

## ***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.***

**Anonymous Referee #1**

Received and published: 24 September 2007

### **GENERAL COMMENTS**

This paper describes the DLT-100 Fast Methane Analyzer (FMA), manufactured by Los Gatos Instruments, and assesses its performance as a methane analyzer incorporated into an eddy covariance system. A fast response, field-stable and high sensitivity instrument which does not require liquid nitrogen has been the “Holy Grail” of the community dedicated to quantifying land-atmosphere exchange of CH<sub>4</sub>. Such an instrument is needed for three reasons: (1) spatial and temporal variability of CH<sub>4</sub> exchange is poorly sampled by traditional chamber techniques; (2) micrometeorological measurements integrate exchange at scales of 10<sup>2</sup> m<sup>2</sup> to 10<sup>4</sup> m<sup>2</sup> – appropriate for landscape scale

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studies; (3) alternative eddy covariance measurements require frequent recalibration and/or liquid nitrogen in the field, which creates logistic limitations to unattended field deployment in remote locations. As such, I believe that this work is important, and the availability of an instrument that performs satisfactorily for eddy covariance  $\text{CH}_4$  measurements would be of huge importance to the biogeochemistry/trace gas community. For this reason, the study falls well within the scope of ACP.

The study has analyzed the instrument's precision, stability, response characteristics and performance in eddy covariance mode. I think the authors have considered the most important scientific issues in the assessment of the instrument for eddy covariance measurements. However, I have some concerns about the conclusions drawn from some of the analyses. I also feel that the paper seems hastily drafted, and would benefit from major reorganization and polishing.

I have therefore evaluated the manuscript as follows, and provide justification for this evaluation below.

### Scientific Significance

Current  $\text{CH}_4$  analyzer technology fall short of being used as eddy covariance detectors for three reasons: (1) response time is too slow; (2) liquid nitrogen is required in the field; (3) inability to resolve 2-5 ppbv  $\text{CH}_4$  differences. The authors present an instrument which they claim overcomes these shortcomings. The present inability to make long-term continuous measurements of  $\text{CH}_4$  emission has hampered efforts to understand the temporal dynamics of  $\text{CH}_4$  emission, and its relationship to environmental drivers. The simultaneous measurement of  $\text{CH}_4$  and  $\text{CO}_2$  exchange would be useful in working out net greenhouse gas exchange ( $\text{CO}_2$  plus  $\text{CH}_4$ ) from terrestrial ecosystems. Long-term eddy flux measurements of  $\text{CO}_2$  exchange are now standard. An instrument with the aforementioned attributes would make eddy flux measurements of  $\text{CH}_4$  exchange also similarly routine.

## Scientific Quality and Presentation

Unfortunately, the presentation of this manuscript needs further work, without which it is difficult to assess the quality of this research. I have provided extensive comments on specific line numbers below.

For those interested in an instrument that can measure  $\text{CH}_4$  in eddy covariance mode, the big question is whether the instrument is sufficiently responsive. Therefore, most researchers will be most interested in Figure 6. This figure could be split into more panels, so that the each family of co-spectra would have its own y-scale. The panel of most interest to those assessing the instrument for eddy covariance mode would be the “Observed  $w\text{CH}_4$ ” family of curves. The reader needs to be informed why (a) there are 5 curves instead of four, (b) whether the fact that there is a variety of negative slopes (some that look like they decline at a rate substantially less than  $-2/3$ ) is significant.

Many other organization, presentation and language issues exist in this manuscript, some which are merely distracting, but others that make it difficult to assess the quality of the science.

## Organization of the Paper

I don't know the flexibility that ACP has in the way that articles are ordered, but this manuscript does not keep to the usual Abstract/Introduction/Methods and Materials/Results/Discussion format. I think the paper would greatly benefit from an organization close to this traditional format. Currently, important issues are raised in the methods and results section, new methods are raised in the results etc. I suggest that the introduction be much more focused on the describing the background and challenges of measuring methane in the field with eddy covariance, so the reader is clear why this issue is important. Currently the first paragraph makes an abrupt transition from talking about atmospheric  $\text{CH}_4$  to talking about eddy covariance. Something should be added about measurement of trace gases fluxes from land surfaces, and approaches to quantifying these.

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The final paragraph of the introduction should describe the objectives of the study, but does not need to include specifics about the instrument or the field site.

I think the materials and methods section could be structured something like:

2.1 Instrument Design

2.2 Assessment of Instrument Precision, stability, accuracy

2.3 Incorporation of the FMA into an eddy covariance system

2.4 Assessment of FMA in measuring CH<sub>4</sub> fluxes by eddy covariance

2.5 Simulation of alternative eddy covariance approaches

The manuscript needs further proofing, consistency with nomenclature and improvements in presentation

## **SPECIFIC COMMENTS**

### ***Abstract***

Brand names shouldn't be used in Abstract, try "Infra-red Gas Analyzer (IRGA)"

Page 11588, Line 7. This sentence is vague and ambiguous

Page 11588, Line 14 replace "emissions" by "emission rates"

Page 11588, Line 19. The other techniques are not necessarily slower, eg, DEA is just as fast but has a lower sampling frequency

### ***Introduction***

Page 11589, Line 3. IPCC is normally cited by the Chapter Author

Page 11589, Line 8. Is GWP on a mass/mass basis?

Page 11589, Line 6. .When defining an acronym, be consistent in how you capitalize the long version. This problem occurs throughout the manuscript.

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Page 11589, Line 16. replace “eighties” with 1980’s

Page 11589, Line 21. replace “refill” with “refilled”

Page 11589, Line 24. replace “proved to be” with “was”

Page 11590, Line 7. Describe Site in Methods

## **Section 2**

Page 11591, Line 5. Would be useful to know how the MRT is controlled. Does the measurement rate (10 Hz or whatever) specify how fast the MRT goes?

Page 11591, Line 14. Change sentence start to “When sampling rates exceed 1Hz, . . .”

Page 11591, Line 10-25. Seems to be a lot of ideas here, can you clarify or split into more paragraphs

Page 11591, Line 17. Make the precision of the figures you cite more consistent, eg, change “3  $\mu\text{s}$  to 3.5  $\mu\text{s}$ ” to “3.0  $\mu\text{s}$  to 3.5  $\mu\text{s}$ ” This problem occurs elsewhere in the manuscript

Page 11592, Line 7. Delete “type”

Page 11592, Line 13. What is the Swagelok part No.?

Page 11592, Line 20. delete “an under”

Page 11592, Line 26 and elsewhere. replace “trough” with “through”

Page 11592, Line 27. use the word “silencer” instead of dampener

## **Results**

Equation 2. Define  $t_1$  and  $t_2$  and say that A refers to a  $\text{CH}_4$  concentration

Page 11593, Line 11. Delete space in middle of purpose

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.

Page 11594, Line 20-24. Can you provide some context for the square root of the Allen Number: what is its significance? What should we compare a given value to?

Page 11595, Line 2-7. Pressure (P) and temperature (T) sensitivity should be tested under controlled laboratory conditions. Measuring T and P under field conditions and relating it to the measured concentration of CH<sub>4</sub> is a confounded analysis.

Page 11595, Line 4. Replace “cel” with “cell”

Page 11595, Line 5. Delete “It can be observed that”. Start with “Relatively. . .”

Page 11595, Line 5. Delete “as well as”. Replace with “ and

Page 11595, Line 6. Subscript “cell”

Page 11595, Line 10. Replace “moments” with “instances” or “occasions”.

Page 11595, Line 6. You state that CH<sub>4</sub> concentrations are independent of P<sub>cell</sub> but at ~13 minutes there is a step change in both P<sub>cell</sub> and CH<sub>4</sub>. Please comment.

Page 11595, Line 16-27. Delete “It can be observed that”. Start with “Relatively. . .”

Page 11596

Line 14.  $\tau$  should be 0.11 not 0.10

Line 19-24. Reword this sentence

Section 3.2. Seems like this should be in methods section.

Page 11597

Line 4-9.

I feel that this section could be expanded because it is crucial to demonstrate that the instrument response to CH<sub>4</sub> is not contaminated by temperature or water vapor

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fluctuations.

Line 12-14. Please clarify: readers (myself included) maybe unfamiliar with this issue.

Line 20. Reference to Figure 6. More clarification is needed. The potential of this instrument to be used in eddy covariance mode depends on whether the sufficient flux is found in the inertial sub-range. How were four 30-minute periods selected? Why are there five lines? Define “comparable”? Not all lines have a slope of  $-2/3$  for the  $w$ -  $\text{CH}_4$  co-spectra.

Page 11598

Line 3. I don't think  $[\text{CH}_4]$  is a conventional abbreviation for  $\text{CH}_4$  concentration in air.

Line 7. Replace “implied” with “indicated”

Line 9. How did you decide on the critical  $u^*$  value of  $0.09 \text{ m s}^{-1}$

Line 9. What are the  $r^2$  of the linear fits

Line 20-24: How were the uncertainties calculated? What proportion are due to instrumental error? What proportion are due to other sources of error?

Page 11599

Line 9. October-3

Line 14 What do you mean by “statistical reliability”?

Line 15. Provide a reference for Eddy Covariance studies that use a lower sampling rate.

Line 22. Replace “220 V” with “an external power source”.

Page 11600

Line 12 . Delete “slow”.

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Line 13. Replace “with  $dt = \dots 0.10$  s” with “at a lower frequency”

Line 24. Replace “matrixes” with “matrices”

Page 11601.

Line 8. Delete “in fact”

Line 18-23. How would you go about calculate the correction factor in other situations?

Line 24-27. The mechanics of sampling (valve switching) in disjunct eddy covariance and relaxed eddy accumulation might introduce additional artifacts. Please comment.

Page 11607

Table 1. It is not clear how the deviation was calculated. For example, in the first row of the table, the FMA measured 125 ppb with the lower concentration calibration gas, which is exactly right, but the deviation is rated at 3.3%. If the deviation is a measure of the analytical precision (eg standard deviation/mean) than this needs to be stated as such.

Also, does the fact that the two calibration gases are so different in concentration (125 vs 2002ppb) have implications for the need to resolve very small concentration differences in eddy covariance mode? Can you provide evidence that the detector responds linearly to concentration (perhaps all that is needed is a three point curve, the first point being at zero concentration, and the second and third at 125 and 2002 ppb).

Figure 1. It would be useful to have some dimension information on this figure, eg, the length of the cell, the path length and average number of passes. Also what does “R,” “T” and “P” mean?

Figure 2. The key (describing parts 1-17) should be in the caption The rest of the figure should be expanded so that it is easier to understand. The length of the tubing and the height of the tower needs to be stated. The LI-7500 is not a temperature analyzer. The direction of the data flow probably is not needed.

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Figure 3. The time series of temperature (sonic) would also be nice to see since this is a would provide a good indication of whether  $\text{CH}_4$  is well correlated to an easily measured scalar. The scale of the y-axis in the lower panel needs expanding. The caption should describe what the  $6.1 \times 10^{-4}$  means. The panels need labeling (a) and (b). Comment on the 40 ppbv “burp” at 220 sec

Figure 4. The left hand axis is ok for  $\text{CH}_4$  concentration but is not correct for hPa

Figure 5.

Don't combine a figure and a table.

Define the abbreviations on the Y-axis

Put the equation in the text.

What is the normalization you refer to?.

Figure 6.

Label panels

What are the slopes that you compare to the required  $-2/3$ ?

How different are the slopes?

What are the different lines? How were they selected from the larger data set?

I don't know how to assess these but I feel that this figure is the most crucial in the paper, and is the definitive proof of whether the analyzer performs adequately for eddy covariance.

Why do some of the lines deviate from the  $-2/3$  slope? Are the deviations significant?

Figure 7.

What are the  $r^2$  of the trends?

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How did you decide on the  $u^*$  of  $0.09 \text{ m s}^{-1}$ ?

How did you define nighttime?

Use  $\mu\text{mol}$  not  $\text{umol}$  for y-axis label.

Figure 9.

How did you propagate your error?

Replace “Additionally,” with “The square marks show”. Delete “is shown”.

**Explicit response to the questions that ACP invite the referees to consider.**

**1. *Does the paper address relevant scientific questions within the scope of ACP?***

Yes.

**2. *Does the paper present novel concepts, ideas, tools, or data?***

Yes, it presents a novel instrument to be employed in eddy covariance studies of land-atmosphere exchange of  $\text{CH}_4$  flux.

**3. *Are substantial conclusions reached?***

Yes, the conclusion is that the instrument is suitable for deployment in eddy covariance systems

**4. *Are the scientific methods and assumptions valid and clearly outlined?***

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Most of the time, but in three aspects more information, or other tests were needed, eg,

(a) the test of whether the instrument responded to temperature and pressure fluctuations should have been made in the laboratory not in the field.

(b) Some of the assumptions (such as the decision of using a  $u^*$  criteria of  $0.09 \text{ m s}^{-1}$ ) were not clearly outlined.

(c) How it was determined that the co-spectra fitted closely to a fall-off line of  $-2/3$  in the inertial sub-range?

The plots (Figure 6) are a log-log axis so small deviations from the  $-2/3$  slope may be significant. I am not qualified to assess how big the deviation may be before is unacceptable. However, I think this warrants discussion. The merit of this instrument will depend largely on whether this instrument has sufficient high-frequency response. There is no description on how the five sets of data were selected (and in fact in the text it says that only four data sets were selected).

**5. *Are the results sufficient to support the interpretations and conclusions?***

Not always. See 4(c) above.

**6. *Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?***

No. There needs to be better description of how some of the quantities were calculated.

**7. *Do the authors give proper credit to related work and clearly indicate their own new/original contribution?***

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Yes

8. ***Does the title clearly reflect the contents of the paper?***

I think the title is a little ambiguous. The word “set-up” should be replaced by “system”, throughout the paper. It is not clear whether stable refers to the eddy covariance system or the instrument itself.

9. ***Does the abstract provide a concise and complete summary?***

Mostly, although some problems listed below.

10. ***Is the overall presentation well structured and clear?***

No. Many problems with figure presentation, language and organization. See comments below.

11. ***Is the language fluent and precise?***

No. See comments below

12. ***Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?***

No. See comments below

13. ***Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?***

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Yes. See comments below

14. ***Are the number and quality of references appropriate?***

Some problems with references. IPCC cited incorrectly (perhaps). Eugster and Penn (1994) reference is missing from reference list.

15. **Is the amount and quality of supplementary material appropriate?**

Yes.

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

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