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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.

P. S. Kroon et al.

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Dear referees,

We would like to thank you very much for your critical notes, useful remarks and advices with respect to the contents of our article. We will improve our final article by means of your comments. We will add some more Figures and texts in the article. Our answers to all your remarks are given below.

Best regards, On behalf of all authors, Petra Kroon

Response in order of the referee numbers:

Answers to comments of referee 1

General aspects

We agree that the criteria should be defined more clearly and that the sequence of these criteria should be systematic throughout the text. Besides, we will evaluate the criteria (continuity, sampling frequency, precision, stationarity) under field conditions.

Major comments

1. We will determine the response time in the field by means of our calibration sessions. Unfortunately, in this case the tube length is shorter than during the real measurements. We will compare the bandwidth belonging to this response time with the given theoretical bandwidth given on page 1147/5. However, we will probably find the same response time and effective bandwidth, since the effective bandwidth should not be a function of the flow rate as long as the pumping speed is constant with cell pressure. The true pumping speed at the cell can be derived by

$$S_{\text{cell}} = \frac{V_{\text{l}}.760}{P_{\text{cell}}} \quad (1)$$

in which Scell indicates the true pumping speed in the cell in $\mathrm{l\,min^{-1}}$, $\mathrm{V_l}$ the flow rate in STP $\mathrm{l\,min^{-1}}$ and $\mathrm{P_{cell}}$ the cell pressure in Torr. By means of the extreme values given on page 1141/18 and page 1145/5, namely 8 standard $\mathrm{l\,min^{-1}}$ at 15 Torr and 22 standard $\mathrm{l\,min^{-1}}$ at 40 Torr; it can be derived that the pumping speed at the cell changes from 405 $\mathrm{l\,min^{-1}}$ to 418 $\mathrm{l\,min^{-1}}$. This is a small change and will not affect the conclusion that the true pumping speed is about 400 $\mathrm{l\,min^{-1}}$. We agree that we made a calculation mistake on page 1147/5. The theoretical bandwidth will be changed into 2

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Hz. Besides, we will indicate in the abstract that the electronically sampling frequency is 10 Hz and the flow response time of 0.07 s corresponds to a bandwidth of 1/(2$\pi\tau$) of 2 Hz.

2. We agree that the content of Table 2 could be conveyed within the text. The main point is that the laser can be operated far enough above the threshold to obtain the necessary short term precision of 0.3 ppb and 3 ppb for N_2O and CH_4, respectively. We agree that the actual laser voltage is specific for this laser and would not be relevant. So, we will omit this statement.

3. In our opinion, the short term precision depends only on alignment and signal level, which should be the same in the laboratory and in the field, but longer term stability of 10 to 100 seconds can be worse in the field than in the laboratory where the temperature may not be controlled as well. In our final article, we will insert an Allan Variance Figure to demonstrate the instrument stability on longer time scales.

4. We agree that a Webb-correction calculation based on latent heat fluxes from analyzers that don't have the same sampling setup is not very appropriate. However, a rough indication can be derived on the magnitude of the Webb-correction for each 30-min flux value. In our opinion, the magnitude of the Webb-correction will be larger for the open path CO_2 system than for our set-up. That's why, an indication is derived of the maximal Webb-correction for our fluxes. A better solution in future experiments will be to dry the sample.

5. The delay time is dependent on the flow in the system and therefore on the pressure in the system. The delay time is determined for each period since it changes due to dirtiness on the inlet filter. So, the main difference between the delay time of the negative N_2O flux and the delay time of the other fluxes is due to the pressure in the system.

6. We agree that it will be better to perform automatic calibrations to avoid doing calibrations always under similar conditions. Unfortunately, we couldn't perform

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automatic calibrations until now. However, we can state the following points: First, we will include two additional Figures to show the dependency of the calibration factor to cell pressure. Second, we will indicate the difference between low, high, low-high calibration factors using both additional graphs and Table 3. Besides, we will make clear that the calibration factors are also dependent on alignment by means of the standard deviation in Table 3. We will improve this Table by using calibration factors measured approximately at the same cell pressure. Finally, we will check if a graph of all calibration factors in time gives a better view of the behavior of the calibration factor.

Minor comments

1. We agree that it will be important to indicate how much one would loose with respect to EC applications if the TE-cooled detectors are used instead of LN2-cooled detectors. We will refer to a nice table in the paper of Nelson et al. 2004 in which TE-cooled and LN2-cooled detectors are compared. The difference in precision is about a factor of three with presently available detectors.

2. The performance will indeed be influenced by the use of a second laser. The use of two lasers affects the electronically sampling frequency. Besides, the penalty in signal-noise ratio would be proportionally smaller by the square root of the duty cycle difference (the amount of measurements). If two lasers are sharing the same sweep equally, the signal-noise ratio on both will be increased by $\sqrt{2}$. The reason for not using the second laser in this case was to maximize the signal to noise ratio, which requires the most precision for flux measurements.

3. We will use consistent syntax for dates.

4. The reviewer is correct. We made a mistake. The $\mathrm{CH_4}$ line is at 1270.8 $\mathrm{cm^{-1}}$ and the $\mathrm{N_2O}$ line at 1271.1 $\mathrm{cm^{-1}}$. The total width of the scan is close to 0.5 $\mathrm{cm^{-1}}$.

5. We will replace QCL-software in TDL Wintel.

6. The water cooling circuit is primarily used to remove the heat generated by the Peltier element, which cools the laser. It also stabilizes the temperature of the pulse electronics to provide a more uniform laser output power, and it stabilizes the position of the laser which rest on the Peltier element.

7. We will define all variables in the final article.

8. We will indicate the minimum in the Allan Variance of our set-up in the final article.

9. The averaging time of 30-min seemed to be long enough by means of spectral analysis.

10. See point six of the major comments

11. The calibration procedure should indeed be performed at the pressure of the measurements seeing that the calibration factors are strongly dependent on the cell pressure (We will include two additional Figures in which this effect will be shown). However, a better match than 0.1 Torr is not required since the uncertainty in calibration factor will be a minor effect when a match of 1 Torr is obtained. This match can easily be obtained.

12. We will refer to both articles in our final article.

Detailed comments on abstract and chapter 1

Page 1138

We will improve page 1138 by means of the detailed comments.

Page 1139

Line 3 and line 6: We agree that we should improve our statement. Our statement in line 6 is not correct. We will modify both lines according to the following statement: "a good TDL-system can obtain similar precision; however, the big advantages of QCLs

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are the better laser frequency stability and freedom from cryogenic cooling cycles that can lead to instability in mode purity and laser frequency."

We will take into account all the other comments.

Answers to comments of referee 3

Specific comments

1. We agree that the extent to which the flux is underestimated is dependent on the Reynolds number of the flow. However, the Reynolds number was higher than 4000 during the total field campaign. The indicated range from 5020 to 1720 is based on the field measurements and some additional test measurements. We will describe this more clearly in our final article. We will separately indicate the Reynolds number range during our field measurements from the total Reynolds number range. Besides, we will better indicate that we prevented the flow becoming less turbulent by regularly replacement of the inlet filters (see page 1145/1).

2. The extent to which the flux is underestimated can be derived from the ogives. This method is also applied in a study of Ammann et al. (2006). The underestimation is indeed dependent on the meteorological circumstances. We will show the extent of the underestimation for some circumstances in our final article. By means of this, it can be concluded that the damping effect can probably be neglected in case of turbulent flow regimes.

3. In our opinion, we need more analysis to investigate the reliability of the negative fluxes. First, the raw data screening could be improved using the Vickers and Mahrt method (Vickers and Mahrt, 1997). Second, the detection limit, based on Wienhold et al. (1995), could be determined for the whole measurement period instead using only one 30-min flux value. Third, the negative fluxes could be analyzed using spectral analysis. We don’t know yet the possible cause of the negative fluxes. However, P.

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Leahy and G. Kiely will submit a paper titled "Micormeteorological observations of nitrous oxide uptake by fertilised grassland" to Biogeosciences within a few weeks in which some possible explanations are given.

Minor corrections

All minor corrections will be taken into account in our final article.

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Thanks a lot for your comments. We will change our manuscript by means of your comments in the following way:

We agree that the manuscript is sometimes a bit confusing. We will try to write everything more clearly in the final article.

Page 1139, lines 13-15: We will clear out the difference between the sampling rate and the response time in the final article.

Page 1142: We will show a schematic representation of the measurement system in the final article.

Page 1143, lines 15-16: We agree that the storage term will go to zero for stationary conditions. We will improve the equation and its description in the final article by means of the following argument. We assume that the circumstances are stationary within a time period of 30-min. Therefore, the average concentration values of CH_4 and N_2O will be determined over each 30-min period. The storage term will be calculated by means of these average 30-min values. In conclusion, a step function will be used to determine the storage term for each 30-min NEE value.

Page 1145, lines 10-13: We will add four co-spectra Figures, namely one Figure which shows the co-spectra of wT and wCH_4 during the night, one Figure which shows the

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co-spectra of wT and wN_2O during the night, one Figure which shows the co-spectra of wT and wCH_4 during the day and one Figure which shows the co-spectra of wT and wN_2O during the day. By means of these Figures, it will be shown that almost no damping effects occurs during these measurements.

Page 1145, lines 18-23: We will add eight ogives Figures belonging to the four co-spectra Figures described by the previous point. We will show four original flux ogives and four flux ogives, by which the same form is given in lower frequencies (Method proposed by Ammann et al. (2006)). However, there will be no big difference between the two types of flux ogives, because almost no damping effects occur in our system.

Page 1146, line 1: The storage term is calculated by means of the average concentration values over 30-min at 3 meter height. Unfortunately, we didn't have additional vertical concentration profile measurements. Therefore, a linear profile was assumed which underestimates the storage term.

Page 1148, lines 14-21: In our opinion, the x-coordinate belonging to the minimum in the Allan Variance plot indicate the point from which laser drift becomes important. Therefore, this x-coordinate, which is indicated by τ_A, represents the stability time.

Page 1148, lines 14-17: We agree that the instrumental drift in the timescales longer than the running mean filter timescale do not affect the flux. Indeed, the instrumental drift could affect the flux in case of using linear detrending or block averaging. The instrumental drift could cause an underestimation or an overestimation, which is dependent on the correlation of the instrumental drift with the vertical wind velocity. We will make this point more clearly in our final article.

Pages 1148-1149, lines 24-4: We will try to express the description of the derivation of the effect of low pass filtering on a more explicit way. Besides, we will change the Figure caption.

Page 1149, lines 20-21: The low and high calibration factors differ a lot. By means of this, it can be stated that there will be a zero offset in our measurement system. Therefore, it will be better to use a high-low standard since the zero-offset should be the same for both. The lowest standard should be lower than the lowest expected atmospheric concentration and the highest standard should be higher that the highest expected concentration. Besides, the calibration factor will be more precise when more calibration standards are used. In our case, we only used two standards.

We agree that the zero offset does not affect fluxes seeing that the zero offset does not affect the gain in the calibration. We will change these sentences.

Page 1150, lines 9-13: No, we have not performed any u* filtering.

Page 1150-1151, lines 15-3: We agree that we should calculate the Wienhold error based on a much longer time period. Besides, we will check if these negative fluxes occur during weak turbulence.

Figure 4: The difference in delay time is caused by a difference in flow rate. We will try to improve these Figures and their captions in our final article.

Page 1151, lines 10-21: We don't conduct the Webb-correction on the whole data, because we have not measured the latent heat fluxes with the same set-up. Seeing that latent heat fluxes derived from analyzers with another sampling set-up may not be appropriate. We could only give an estimation of Webb-correction for each 30-min flux value derived by means of an open path CO_2 system.

Page 1152, lines 17-23: See one point above

Page 1153, lines 5-6: We will make a better distinction between sampling frequency and response time. Besides, we will include both aspects in the abstract and in the conclusion.

Table 2: V$\sim$80 mV and V$\sim$180mV mean the detector signals in field 1. However, we will obey the suggestion of referee 1 to omit this Table from our article, because these values are specific to this instrument and not relevant for other users. The Table

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content will partly be conveyed within the text. The main point which we will make is that we can operate the laser far enough above threshold to obtain necessary short term precision of 0.3 $\mathrm{ppbHz}^{-1/2}$ and 3 $\mathrm{ppbHz}^{-1/2}$ for $\mathrm{N_2O}$ and CH_4, respectively.

Some general comments

All general comments will be taken into account in our final article.

Minor corrections

All minor corrections will be taken into account in our final article.

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