Atmos. Chem. Phys. Discuss., 7, S6659–S6668, 2007
www.atmos-chem-phys-discuss.net/7/S6659/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "A compact and stable eddy covariance set-up for methane measurements using off-axis integrated cavity output spectroscopy" *by* D. M. D. Hendriks et al.

D. M. D. Hendriks et al.

Received and published: 8 November 2007

Dear referee,

We would like to thank you a lot for reviewing our paper A compact and stable eddy covariance set-up for methane measurements using off-axis integrated cavity output spectroscopy. All comments have been read carefully and when necessary we adjusted text and figures according to your comments. Below, you will find all our responses to your remarks. First, our responses considering contents, analyses, tables and figures are discussed. Second, our replies to the merely textual comments are listed. We think that the manuscript has improved a lot from all the adjustments.

Best regards,

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Interactive Discussion
Discussion Paper

On behalf of all authors, Dimmie Hendriks

General comment

As suggested by the referee, the organization, presentation and language are adjusted in order to increase the clarity of the paper. Additionally, the spectral analyses is improved and clarified as well as the other issues mentioned by the referee. All comments of the referee are taken into account and our response is listed below.

Organization of the paper

The organization of the paper is changed to the traditional Abstract/Introduction/Methods and Materials/ Results/Conclusions and Discussion format. Paragraph two is divided in seven sub-paragraphs: 2.1 Instrument design 2.2 Site description 2.3 Assessment of instrument stability, precision and accuracy 2.4 Incorporation of FMA in eddy covariance system 2.5 Assessment of FMA in measuring CH4 fluxes 2.6 Data processing 2.7 Simulation of alternative flux measurement approaches Paragraph three, is divided in three sub-paragraphs: 3.1 First data series 3.2 Intercomparison with flux chamber data 3.3 Simulation of alternative flux measurement approaches

In order to require the correct order of subjects, blocks of text are shifted. These shifts were applied after all other changes due to specific comments of both referees were made.

Specific comments

Page 11590 Line 7: Description of the site is extended to the passage below and is shifted to the methods section. Besides testing in the laboratory, the FMA was tested in an eddy covariance set-up at the Horstermeer measurement site. This site is located in a eutrophic peat meadow area in the central part of the Netherlands and was described extensively by Hendriks et al. (2007). The area has a flat topography and vegetation consists of grasses, small shrubs and reeds. Before, CH4 fluxes in the

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area were measured with the flux chamber technique and variation between three land elements was observed: emissions from the saturated land and water surfaces were high compared to the relatively dry land. The annual weighted CH4 emission at the site over 2005 and 2006 was estimated 83.95 ś 54.81 nmol m-2 s-1 (Hendriks at al., 2007).

Page 11591 Line 10-25: Seems to be a lot of ideas here, can you clarify or split in more paragraphs? - Text is reorganised and split in more paragraphs in order to improve clarity.

Page 11592 Line 13: What is the Swagelok part No.? - This is part No.: SS-4FW4-2. Information is added to the text. Equation 2: Define t1 and t2 and say A refers to a CH4 concentration. - The sentence in line 12 is extended to …. calculating the time averaged product (over the period t1 to t2) of the deviation (s’) of the….. We think this remark on A concerns Equation 3 instead of 2. The sentence on line 7 is extended to In this equation A is an average of the CH4 concentration, ….

Page 11594 Line 19: The y-axis interception point implies a slope. This is just a minimum value, so does not need to be extrapolated to the Y axis. - Interpolation is removed from the figure. Line 20-24: Can you provide some context for the square root of the Allen Number: significance? What should we compare a given value to? - The following sentence is added at line 20 on page 11593: The precision of the concentration measurements should be at least a few parts per thousand of the ambient mixing ratio. In the case of CH4 measurements this is approximately 4 ppb for a mixing ratio of approximately 1800 ppb (outside air) (Kroon et al, 2007). The following sentence is added at line 23 on page 11594: Precisions of 0.3 ppb ppb Hz-1/2 (Nelson et al., 2004) and 2.9 ppb Hz-1/2 (Kroon et al., 2007) were found in previous studies of QCL instruments.

Page 11595 Line 2-7: P and T sensitivity should be tested under controlled laboratory conditions. - additional analyses was done over three hours in the laboratory with rising

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Tcell and fluctuating P. - Passage to describe results is changed in accordance to the new figures:

The influence of changing temperature and pressure conditions in the measurement cell (Tcell and Pcell) on CH4 concentration measurements was assessed in the laboratory. During a time series of continuous measurements, a step change in Pcell was induced, while Tcell increased steadily. An effect of the increase of Tcell on CH4 concentration was observed neither from the time series, nor from the correlation plots (Fig. 3). A rapid decrease in the CH4 concentration data was observed 990 sec after the sharp decrease in Pcel. However, this feature did not result in a clear correlation between CH4 concentration and Pcell. Also, this type of Pcell changes did not occur under normal circumstances when Pcell had a stable value near 210 hPa.

Page 11596 Line 14: τ should be 0.11 not 0.10 - Passage about instrument response time is clarified, according to remarks of referee #2.

Page 11596 Line 4-9: Section should be expanded to demonstrate that the instrument response to CH4 is not contaminated by temperature and water vapour fluctuations. - The FMA uses a distributed feedback (DFB) diode laser with a wavelength of nearly 1.65 μm. This DFB diode laser offers narrow line width and features a grating structure within the semi-conductor, which narrows the wavelength spectrum and guarantees single-frequency emission. Water vapour will therefore not interfere with the CH4 measurements, which might be the case with some older techniques. Furthermore, if much water vapour would accumulate in the measurement cell, this would have an effect on the reflectivity of the mirrors and automatically, the MRT would decrease which can be observed at the screen and in the data files. When the MRT drops below 3.0 to 3.5 μs, the measurement cell and mirrors should be cleaned. The effect of temperature in the measurement cell is discussed in the paper and in figure 4. This explanation is improved, see above. Additionally, to correct for density fluctuations arising from temperature and water vapour fluctuations the Webb corrections for closed path eddy covariance are applied, see explanation and references on lines 10-20.

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Line 12-14: Please clarify - It was described by Leuning and Moncrieff (1990) that when the tube length $>$ 1000 times the tube diameter the air brought into the measurement cell can be considered at a constant temperature and the Webb-correction for temperature is not necessary. Here, the tube diameter = 0.0064 m and tube length = ś7 m. - The reference Leuning and Moncrieff (1990) is added to the text.

Line 19-24: are reworded to: The travel time of the air in the closed path set-up from the inlet filter to the FMA, caused a time lag with respect to the in situ measured wind data. For 20 half hour periods, the covariance w’[CH4]’ of w’ at instance t = 0.0 s and [CH4]’ at t = 0.0 s was determined, as well as the covariance of w’ at t = 0.0 s and [CH4]’ at the t = 0.1 s, t = 0.2 s, t = 0.3 s, …, t = 2.0 s. For all half hour periods the highest value of w’[CH4]’ occurred with [CH4]’ at t = 0.6 s. For the calculation of the actual covariance the time lag of the CH4 measurements compared to the wind measurements was therefore taken as 0.6 s.

Page 11598 Line 20-24: How were uncertainties calculated? - We decided to focus on the types of gaps occurring (initial gaps due to instrument failure, removed spikes and gaps due to u* correction). Please find the adjusted text at the response to referee #2. Additionally, we decided that gap filling and thorough analyses of uncertainty percentages are beyond the scope of this paper. Gaps will not be filled and further uncertainties were probably very small regarding the very good stability and precision of the instrument. Line 9: How did you decide on the critical u* value? What are the R2 of the linear fits? - For more clarity, the figure is changed slightly. Instead of linear fits, the w’[CH4]’ data are binned over u* categories of 0.02 m s-1. In the graph a drop in w’[CH4]’ values at ˜0.09 m s-1 can be observed. CO2 values are left out, since this was described in other papers and a reference is provided: Dolman et al. (2004).

During night time periods with low friction velocity (u*) the turbulence of the atmosphere can become too low for the performance of eddy covariance measurements (Wohlfahrt

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et al., 2005; Dolman et al., 2004). In order to determine the critical u* value for CH4 eddy covariance measurements at this specific site, the CH4 flux data and u* data from periods with incoming shortwave radiation (SWin) $<$ 20W m-2 were selected (Dolman et al., 2004). The nightly CH4 fluxes showed a significant decrease for periods with u* $<$ 0.09 m s-1 (Fig. 7). This result is similar to the critical u* value of 0.10 m s-1 found for CO2 fluxes at the same site (Hendriks et al., 2007).

Page 11599 Line 14: What do you mean by statistical reliability? - This means that there are more samples over the same period of time, generating a more precise observation of the wind velocity or atmospheric scalar over this period. Line 15: Provide a reference for EC studies with lower sampling rate. - (Rinne et al., 2000 and 2001; Graus et al., 2006; Businger and Oncley, 1990) is added to the text here.

Page 11601 Line 18-23: How would you go about the correction factor in other situations? - The b-value for the momentum of CO2 and water vapour fluxes (both measured by the open path Licor) was similar to that for the CH4 fluxes measured with the FMA. Since these variables are often measured at eddy covariance sites even where an external power source is not available, the b-value found with manipulation of those data will give a good indication of the b-value needed for the REA system. - Information on this subject will be added to the text and in Table 2.

Line 24-27: The mechanics of sampling (valve switching) in disjunct eddy covariance and REA might introduce additional artefacts. Please comment. - Finally, it should be taken into account that the ‘snap sampling’ instrument for disjunct eddy covariance and the valve switching system for REA will introduce additional artefacts, which also requires certain amounts of power (Rinne et al., 2000; Kuhn et al., 2005; Graus et al., 2006). is added to the text at the end of the Conclusions an Discussion section.

Page 11607 Table 1: It is not clear how the deviation was calculated. - To clarify, the following passage is added to the text:

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The deviation from the standard values of CH4 (MD) was calculated for each measurement by Eq.(4):

MD = (4) where M is the measured CH4 concentration in ppb and S is the CH4 concentration of the standard gas in ppb. A two-point calibration factor (Fcal) was calculated for each set of measurements by Eq. (5):

Fcal = (5) where SH and SL are the low and high standard values of CH4 in ppb and MH and ML are the high measured values of CH4 in ppb. Although the measured concentration sometimes varied one or two ppb from the standard values over the experimental period for both gases, no actual drift was observed in the instrument (Table 1). Fcal was 1.000 on average with fluctuations $<$ 0.30%, indicating high accuracy of the FMA.

Table 1: Does the fact that calibration gases are so different in concentration have implications for the need to resolve very small concentration differences? Can you prove that device responds linear to concentration? - In order to check the stability and precision of the instrument at shorter time scales and small concentrations differences, the Allan variance graph was calculated, showing no effects from drift of the instrument and a high precision required for the determination of small concentration fluctuations. Unfortunately, a third calibration gas was not available. However, the measured fractional absorption of light at the methane resonant wavelength is an absolute measurement of the methane in the cell and since the calibration factor of the two-gas experiment changed only very slightly we think it is reasonable to conclude that no drift occurs and the instrument responds linear to concentration.

Figure 1: Dimension information is added to the figure. Path length can be found in the text (page 11591 at line 2). Average number of passes (1 – 10Œ104 passes) is added to the text on page 11591 at line 2. R, P and T are respectively the reflectivity of the mirrors, the pressure sensor and the temperature sensor. This is added to the caption.

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Figure 2: We think that the description of the parts is clearest as a list next to the picture. Figure is extended and information on length of tubing and dimensions is provided in the figure.

Figure 3: Indeed Time series of temperature in comparison with methane (fluxes) is very interesting. However, we decided not to go into the physical processes behind methane emissions, but explore this in a next paper because the topic is too large and comprehensive to take up in this paper. The scale on the y-axis of the lower panel is expanded and the panels are labeled (a) and (b). The burp at 220 sec. We do not precisely know what caused the burp at 220 sec. It might be caused by some air coming in the laboratory from the technical working space adjacent to the laboratory.

Figure 4: The pressure-axis is corrected. Additionally, this figure is split up into several sub figures: time series of CH4, Tcell and Pcell as well as scatter plots for Tcell vs CH4 and Pcell vs CH4.

Figure 5: Table is removed and equation is shifted to text (see response to referee #2). Abbreviations can be explained as: dCobs is the increase of [CH4] at time t : [CH4]t - [CH4]t=0 and dCincr is difference between the final [CH4] and [CH4] at the starting time: [CH4]final - [CH4]t=0. By this transformation the data were normalized. The following is added to the text: The data in the graph (Fig. 5) are normalized by the transformation dCobs/dCincr, where dCobs is the increase of [CH4] between the starting time and time t and dCincr is difference between the final [CH4] and [CH4] at the starting time.

Figure 6: This analysis is refined to power- and co-spectra averaged over longer time periods which are compared to the theoretical spectra. There are now three graphs in the figure: the w power-spectrum (observed and theoretical), the [CH4] power-spectrum (observed and theoretical) and the w’[CH4]’ (observed and theoretical).

Figure 7: The figure is changed slightly. Instead of linear fits, the

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w’[CH4]’ data is binned over u* categories, showing a drop in w’[CH4]’ values at ˜0.09 m s-1. Nighttime is defined by all periods with incoming short wave radiation (SWin) $<$ 20 W m-2. The caption is changed Results of the analyses of the effect of low turbulence on nightly CH4 fluxes, showing a drop in flux magnitude below of u* of 0.09 m s-1. Data are binned over u* classes of 0.02 m s-1 and error bars show the standard deviation per class.

Figure 9: How did you propagate your error - See response to referee #2 - no error is introduced here (see response to referee #1). Last sentence of the caption is changed to The square marks show the weighed average of the flux chamber measurements.

Text editing (Since referee #2 made more remarks considering text editing, the remarks of referee #2 are dealt with first. Remarks that were made by both referees are not repeated in the referee #1 part.)

Abstract Line 6-7: Statistical testing, a calibration experiment and comparison with high tower data showed high precision and very good stability of the instrument. is changed to Statistical testing, a calibration experiment and comparison with tower data showed high precision (7.81Œ10-3 ppb) of the instrument, while no drift was observed. Line 14: emissions is changed to emission rates. Line 19: slower measurement rates is changed to lower sampling frequencies.

Page 11591 Line 3: (IPCC, 2007) is changed to (Foster et al, 2007). Line 8: Global Warming Potential (GWP) of methane is changed to Global Warming Potential (GWP) of methane expressed as CO2-equivalents. Line 24: proved to be is changed to was.

Line 5: might change as a result of changes in reflectivity of the mirrors in the cell is changed to is determined by the reflectivity of the mirrors in the measurement cell. Line 10-11: is claimed to have an accuracy of at least 1.0% and a precision of 0.1% and to operate autonomously. is removed. Line 14: In case of a measurement speed higher than 1Hz is changed to When sampling rates exceed 1Hz. Line 17: The text The MRT cannot … a dust-free environment. is shifted up to line 19.

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Page 11592 Line 7: type is deleted. Line 20: an under is deleted. Line 27: noise dampener is changed to silencer.

Page 11595 Line 5: It can be observed that is deleted. Line 5: as well as is changed to and. Line 10: moments is changed to instances.

Page 11597 Line 12-14: The method of Nakai et al. (2006) was used is complemented and is therefore changed to Since 3-axis ultrasonic anemometers were found to under measure wind speed at large angles, the method of Nakai et al. (2006) was used…

Page 11598 Line 7: Implied is changed to indicated.

Page 11599 Line 22: 220 V is changed to an external power source.

Page 11560 Line 12: slow is deleted. Line 13: with dt = 1.0 s instead of 0.10 s is changed to at a lower frequency. Line 24: matrixes is changed to matrices.

Page 11561 Line 8: in fact is deleted.

Slow 1Hz eddy covariance is changed to 1Hz eddy covariance throughout whole text.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 11587, 2007.