Author Comment with regard to:

Comparison of two closed-path cavity based spectrometers for measuring air-water CO2 and CH4 fluxes by eddy covariance

by M. Yang et al.

26 October, 2016

Many thanks for the thoughtful *comments and suggestions from Referee B. Butterworth*. Below we present each comment (in *italic*), followed by our reply. All of our replies are incorporated into the revised manuscript where appropriate, unless indicated otherwise.

General comments

In "Comparison of two closed-path cavity based spectrometers for measuring air-water CO2 and CH4 fluxes by eddy covariance" the authors compare the performance two commercially available gas analyzers (Picarro G2311-f and LGR FGGA) used to measuring CH4 and CO2 fluxes. As a research article on the intercomparison of gas measurement instruments this manuscript falls squarely into the scope of AMT. As stated in the article, instrument performance over terrestrial sites does not necessarily apply to ocean studies as the fluxes of CH4 and CO2 over the ocean are much smaller, justifying a study looking at air-sea fluxes. The findings will be of interest to anyone at tempting to make such field measurements, specifically for deciding which instrument is best suited to their needs. Paper provides a thorough description of the Picarro and LGR in action, highlighting certain technical characteristics of which future users should be aware (e.g., lack of line broadening correction applied internally on LGR, positive offset in dry CO2 mixing ratio in LGR, etc.).

The findings are scientifically sound, following the necessary instrumental and processing techniques that have been developed over the past couple decades. The stationary platform avoids need to correct for motion. Intervals were filtered for non-stationarity. Estimates for high frequency flux loss were performed and magnitudes compare well with previous studies. Flux quality is verified by showing the mean cospectra for CO2 and CH4 fluxes against the momentum flux cospectra. Drying the airstream to at least one of the systems (as was done here) was important to rule out spurious CO2/CH4 flux due to water vapor. The paper was thorough, using different methods to estimate performance when possible (e.g., theoretical and empirical estimates for flux detection limits, filter function and ogive estimates for high-frequency loss, etc.).

With regard to the main thrust of the paper (comparing the performance of measuring flux from the two instruments) the results support the conclusions. The interpretations on the effectiveness of the numerical corrections for water vapor are not as strongly supported by the data. The paper should acknowledge the limitations of the dataset for specifically assessing the true impact of water vapor. An in-depth assessment of the effectiveness of the numerical corrections was not required for this paper, and could be the subject of another study. Some other minor issues/comments are discussed below.

We are happy to hear that the referee finds out contribution useful. Our answers below and changes in the manuscript address the referee's specific questions.

Specific Comments

Line 101 – Inner diameter is more useful if anyone wanted to run the numbers (e.g., Reynolds number, expected flow rates, etc.).

Thanks for the suggestion. The inner diameter is ¼''.

Line 107 – It's unfortunate that the flushing time is slower than 10Hz. It would be good to know how much improvement the LGR might see at faster flow rate.

A faster flow rate should reduce the high frequency flux loss (which is already relatively small, i.e. <=10%). It would not significantly improve the instrument noise or flux detection limit, however.

Line 127 – Not clear how you know that the LGR only applied dilution correction (and not line broadening) internally.

This is clear because the C_{CO2_d} value reported by the LGR is exactly equal to C_{CO2} / (1 + a * C_{H2O}), where a = -0.01. The same is true for CH4.

Line 177 – I don't understand how Figure 3 shows that "cross-sensitivities between CO2 and CH4 vs H2O fluxes are well accounted for by Equations 1 and 2." It seems to show only the impact of the corrections, not real information about how water vapor is influencing the flux measurements. The absence of "additional non-linearity" just means that the numerical corrections are generally linear (i.e., coefficients b and d are effectively zero).

We made this statement because the slopes between the fluxes in Figure 3 are essentially equal to the slopes expected between the gas mixing ratios due to the numerical H_2O corrections. In other words, a +1% change in the mean CO2 mixing ratio due to the H2O correction appears to result in a ~ +1% change in CO2 flux, rather than a change of +5% or -10%, for example. This is because the spectral corrections are small (i.e. coefficients b and d are near zero), as the referee suggested.

To determine the impact of water vapor would require having simultaneous flux measurements using identical instruments, with one dried and one undried. In Figure 2 it's clear that the difference between the LGR numerically corrected fluxes and LGR ambient fluxes is small compared to the difference between LGR and Picarro fluxes. Because the difference between the instruments is greater than the numerical correction for H2O, it is difficult to assess the true impact of water vapor on the measurements. Presumably the impact of water vapor on the measured fluxes is not large due to the attenuation of the water vapor fluctuations in the long tube line (as stated in lines 147, 211) – the larger lag time in H2O should cause any real correlation between fluctuations in H2O and CO2/CH4 to become uncorrelated. But I would be wary

of concluding that the numerical corrections are functioning properly when there is no actual way to verify with the dataset.
Line 198 – Same general comment as above – no clear proof that water vapor is not a factor.

Thanks for the comment. We agree that concurrent measurements using two identical instruments, one dried and one undried, would provide more accurate estimates of the impacts of the numerical H2O correction. In our case the impact of the H2O correction is masked by (i) instrumental differences between the Picarro and LGR; and (ii) severe attenuation of the H2O flux in the long inlet tubing. We have toned down our conclusions on the validity of this correction in the manuscript where appropriate.

Line 218 – Interesting that the scatter in hourly CO2 flux from LGR was only 50% higher than Picarro, while Fig. 7 shows order of magnitude greater variance for LGR compared to Picarro. Any idea why this is? Is this just due to averaging?

As described in Section 3.2, scatter (and also the flux detection limit) in the hour flux depends on both high frequency instrumental noise and ambient variance of the mixing ratio. When the ambient variance is large, the impact of instrument noise on the measurement scatter becomes relatively small. Conversely, over the open ocean when the ambient variance is often smaller than at this coastal site, we expect the impact of instrument noise to be greater.

Line 286 – While not large, tubing can cause some high frequency attenuation of CO2 fluxes (Goulden et al. 1997, Ibrom et al. 2007). With an 18m tube it may not be insignificant. Of course, for the comparison of the two instruments it doesn't really matter, since both will be measuring the same air.

Thanks for the comment. Estimates of flux attenuation by spectral similarity represent the combined effect of the tubing (and Nafion drier/filter, if used) and instrument response time. We agree that there will be some finite attenuation caused by the tubing itself. Given the fully turbulent flow, the attenuation should be rather small for non-sticky gases (Lenschow and Raupach, JGR 1991). Earlier works (e.g. Goulden et al 1997) that employed slower-responding CO2 sensors observed greater flux attenuation, which were probably not primarily caused by the long tube. Ibrom et al. 2007 observed a ~4% high frequency attenuation for CO2 with a 50 m inlet tube, much longer than ours.

Line 289 – It's not clear to me why reducing the flow rate will show response time. Reducing the flow of the calibration gas should not change the mixing ratio of the gas in the chamber. How does this work?

Sorry for not making this clearer. In our setup, the concentrated calibration gas was 'tee-ed' into the flow that went into the instrument. Thus changing the flow of the calibration gas changed the final mixing ratios seen by the instrument.

Line 305 – Does the Picarro (undried) show the same high frequency loss in the

cospectra as Picarro (dried)? If so, the high frequency loss may be attributable to the long tube line, not just the nafion.

Unfortunately we only have <1 d of the Picarro (undried) flux measurements before the laser control in the Picarro failed. This precludes an accurate estimate of the high frequency flux loss in the (undried) Picarro cospectra. However, the volume of the Nafion dryer (PD-200T-24M) is large and previous use of this dryer in DMS flux measurements (see Blomquist et al, AMT, 2010) showed that the dryer caused much more flux attenuation than the inlet tube, consistent with our Fig. 8.

Based on the shorter lag time here than for the maximum covariance lag used in the CO2/CH4 calculations it appears that the time constants were found using a short bit of tube from the cal gas tank - through the nafion - then through the gas analyzer? Future measurement campaigns from this site would benefit from inlet testing (timed release of calibration gas in front of the inlet tube). Time constants for the whole system could be obtained, and used to estimate an overall high frequency flux loss. Also lag times could be measured directly, and be used to verify the maximum covariance lags.

Thanks for the suggestion. Indeed in our test the CO2/CH4 gas standard was introduced inside of the observatory hut, rather than at the tip of the inlet on the mast. We agree with the referee that adding the gas standard from the tip of the inlet would provide a more complete picture of the flux losses and a more accurate determination of the lag time. We plan to implement this in the future.

Technical Corrections
Line 55 – is rigorously and quantitatively confirmed was rigorously and quantitatively confirmed

Suggestion accepted.

Line 108 – "LPM (at atmospheric pressure)" same as "SLPM", which was the notation used in the previous paragraph. Why not stay consistent?

SLPM indicates mass flow measured by a mass flow meter. LPM is an approximate volumetric flow inferred from the instrumental manuals.

Line 187 – Staionarity Stationarity

Corrected.

Line 218 – In this one sentence you report CH4 first, then CO2. Every other time it's CO2 followed by CH4 (even the next sentence). I would keep it consistent.

Corrected.

Line 234 – Switched from mmol m-2 d-1 to mmole m-2 d-1 for this section. Keep

consistent.

Corrected.
Line 273 – (Kort et al. (2012) (Kort et al. 2012)

Corrected.

Line 351 – Unnecessary ")" at end of CD10N equation.

Corrected.

Line 358 – Water vapor correction based on Schotanus et al. (1983)?

Yes. Reference added in manuscript.

References
Schotanus, P., F. T. M. Nieuwstadt, and H. A. R. de Bruin, 1983: Temperature Measurement with a Sonic Anemometer and its Application to Heat and Moisture Fluxes. Boundary- Layer Meteorol., 26, 81–93.
Goulden, M.L., B.C. Daube, S.-M. Fan, D.J. Sutton, A. Bazzaz, J.W. Munger, S.C. Wofsy, 1997: Physiological responses of a Black Spruce forest to weather. J. Geophys. Res., 102, 28,987–128,996.
Ibrom, A., E. Dellwik, H. Flyvbjerg, N. O. Jensen, and K. Pilegaard, 2007: Strong low-pass filtering effects on water vapour flux measurements with closed-path eddy correlation systems. Agric. For. Meteorol., 147, 140–156.