Biogeosciences Discuss., 4, S583–S589, 2007
www.biogeosciences-discuss.net/4/S583/2007/

Biogeosciences Discussions

Interactive comment on "Suitability of quantum cascade laser spectrometry for CH_4 and N_2O eddy covariance measurements" *by* P. S. Kroon et al.

Anonymous Referee #1

Received and published: 29 May 2007

1 General aspects

This paper investigates the suitability of quantum cascade laser (QCL) based spectrometry for eddy covariance (EC) flux measurements of N2O and CH4. The paper addresses a relevant scientific question and fits well within the scope of BG. EC has a number of advantages over other flux measurements as has correctly been pointed out by the authors, and instrumentation based on QC lasers is expected to provide the necessary analytical quality. The authors also correctly state that only few publications describe field applications, and that it is important to fill this gap. One of the main interests of this paper would be to check the performance that was determined in the lab (e.g. by Nelson et al.) in a field application. The criteria (continuity, sampling frequency, precision, stationarity) are, however, badly defined. Furthermore, the study evaluates

precision, stationarity and sampling frequency again in the lab, which is nothing really new. It would be relevant to know what the values for these performance criteria are under field conditions. It should also be noted, that the authors strongly rely on the criteria suggested by Nelson et al., in paper which is focused on spectroscopy, and not on EC flux measurements. The sequence of these criteria is not systematic throughout the text, which makes it unnecessary difficult to read. The language is often not precise enough. As a general impression, the paper should very carefully be reviewed for content and style before publication. I have made a detailed list of remarks for the abstract and the first chapter. Some of these are personal preferences; however I believe that the whole paper should be reviewed by the (very competent) authors at this level of detail.

2 Major comments

1. One of the key factors for EC flux measurements is the response time of the whole system (sampling + analyzer). Unfortunately, it seems that this was not determined experimentally in the field. This should be made clear, and the statement of a 10 Hz sampling frequency avoided at least in the abstract, because it gives a completely false impression. Can you derive the bandwidth based on the field data, or indicate why it was not possible to determine this experimentally for the field setup? The bandwitdth given on page 1147/5 is a good approach. However, based on the values (400 l min-1 pump; 0.5 l cell) one would under optimal conditions (without sampling) get a value of 2.1 Hz, and not ca. 3 Hz as stated.

2. Table 2 and the corresponding text are not clear enough. They are probably only meaningful for this specific instrument and can be left out.

3. Precision was found to be in agreement with the original paper (Nelson, 2004). This is nice, but it would be interesting to know what the precision was in the field. Are the calibrations long enough to be used for Allan plots, or would you have other field data to do this?

BGD
4, S583–S589, 2007

EGU

4. The Webb-correction for water vapour is critical. A very significant part of the values given in Fig. 3 are within the boundaries given by the estimation of the maximal Webb-corrections (page 1151/11-15). Using latent heat fluxes from analyzers that do not have the same sampling setup may not be appropriate. Another option would be to dry the sample. Why was this not done? What would the limitations be?

5. In Fig. 4, the minimum for the delay time is distinctly shifted for N2O (negative flux). How can this be explained?

6. The information given in Table 3 should be replaced by a graph giving the values and standard deviation of all calibration sessions. This would give a much better idea of the stability of the system. There is the danger of doing calibrations always under similar conditions, e.g. same time of the day, after new alignment, etc. Therefore, it would be very helpful to have frequent measurements of a constant sample (gas bottle) with typical measurement conditions (e.g. without having a fresh background each time) and over periods with marked temperature changes (day/night).

3 Minor comments

1. One of the major advantages of QCLs is that they can be TE cooled. Still, the authors use N2(l) for detector cooling. It would be important to know how much one would loose with respect to EC applications if the TEC detectors were employed.

2. Only one laser was used in the system. Would the performance be influenced by the second laser? Was there any specific reason why no second laser was used?

3. Use consistent syntax for dates (e.g. page 1140/18)

4. The values given on page 1141/27 imply a tuning range of 3 cm-1. This seems very large for QCL in the described setup.

5. Page 1142/5: QCL-software should be replaced by TDL Wintel.

6. Page 1142/20: what exactly was stabilized by the water circuit?

BGD
4, S583–S589, 2007

EGU

7. Page 1143/15: define variables

8. Page 1148/8: it would be better to use values determined for the instrument used in this study, and not based on a similar instrument used for the publication of Nelson et al.

9. Page 1149/1: How was it proven that the averaging time of 30 min was long enough?

10. Page 1149/23-25: The interesting part would be: how much do the calibration factors (and standard deviation) change between calibrating (and tuning) of the instrument.

11. Page 1150/4: Was a 0.1 Torr stability required and/or obtained?

12. It might be interesting for the reader to have references to the very recent (parallel) publications be Eugster et al. (BGD) and Neftel et al. (Tellus B).

4 Detailed comments on abstract and chapter 1

Page 1138

Title: ...eddy covariance flux measurements

Line 1: eddy covariance flux measurements

Line 2: continuous field measurements

Line 5: All four required criteria...related to continuity, sampling frequency, precision and stationarity were checked.
Do you mean: Various criteria related to ... or: The four criteria continuity, sampling frequency ...

Line 6: The system had been running...
Check tense

Line 9: A sampling frequency of 10 Hz... using a 1 GHz PC system.
Sampling is ambiguous in this context. Do you mean: absorption spectra were ana-

BGD
4, S583–S589, 2007

lyzed for CH4 and N2O at 10 Hz. Is the clock rate of the PC of such importance that it should appear in the abstract?

Line 10: A precision of... was obtained for CH4 and N2O, respectively.
Since you talk of field measurements just above, it would be important to specify that this was in the lab.

Line 11: Drift in the system was removed...
Was drift really removed or is the filter not rather a method to reduce drift induced effects?

Line 14: reread sentence (use of average).

Line 14: flux units
Is it important to have two different units in the abstract, and if so could you make the sentence more reader friendly? The reader has to deduce from ngC m-2 s-1 that this is CH4, and that the next value in brackets also refers to CH4.

Line 15: average
Given that 40

Line 17: no capitals for methane and nitrous oxide

Line 17 and 20: repeated use of “major”.
Is the term appropriate for both statements?

Line 24: the term obtain is not well chosen. Other options would be to “quantify emissions” or "determine emission rates".

Line 25: repeated use of “obtain”

Line 26: convenient does not seem to me a well chosen attribute for this type of instrumentation

Line 26: ..... instrumentation... meets...

BGD
4, S583–S589, 2007

Page 1139

Line 1: eddy covariance operation is somewhat awkward. Eddy covariance measurement would be better.

Line 3: Pointing towards the limitation of TDLS is very critical here, because it creates the research gap that this paper should fill, i.e. the QCL system used here should be better than the TDLS analyzers used in the referenced papers. I would suggest to rethink again about the two criteria (drift and insensitivity) that are considered in this context, because (i) drift is a limitation that the instrument used for this study seems to suffer strongly from too, and which is very badly quantified by the presented data, and (ii) insensitivity is a rather vague term, which is not further defined in this context. If you mean precision, then it should be noted, that the cited studies have partly used instruments with a precision that is very comparable to the QCL system.

Line 6: QCL is typically used as an abbreviation for quantum cascade lasers (and not ... spectrometers). This is not precise throughout the text. Check all uses of QCL and replace (where appropriate) by QC laser or QCL spectrometer, etc.

Line 6: This statement about instruments is simply wrong. It is true that QC lasers are inherently more stable than lead salt lasers. They tend to have more power and show no mode hopping (to cite just a few advantages). However, this does not make all QCL based instruments more sensitive than TDL-systems, especially if you consider the spectroscopic options that the cw operation of TDLs offers.

Line 8: It is certainly appropriate to cite the Faist (1994) paper. However, is this really a publication about spectroscopy?

Line 8/9: The cited papers (Nelson, Jimenez) do not give detailed descriptions of the working principles of quantum cascade lasers.

Line 16: ... quantum cascade lasers can meet these criteria...
No, quantum cascade lasers are nothing else than (excellent) light sources.

BGD

4, S583–S589, 2007

Interactive Comment

Line 17: ... this conclusion... based on technical and theoretical considerations.
What are technical considerations in this context? You should state that Nelson's conclusions were also based on experimental results. From there the text should then more clearly state what the reader can expect as additional information from this study.

Line 25: Finally... the discussions are presented...
Awkward

Interactive comment on Biogeosciences Discuss., 4, 1137, 2007.