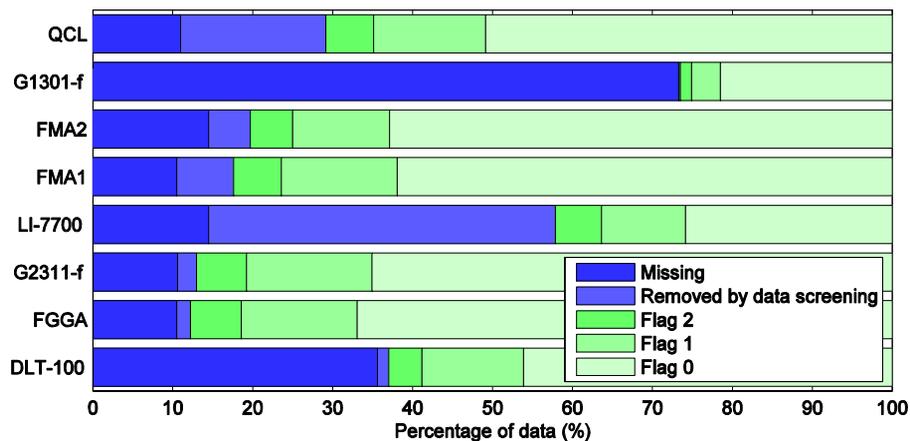


Fig. 3. Distribution of quality flags for methane fluxes during the campaign. Quality flags were given according flux stationarity criteria (Foken and Wichura, 1996). Flag 0 (flux stationarity below 0.3) corresponds to highest quality, flag 1 (flux stationarity between 0.3 and 1) to mediocre quality and flag 2 to lowest quality (flux stationarity above 1). Green bars show the amount of accepted data and blue bars shown the amount of omitted data.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

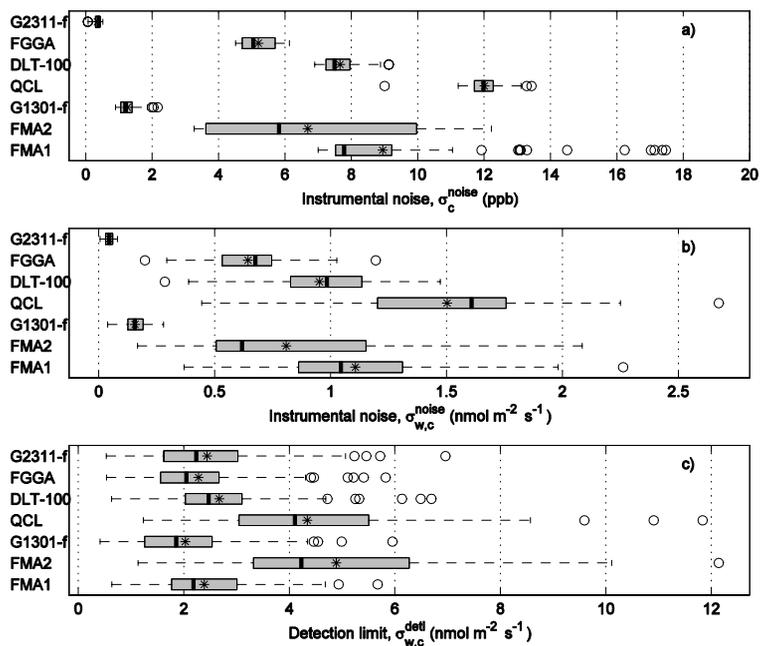


Fig. 4. Boxplots of instrumental noise (upper two plots) and detection limit (bottom plot) calculated with methods described in Sect. 3.5. Only periods when all the instruments were working and the CH₄ flux was below 30 nmol m⁻² s⁻¹ were used (129 points) for the detection limit plot and for the instrumental noise plots periods when all the instruments were working and the instrumental noise was estimated successfully were used (99 points). Grey boxes show the interquartile range, vertical lines within the boxes show medians, stars show the means, dashed whiskers show the limits for outliers and circles show values outside these limits. LI-7700 is not shown due to the data logging problem.

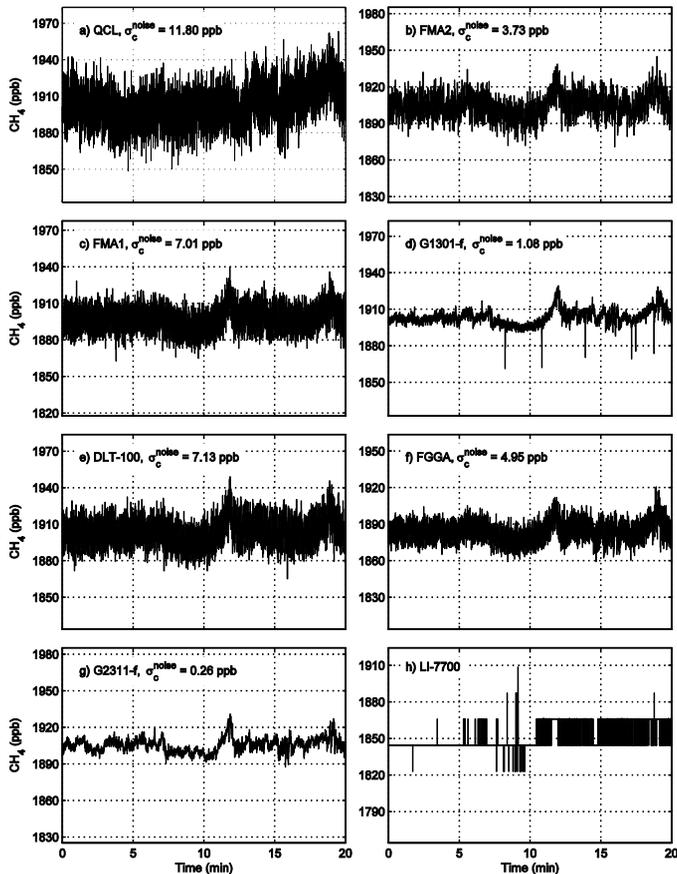


Fig. 5. Example of raw CH_4 concentration data from each instrument. Data was measured in 22 June 2012, between 10:40–11:00 LT. Corresponding instrumental noise estimates are also shown in the figure, except for LI-7700. The LI-7700 data are clearly affected by the data logging problem.

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

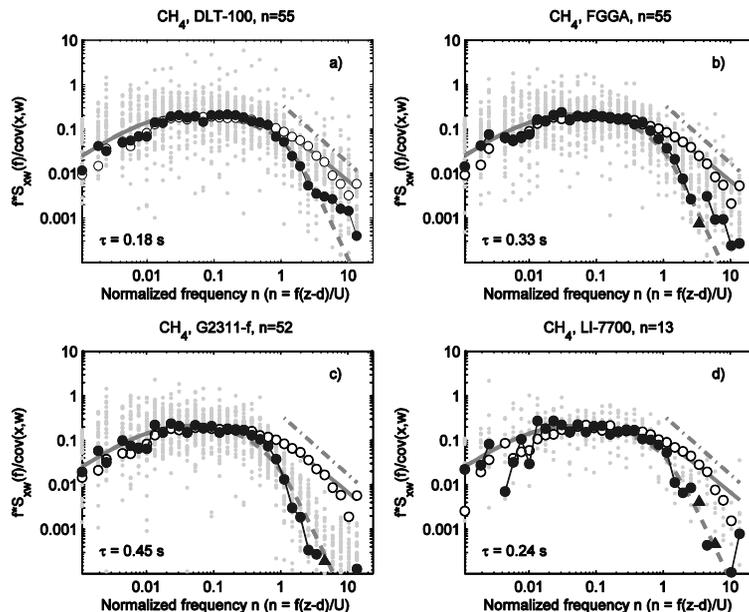


Fig. 6. Normalised, frequency weighted methane co-spectra between methane and vertical wind velocity for the different instruments associated with the anemometer METEK1 (black dots show positive values, black triangles negative values), corresponding temperature cospectra (white dots), theoretical slope $-4/3$ in the inertial subrange (dash-dotted line), model cospectrum (grey line) and model cospectrum multiplied with transfer function describing high frequency attenuation, Eq. (3) (dashed grey line). Response time describing high frequency attenuation is given in the figure for each gas analyser. Small grey dots show individual cospectra from which the mean cospectrum (black markers) is calculated. For this figure data were selected from the period 19 to 27 June, using only unstable periods when (1) methane flux was directed upwards, (2) wind speed was between 2 ms^{-1} and 6 ms^{-1} , (3) CH₄ fluxes were flagged with quality flag 0 and (4) more than 60 % of the points in the normalized cospectra were positive.

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

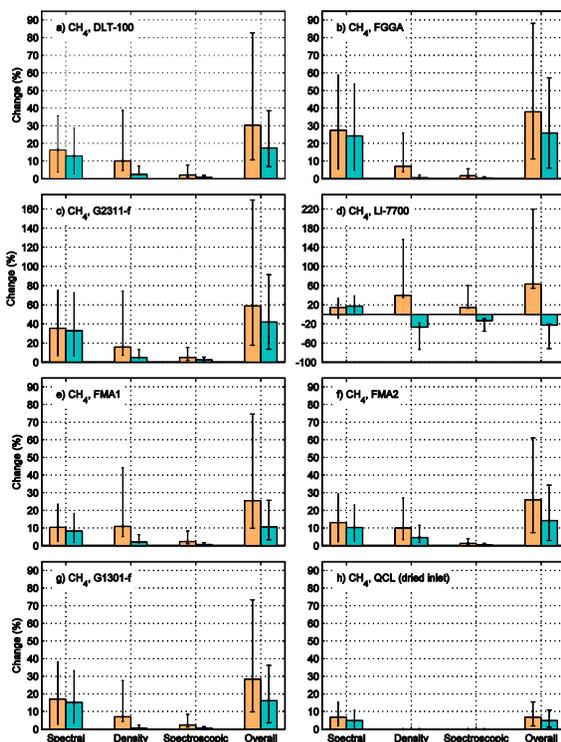


Fig. 8. Change in methane flux after applying different corrections. The values are given as a percentage of raw uncorrected flux. Bold bars give the median and error bars give 25th and 75th percentile values. Light brown bars show daytime data (sun elevation angle > 0°) and light blue bars show night time data (sun elevation angle < -3°). Bars on the far right show the overall effect. Negative value means that the correction is increasing downward fluxes, positive that it is increasing upward fluxes. Note the changing scale on y-axis.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

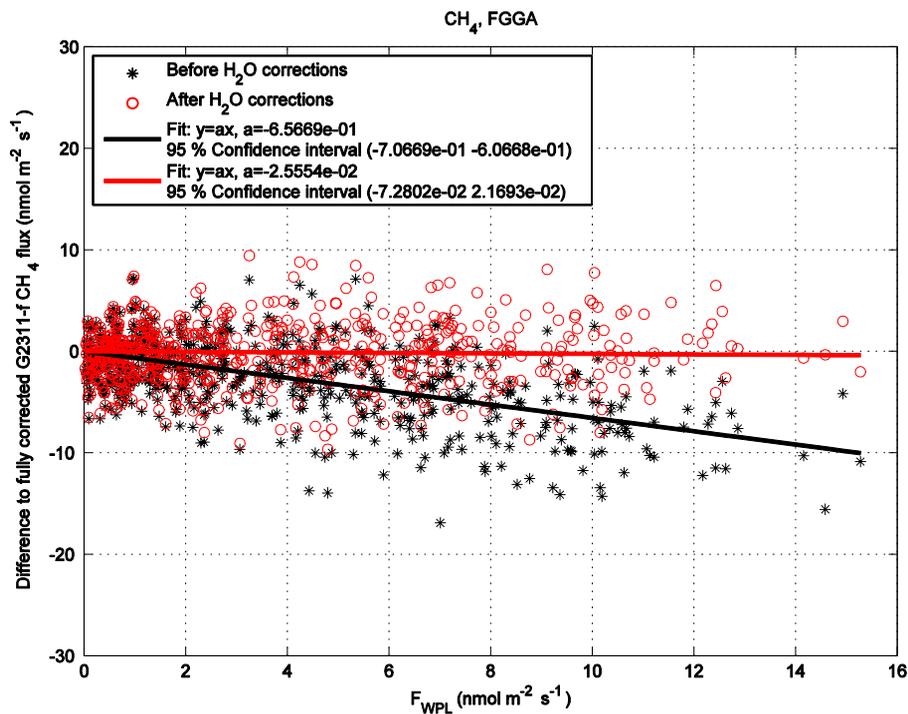


Fig. 9. Difference between FGGA methane fluxes and fully corrected G2311-f methane fluxes before (black markers) and after (red markers) H₂O corrections are applied to FGGA CH₄ data. F_{WPL} was calculated with H₂O covariance, $w'\chi'_V$, maximized.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

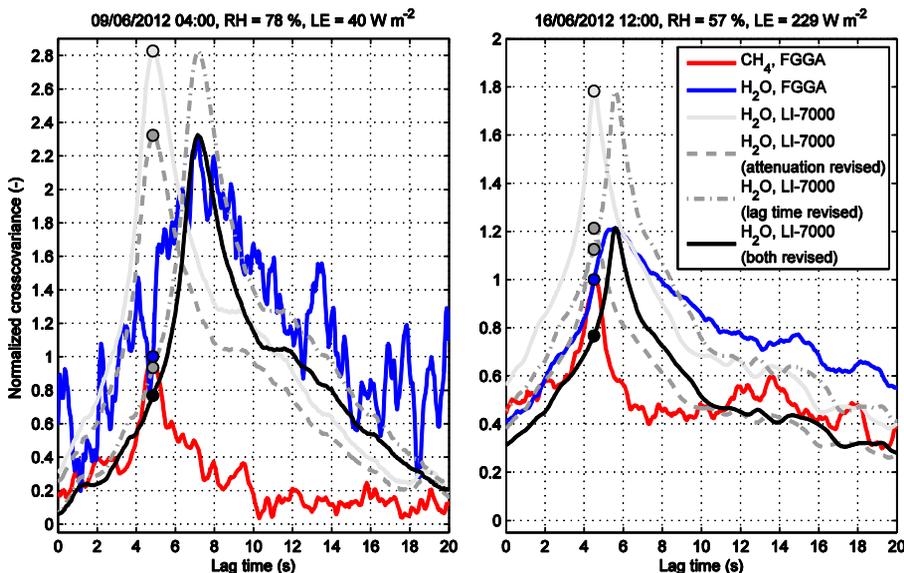


Fig. 10. Two examples of cross-covariances between w and CH₄ (FGGA), H₂O (FGGA) and H₂O (LI-7000). Location (lag time) and amplitude (attenuation) of H₂O (LI-7000) cross-covariance is adjusted in order to replicate H₂O (FGGA) cross-covariance. H₂O cross-covariances were normalized with the values that should be used in Eq. (5) in order to illustrate the magnitude of the relative error made in density and spectroscopic corrections (Eq. 5) if different values for $w' \chi'_V$ are used. The dots show the values which would be used in the correction (Eq. 5).

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



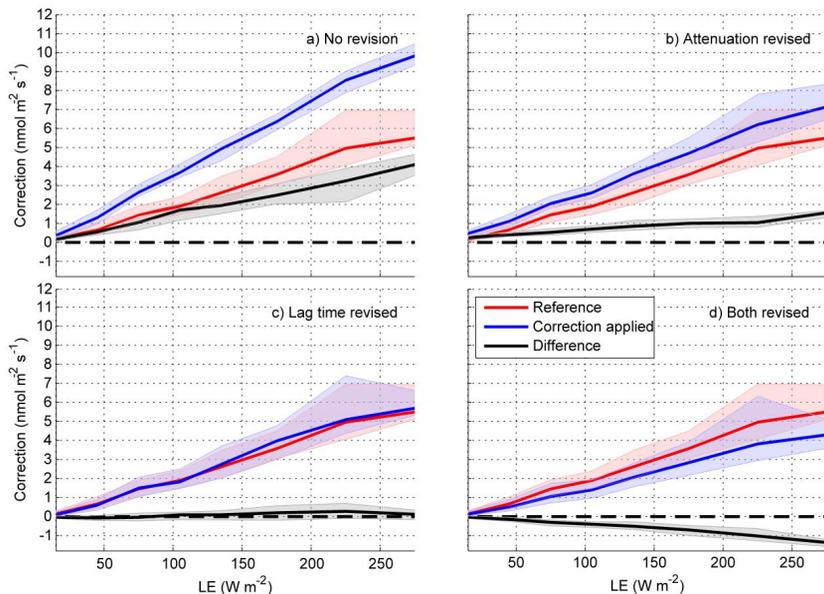


Fig. 11. Combined error in density and spectroscopic corrections as a function of latent heat flux if external water vapour measurements are used in Eq. (5). FGGA CH_4 fluxes were corrected with LI-7000 H_2O measurements (blue colour) and with FGGA's internal H_2O measurements (red colour) which yield correct values and are shown for comparison. The difference between these two, i.e. error in the correction using external H_2O is plotted with black colour. The lines show medians and the areas show interquartile range around the medians. **(a)** The correction is calculated with maximizing LI-7000 $w' \chi'_v$. **(b)** Maximum of LI-7000 $w' \chi'_v$ is adjusted to match maximum of FGGA $w' \chi'_v$. **(c)** Lag time of LI-7000 $w' \chi'_v$ is adjusted to match the lag time of FGGA $w' \chi'_v$. **(d)** Both lag time and maximum of LI-7000 $w' \chi'_v$ are set to match FGGA $w' \chi'_v$. Data measured during the whole campaign (635 points) was divided into 8 latent heat flux bins before plotting. The dashed lines highlight the zero line. See also Fig. 10 for examples from two contrasting averaging periods.

Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

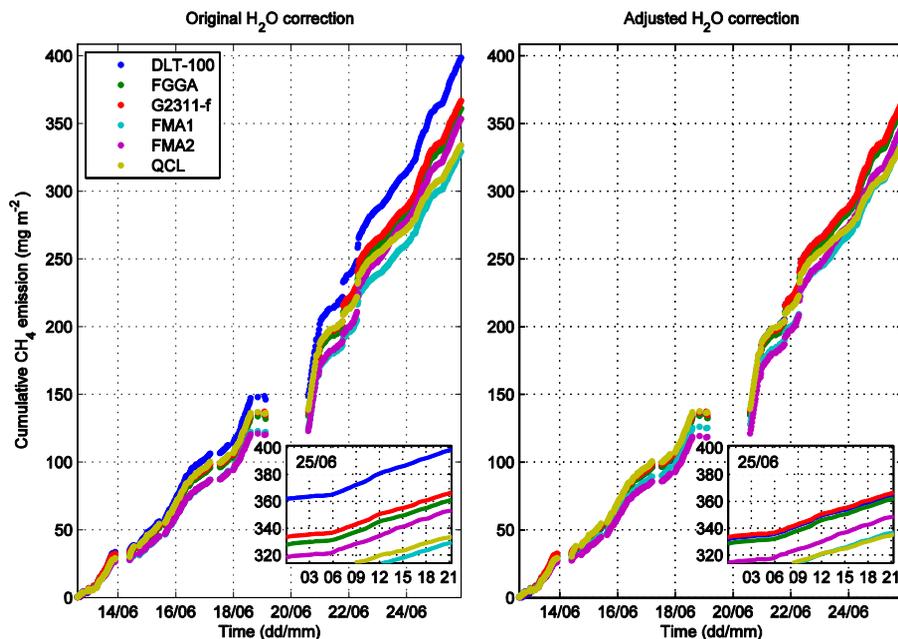


Fig. 12. Cumulative sums of CH₄ fluxes during a part of the campaign (from 12 to 25 June). The time series are not gap filled and they all contain the same amount of points. The left plot show fluxes calculated with the original H₂O corrections and the right plot shows cumulative CH₄ fluxes after adjusting the H₂O corrections with the slopes given in the right in Table 3, in other words by adding $-kF_{\text{CH}_4}^{\text{WPL}}$, where k is the slope. Thus, after this adjustment the difference between G2311-f CH₄ fluxes and other CH₄ fluxes do not depend on $F_{\text{CH}_4}^{\text{WPL}}$. Data from LI-7700 and G1301-f are not shown. The insets show a close-up of the last day.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Evaluating the performance of EC CH₄ analysers

O. Peltola et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

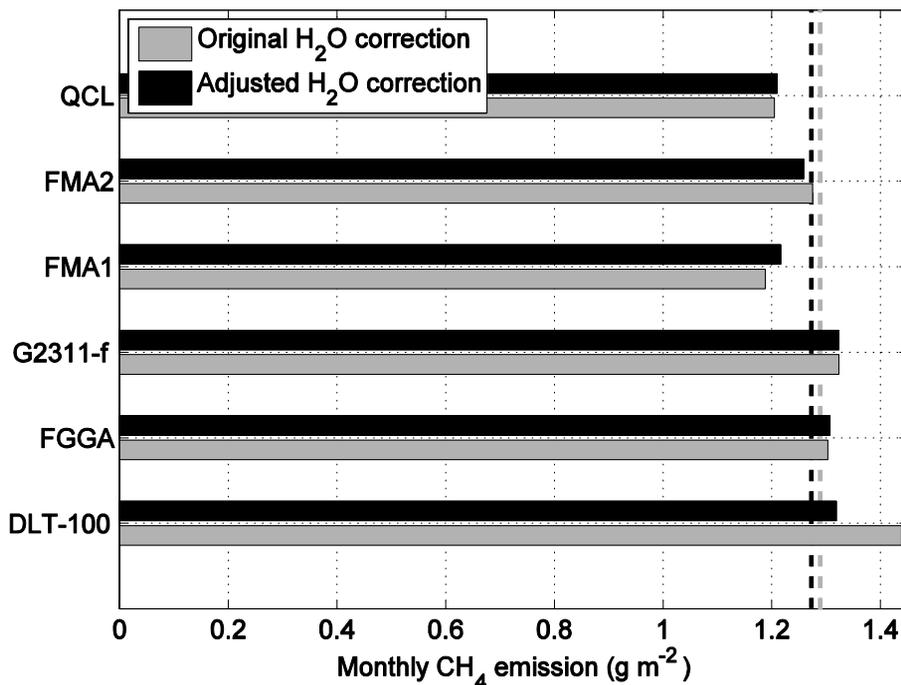


Fig. 13. Monthly CH₄ emissions estimated from mean CH₄ fluxes observed during part of the campaign (from 12 to 25 June). Data corrected with the original H₂O correction are shown with grey bars and with the adjusted H₂O correction ($-kF_{\text{CH}_4}^{\text{WPL}}$ added to the fluxes) are shown with black bars. Vertical dashed lines show the mean values.