Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2019-278-AC2, 2020

Atmospheric Measurement Techniques Discussions

Interactive comment on "First eddy covariance flux measurements of gaseous elemental mercury (Hg^0) over a grassland" *by* Stefan Osterwalder et al.

Stefan Osterwalder et al.

stefan.osterwalder@univ-grenoble-alpes.fr

Received and published: 24 January 2020

Comment 1 from the Referee: This manuscript is very interesting and presented a very promising methodology significantly improved the Hg(0) flux measurement. Accurate measure Hg(0) flux has been a challenging issue over decades due to the limitations of available methods. As I read this paper, I had an impression that paper proposed a very promising trail work to significantly advance Hg(0) flux measurement. Overall, I support the publication of this manuscript on journal AMT. Line 14 to 15. The statistical estimate... (50% cut-off). Move this sentence to the present line 18. The field campaign based detection limit should be described in the context of the campaign

Author's response: We moved the sentence to line 18 as suggested.

Comment 2: Line 18, 24 in the abstract and throughout the manuscript (section 3.3), replace "re-emission" with "emission" in general. "Emission" represented the measured results better.

Response: We agree with the Referee and replaced "re-emission" with the more general term "emission".

Comment 3: Line 176-181. Three auto-calibration strategies were performed through the course of field campaign. It seems that frequent auto calibration relaxed the baseline drifting (Fig.2d), and the authors do used same data processing steps to detrend the time-series data. So which measurement routine is better for achieving high quality data? Meanwhile, increased calibration frequency resulted a significant loss of online data, linear interpolation is mentioned to fill the gap before flux calculation: (1) will this increase the uncertainty compared to a less frequency of calibration, in terms of different proportion of high-frequency data gap filling? (2) the methodology of gap filling of high frequency data was not easy to understand in the present manuscript.

Response: The Referee addresses an important issue: how to find an optimum between long continuous measurements with infrequent quality control of the drift of the instrument away from the calibration target vs. frequent calibration at the expense of losses in the time series due to gaps. The potential effects this has on flux estimations is best explained using Fig. 4 in the manuscript, which shows a Fourier-transformed example spectrum of the Hg(0) measurements (Fig. 4a) and an example cospectrum of the Hg(0) eddy covariance flux (the so-called cospectrum). Typically, the characteristic time of turbulent turnover in the daytime atmosphere is on the order of one hour, hence the standard is to use 1-hour continuous time series for such analysis. If we calibrate every 24 hours, the Nyquist cut-off frequence at the low frequencies is 2/24 hours = 2/86400 s = 0.000023 Hz, thus if our instrument runs stably over such a period, we would expect no effects of calibration on the fluxes (except for the hours when the calibration took place). Contrastingly, if we perform a calibration every 4 minutes we have a low-frequency cut-off at 2/4 minutes = 2/240 s = 0.0083 Hz. This frequency in Figure 4 falls between the tick of 0.001 and 0.01 Hz and eliminates all information on the left hand side of a vertical line that we could draw at 0.0083 Hz. The information (spectral or cospectral densities) at these low frequencies are always associated with a relatively high uncertainty, independent of the measurement, since the statistical coverage becomes poorer the lower the frequency of cyclic behavior of turbulent eddies. For example: a cyclical pattern with a return interval of once per 4 minutes is only covered with 15 “data points” in a 60-minute averaging period. Contrastingly, some turbulent mixing happening at 0.1 Hz (cyclical behavior assumed) is covered by 360 data points and thus statistical uncertainty is much smaller at high frequency than low frequency in eddy covariance flux measurements.

Now, to address question (1) these considerations mean that all the data points to the left of 0.0083 Hz do not exist because they were not measured, and the best we could do is to assume ideal behaviour as shown by the blue idealized cospectrum in Fig. 4b. But note the logarithmical y-axis: cospectral densities at 0.0083 Hz and lower frequencies are already more than one order of magnitude smaller than in the energy containing range (with the maximum of the blue line at ca. 0.23 Hz), and thus the answer is: yes, this increases uncertainty as in any system where less data are associated with higher uncertainty than more data of the same observational period; but at the same time it can be said that this additional uncertainty is not of great concern since the contribution of the low-frequency parts shown in Fig. 4 are small in comparison to the resolved part of the spectrum and cospectrum.

For scientific reasons it should however also be mentioned, that when testing equipment one tends to focus on ideal cases and golden days. Thus we could say: for ideal cases and golden days calibrating every 4 minutes is not a big problem. But in meteorology most cases are not ideal and do not fall in the category of golden days, and thus, with too frequent calibration we would lose part of that information about (possi-

bly non-ideal) contributions of the low frequencies. Question (2) addresses the issue of gap-filling the calibration periods with a linear interpolation. This obviously was not easy to understand. We must admit that because this is only relevant in our paper to convince the reader that it is not really the relevant aspect, we did not phrase the text carefully enough. We will of course improve that text in our revision.

Here for explanation (this addresses both question 1 and 2) we present the cospectrum of the period shown in Fig. 3b,d,f (with 4-minute calibration) for comparison with an ideal period as shown in Fig. 4b (see Figure E1 in the supplement to this comment). If we calibrate every four minutes but want to calculate hourly fluxes and cospectra, we have to deal with the gaps in one or the other way. A typical approach if a time series is too short (e.g. only 40 minutes in a 60 minute interval), tapering on both sides with the mean of the data is a conventional approach in time series statistics since this has the least effect on the shape of the spectra and cospectra calculated from such data. In our case, the mean of the entire period however would introduce additional unwanted artefacts because variations at lower frequencies are still seen after detrending. In our example in Fig. 3d you see that the junks of continuous data are not perfectly fluctuating around a common mean value. Even after drift removal some variation with repetition intervals of 10 to 20 minutes are seen. Thus, we decided to make a simple linear interpolation instead of using a constant value to bridge the gaps during calibration passes.

This means that we removed a substantial part of the high-frequency variablity by doing so. In Figure E1 this leads to the artefact that the scaling of the cospectrum goes too far away from the idealized cospectrum (blue dashed line in Figure E1 in the supplement to this comment). Although the qualitative shape is still surprisingly fine, the frequency range with highest relative contribution to the normalized cospectrum is no longer centered around 0.23 Hz (where we expect it based on idealized empirical cospectra published in the literature that were used for the dashed blue line), but shifted towards lower frequencies. As a short summary of our long response: the final test we made with frequent calibration at 4-minute intervals clearly shows that there is no dramatic improvement in doing so with respect to the quality of the fluxes and hence our conclusions suggest to improve the instrument so that standard eddy covariance processing fits the purpose, with less frequent calibration. We will revise the text in Sections 3.2.1 (High-frequency signal analysis) and 3.4 (Suggestions to improve the Eddy Mercury system) accordingly.

Comment 4: Line 439, replace dial flux cycle with diel flux pattern.

Response: Done.

Please also note the supplement to this comment:
https://www.atmos-meas-tech-discuss.net/amt-2019-278/amt-2019-278-AC2-supplement.pdf

---

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2019-278, 2019.