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Atmospheric Measurement Techniques Discussions

Interactive comment on "Eddy covariance carbonyl sulphide flux measurements with a quantum cascade laser absorption spectrometer" *by* Katharina Gerdel et al.

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Reply to Anonymous Reviewer #1: We thank reviewer #1 for his/her critical comments to which we reply below in a point-by-point fashion.

Reviewer comment: Gerdel et al. present a careful consideration of eddy covariance data processing in the specific case of carbonyl sulfide (OCS) data from a popular, commercially available instrument. It is important to pay attention to this kind of methodological detail, and I respect the authors' work here. However, as to the manuscript itself, I have to say that I do not think it presents substantial new concepts, ideas, methods, or data, and therefore I cannot recommend it for publication in AMT. Please do not take the following explanation as demeaning the authors or their work;

that is not my intent. I just need to be clear about why I would reject this manuscript.

Author reply: Not surprisingly we disagree with the reviewer regarding the novelty of our manuscript. Eddy covariance COS flux measurements have become possible since a few years (as of this writing we are aware of a total of 4 published studies) – so far these studies have provided minimal methodological detail on the eddy covariance data processing, making it very hard to reproduce the work. While QCLAS instruments have been used previously for eddy covariance flux measurements of several scalars, it cannot be assumed a priori that experience gained, e.g. for N2O and CH4, holds for COS as well, due to differences in e.g. line strength (as mentioned by this reviewer below). At a recent COS workshop in Finland in September 2016, the lack of clear guidelines for making COS flux measurements and processing the resulting data was identified as a key gap for progress on understanding ecosystem-scale COS exchange. We thus believe there is the need for a study which carefully analyzes and compares the various possible post-processing options associated with eddy covariance COS flux measurements obtained by the Aerodyne QCLAS, which is the only instrument suitable for eddy covariance so far, and thus provides future studies with a reference – this is what motivated this study.

Reviewer comment: The manuscript aims to assess whether the instrument in question can make "defensible EC COS flux measurements", but what does "defensible" mean? The authors do not report the accuracy of the EC OCS fluxes, and so I came away from the manuscript with no more or less confidence in them than I had before. The validation (by comparison to another type of instrument) is restricted to CO2 and H2O measurements. Instead of considering EC OCS accuracy, the authors focus on random noise, and the real question that the manuscript addresses is: can the noise in EC OCS measurements with this instrument be reduced via high- and low-pass filtering? The answer is of course yes: filtering out noise makes data less noisy. If the noise filtering techniques were new and innovative, their performance might be worth reporting, but as the manuscript itself says, the techniques are common.

Author reply: Accuracy is typically defined as the deviation of a measurement compared to some established standard or reference. For example, most of the COS community uses a gas standard derived from NOAA to reference COS concentration measurements. For eddy covariance flux measurements (of a scalar or vector quantity), such a standard or reference does not exist, and it is in fact difficult to imagine how such a standard would look like (this would require a reference surface with a known source/sink strength). As a consequence, the concept of accuracy does not apply to eddy covariance flux measurements. In contrast, it is common to quantify and report the systematic and random uncertainty (Moncrieff et al., 1996; GCB 2, 231-240), as for example in Sturm et al. (2012; AFM 152, 73-82). In the present manuscript we do not repeat previous analyses of systematic uncertainty due to e.g. the choice of the coordinate rotation, but instead focus on two sources of systematic uncertainty specific to the use of the QCLAS that is instrument drift, and the consequences that different approaches of high-pass filtering have, and high-frequency effects. The term "defensible" was meant to convey that we are not able to compare to an absolute standard, but instead have to quantify systematic uncertainty by exploring different processing options. We realize that this may have been not entirely clear and will explain in the revised manuscript what we mean by defensible through introducing the concept of systematic and random uncertainty following Moncrieff et al. (1996).

Reviewer comment: The closest the authors come to the issue of bias (and therefore, in my mind, defensibility) is when they imply briefly in Section 3.3 that the high-frequency noise in the OCS mixing ratio tended to be correlated with the vertical wind velocity signal such that the cospectra were biased high. But they do not offer any evidence for this surprising claim, or any ideas for how such a thing might occur. The figures do not illustrate it.

Author reply: Based on a comment by reviewer #2 we have completed a comprehensive (co)spectral analysis; in contrast to what we had observed on single half-hourly (co-)spectra, it turns out that averaging (co-)spectra by bins of stability and wind speed,

removes the observed behavior, suggesting it to be random noise. We will revise the corresponding text and present new figures showing the results of the new (co-)spectral analysis, as shown in the reply to reviewer #2 (Fig. R2_1).

Reviewer comment: The characterization of the noise in the OCS mixing ratio reported by the instrument (i.e. the Allan plots in Fig 2) does not appear to be new either; I believe the manufacturer itself has done this kind of analysis and freely shares it. That the characterization was done in the field here might add some novelty, but the authors do not say whether the noise was different in the field than in the manufacturer's labs.

Author reply: We thank the reviewer for this suggestion and we will certainly extend the paper by adding information about possible differences between the Allan variance plots reported by the manufacturer and our field findings.

Reviewer comment: This work seems to belong in the methods section of an article that actually makes use of the calculated EC OCS fluxes. In that case, details of how the data processing was done would be important to enable others to reproduce the work. I hope the authors have such a manuscript in the pipeline; I would look forward to reading it.

Author reply: What the reviewer suggests is the approach that has been taken in the few available COS flux papers so far, resulting in exactly the opposite effect – it is close to impossible to reproduce the published work based on these (necessarily within the frame of a paper not having a methodological focus) short descriptions. In addition, within a paper focused on "the science of COS fluxes", it would be impossible to present the details on different processing options and their consequences for QA/QC as present it here. We thus believe that there is a need for and value in such a detailed study in order to pave the way for future work focusing on the science. We though realize that we have missed out to demonstrate the consequences of the various processing options we have explored for the actual application of COS as a tracer for canopy photosynthesis and stomatal conductance and will add corresponding material

to the revised manuscript, as shown in Figure R1_1 below, which demonstrates the relationship between the ecosystem relative uptake rate (ERU – the ratio between the CO2 to COS deposition velocity) as a function of incident photosynthetically active radiation. The figure demonstrates that the ERU deviates from a constant value at low PAR values, when the uptake of CO2 decreases faster than COS due to biochemical limitations of photosynthesis. Important within the context of the manuscript is that the three high-pass filtering options all capture the same response.

Reviewer comment: I was a little concerned about how all laser spectrometers (for CO2 isotopes, N2O, CH4, OCS) were lumped together at times (e.g. page 8, lines 15-17), as if the present analysis ought to apply to all of them – but not, say, to an IRGA. What matters here is the noise and drift in the mixing ratio measurement, not whether the infrared light in the instrument came from a heated filament or a laser. The noise and drift considerations for Patrick Sturm's CO2 isotope QCLAS were very different than those for the present OCS QCLAS, which is measuring a comparatively tiny spectral line.

Author reply: Actually, as we argue above, it is exactly NOT our intention to lump all laser spectrometers together, rather this paper starts out to investigate whether the instrument used and the required processing steps are in any way different from what is known for other laser spectrometers. We will clarify this in the revised manuscript.

Reviewer comment: Regarding the high-pass filtering: an alternate approach is to correct the drift in the OCS mixing ratio before beginning EC calculations. The OCS mixing ratio drift results from slow changes in the spectral baseline (i.e. the zero offset), which is reset periodically by the QCLAS's auto-background feature but changes in between those resets. By comparing measurements of the same gas (a standard tank or even the atmosphere) just before and just after an auto-background reset, one can determine how much the OCS zero level had drifted since the previous reset and make a linear correction (though the drift might not always be so linear). When it comes to the EC fluxes, this method is probably similar in effect to using linear detrending but

ought to be better because real trends in the OCS mixing ratio would not be filtered out.

Author reply: We thank the reviewer for this useful suggestion which we have implemented thanks to the conducted half-hourly background measurements. The results of this approach will be compared against the high-pass filtering approaches already tested.

Reviewer comment: Regarding the approach of Wienhold et al. (1995) to quantify the SNR, it always seemed fundamentally flawed to me. If you look at a plot of covariance vs lag time, you typically see oscillatory patterns because the eddies are quasi-periodic structures. So the variability in the covariance as you scan the lag time is not merely noise: much of it results from the quasi-periodic nature of the signal. You should be able to test this by comparing the noise estimate obtained this way when the signal (i.e. the real eddy flux) is large (e.g. at midday) to when it is near zero (e.g. at night for H2O and OCS). If the noise seems to decrease as the signal decreases, then the method may be flawed in the way I suggest.

Author reply: The reviewer is correct in pointing out that cross-correlation plots feature repeating patterns due to the quasi-periodic structure of the main transporting eddies. The term SNR is thus not really appropriate and we rather should call it a flux detection limit test (as originally done by Wienhold et al. 1995). Based on the reviewer comment we have adopted a second flux detection limit test not based on a cross-correlation analysis – the approach by Pihlatie et al. (2005; BG 2, 377-387) and compare it against an improved version of the Wienhold approach put forward by Rannik et al. (2016; AMT 9, 5163-5181) as suggested by reviewer #2.

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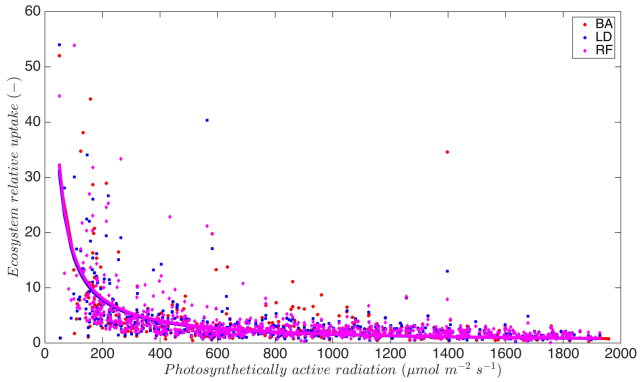

Figure R1_1 Ecosystem relative uptake rate as a function of incident photosynthetically active radiation for three high-pass filtering options. Lines represent non-linear fits to the data (symbols) using the equation y = a / (b x).

Fig. 1.