

Answer to referee comment 2 for “Quantifying agricultural N₂O and CH₄ emissions in the Netherlands using an airborne eddy covariance system”

We thank the reviewer 2 for the recommendations for improvement of our manuscript. Below we list our answers to the comments and corresponding revisions in the manuscript. Referee comments are written in normal font, our answers in italic font.

The authors describe high-speed airborne in situ measurements of N₂O and CH₄ using a new commercial spectrometer. These measurements are used to calculate vertical fluxes of N₂O and CH₄ via eddy covariance, and results are described for four flights during the GHGMon campaign in 2023. Results show a persistent level of CH₄ flux consistent with ruminant emissions and increased N₂O flux within a day of a significant rain event, with evidence that N₂O flux increases is related to an increase but not amount of soil moisture. I find this work to be novel, well written, and worthy of publication in Atmospheric Chemistry and Physics. I recommend publication after the authors address a few minor comments:

Section 2: Placing the instrument and campaign description subsections before the details of the eddy covariance analysis may be a better way to organize the paper. I found myself jumping down to the instrument & campaign section and finally just read it totally before going back to the EC section.

The positioning of subsections in the methodology part of our manuscript (Eddy covariance theory vs. instrument and campaign description) was indeed under debate among the participating authors. Since we don't assume Eddy covariance to be very well known within the expected audience, we decided in the end to start with the principles of Eddy covariance. We are aware that this may limit the smoothness of readability somewhat, but this would likely be the same, when discussing e.g. measurement strategies and designated eddy covariance flight patterns (in the campaign section) before introducing the eddy covariance methodology itself.

As a compromise and to improve the reading flow, we expanded the short summary/introduction of the overall methodology Section (Section 2), to give all relevant information on the instrumentation to the reader which is necessary to go through the Eddy covariance Section more smoothly.

Line 135: You denote stationarity, horizontal homogeneity, and well-developed turbulence as core EC assumptions, and I think you demonstrate each of these at different points, but the explicit terminology doesn't come back. I'm not sure the exact place(s) where it would be appropriate, but it would be good somewhere in the manuscript to denote exactly where you are justifying these assumptions (e.g. sect 3.1 for stationarity, see comment below).

Thank you for this hint, we added discussion to revisit the underlying concepts and explicitly state how they are fulfilled in the corresponding Sections.

- You are right, we demonstrate (quasi-) stationarity using the cospectral analysis in conjunction with our method to estimate the cut-off frequency between turbulent and non-turbulent, larger scale motions. We don't find significant low-frequency, large-scale contributions, from e.g. mesoscale motions, which would have led to a break-down of the stationarity assumption.*

- *Horizontal homogeneity is now discussed in more detail in the context of the moving window approach, where only local homogeneity is required within each single window. By comparing fluxes of consecutive windows, possible spatial heterogeneity of the flux field can be determined with this method, instead of leading to a violation of the homogeneity assumption.*
- *We used the convective velocity scale w^* to define a threshold for well-developed turbulence of individual flux legs. By inspecting variance spectra of vertical wind and scalars we find a pronounced inertial subrange spanning a broad range of frequencies following the expected $-5/3$ law, indicating that turbulence is well developed with energy cascading down from the energy containing range to the dissipation range. Under these conditions, the covariance calculation provide a meaningful representation of the flux.*

Specifically, we have included (inter alia) the following paragraphs to

Section 4.1:

“Repeated flux legs over the same ground scene, but flown during different times of the flights, and even at different times of the day (morning flight 21a and afternoon flight 21b), yield similar emission rates. This reproducibility indicates spatial coherence and temporal consistency of the flux signals, hence stationarity. We observe smooth transitions in fluxes of single flux legs between regions of low fluxes and regions of high fluxes and since consecutive flux legs have a large spatial overlap with corresponding similar footprint, local homogeneity within the flux legs can be assumed. Thus, the variability of N_2O fluxes across the target area, from close to zero in some parts of the area up to emissions of around $1 \mu g m^{-2} s^{-1}$ (in the central part of the patterns) demonstrates the ability of our airborne EC setup to detect and hence, spatially resolve small-scale (i.e. 1–10 km) emission hotspots within a relatively homogeneous landscape.”

Section 3.1:

“This marginal shift of dominating eddy scale sizes after applying the high-pass filter indicates that contributions from non-turbulent, large-scale motions are negligible and do not distort stationarity during the time of the flight, which is a core assumption of EC. Nonstationarity would have led to incomplete averages, with overshoots or oscillations.”

“The dominant flux-contributing scales lie within the inertial subrange, which in this case is well pronounced over a broad range of frequencies for all scalars and w , demonstrating well-developed turbulence and hence consistency with EC theory.”

Line 160: Upcoming manuscript that by now may be citable? Or an upcoming experiment? It may suffice to simply state that you used Equation 4 and deeper footprint analysis is deemed outside the scope of this manuscript.

We changed our comments on the footprint calculation as suggested. We are working on more advanced footprint analysis currently, for future comparison of the GHGMon data with emissions inventories at high spatial resolution. But as you point out, those activities are out of the scope of this manuscript, which aims for the introduction of the airborne eddy covariance system and the demonstration of its suitability to study agricultural GHG emissions.

Line 167: This line about the timing is important to include, but was confusing placed here in the middle of a discussion of empty data treatment (made me ask if you were interpolating GPS time synchronization data).

We adapted the corresponding description of clock synchronization for improved clarity. We were not interpolating GPS time stamps, but due to small divergence between GHG and METPOD clock, we had to interpolate single GHG data values a few times per flight.

Line 240: Should “no” be “minimal” or “no significant”? ogives give no further contributions only when they reach the highest/lowest frequency at 1 or 0...so maybe once contributions reach N%?

That is true, our initial statement that “ogives don’t show further contributions” implies a misleading impression of exactness. We changed this accordingly and used the formulation of “no further significant contributions”.

Line 301: What WMO scale is your dataset traceable to, presumably X2006A?

Yes, it is X2006A. We included this information in the manuscript.

Line 310: Have you examined whether there is any artifact from aircraft motion in the measurements? If there is, then preferentially calibrating in turns could bias the dataset.

We did visual inspection of data quality during aircraft maneuvers, including turns, climbs, descents and at higher flight levels with lower ambient pressure. We could not find a dependency (bias) of any of the measured GHG concentrations related to these maneuvers, nor were calibration gas measurements in turns or during climbs and descents of less quality (like enhanced noise (spread)).

Line 311: Have you evaluated whether transferring standard gases affects the in-flight concentrations? Usually when standards are filled, there is some amount of settling time.

We are aware that the transfer of reference gases can lead to a reduced quality of the calibration and has to be done with caution. During GHGMon, the refilling of gases was done thoroughly by flushing the pressure reducers of the involved cylinders and the transfer-pipes at least five times generously with calibration gas. Furthermore, the same large gas cylinder was used for the replenishment of the small cylinders during the whole period, hence no residual gas contamination was possible. In the comparison of the GHG analyzers concentration measurement with the flask samples of JAS (Appendix), we don’t see significant flight to flight discrepancies, which would elucidate a possible contamination of calibration gas due to gas transferring. We have therefore high confidence that our calibration routine does not suffer from the gas transfer.

Line 315: Did you evaluate whether a calibration slope correction is needed, even if you could not perform this in the air (e.g. in the lab, on the ground with other calibration standards).

We acknowledge that a slope correction (with a two-point calibration) leads to a higher accuracy in most cases, but as mentioned, we were not able to apply a two-point calibration during the GHGMon campaign. We indeed made experiments with multiple reference gas bottles of different N₂O concentrations in the laboratory and derived corrections. We found that at least between 300ppb and 400ppb mole fractions of N₂O, a linear correction (slope correction) fits the observations versus reference standard best. Those slope corrections were relatively constant over repeated experiments with a couple of weeks in between. We observed variability of slopes between minimum 1.004 and maximum 1.028 (without systematic trend between minimum and maximum slopes). Changes in the slope are a consequence of changing sensitivity, which indeed can affect eddy covariance fluxes.

A slope correction propagates directly into the flux calculation according to:

$$F_{true} = \overline{w'c'_{true}} = \overline{w'(m \cdot c_{meas} + t)'} = m \overline{w'c_{meas}'} = m F_{meas}$$

With subscript “true” denoting real flux or concentration (with slope correction), “meas” denoting raw flux and concentration (without slope correction), m the slope of the linear fit, and t the intercept. t does not contribute to the covariance term, since it is only a constant added to the concentration. In the case of slope $m=1$, the calibration has no impact on the fluxes ($F_{true} = F_{meas}$). If $m \neq 1$, the relative flux error due to a missing slope correction can directly be inferred from the slope. Our laboratory experiments revealed slopes slightly deviating from 1 (between 1.004 and 1.028), therefore, and under the assumption that the sensitivity of the GHG analyzer would have shown similar behavior during the GHGMon campaign as it did in the laboratory, the associated additional relative flux error would only have been 0.4 – 2.8 %. This assumption is confirmed by the comparison of the GHG analyzer with the flask samples (JAS), where a slope of 1.04 was found, which also only slightly deviates from 1, but additionally includes effects of limited accuracy from JAS (see Appendix B).

Due to the expected small flux error contribution of a missing slope correction compared to other flux error terms, and due to the fact that we could not perform a slope correction during the campaign, hence could not quantify this error term, we decided to not include it in our calculations. Since we acknowledge that a slope correction would slightly improve our flux estimates, and since this information might be of use for similar approaches, we included our statement in the Airborne EC uncertainty analysis Section in Appendix A:

“In the case of N_2O , we furthermore note, that our limitation to a simple offset calibration will introduce an additional flux error, directly proportional to the difference of the slope from one. During repeated laboratory experiments with different reference gases, we found slopes only slightly deviating from one (maximum 1.028), hence the corresponding errors are expected to be very small compared to other flux error terms (<3-4%). Therefore, because of other flux error terms being an upper limit approximation, and because we could not determine this uncertainty contribution quantitatively, we decided to not include it in our uncertainty assessment.”

Line 322: This seems important to evaluate the measurements, there should at a minimum be a reference to this other publication, at least a conference proceeding/presentation.

The water vapor correction is definitely important to consider for the flux calculations. There will be a separate publication on this topic in the near future, but by the time of the final submission of this manuscript, this separate publication is not citable. We inserted instead a reference to a conference presentation, where we addressed the water vapor correction.

Line 334: “measured with a time resolution” -> “reported at”, 100 Hz is not the sampling rate of any of the METPOD suite, just the sampling rate.

This was a mistake by the authors. 100 Hz is just the rate at which meteorological and wind measurements are reported. This was corrected in the manuscript accordingly, for example:

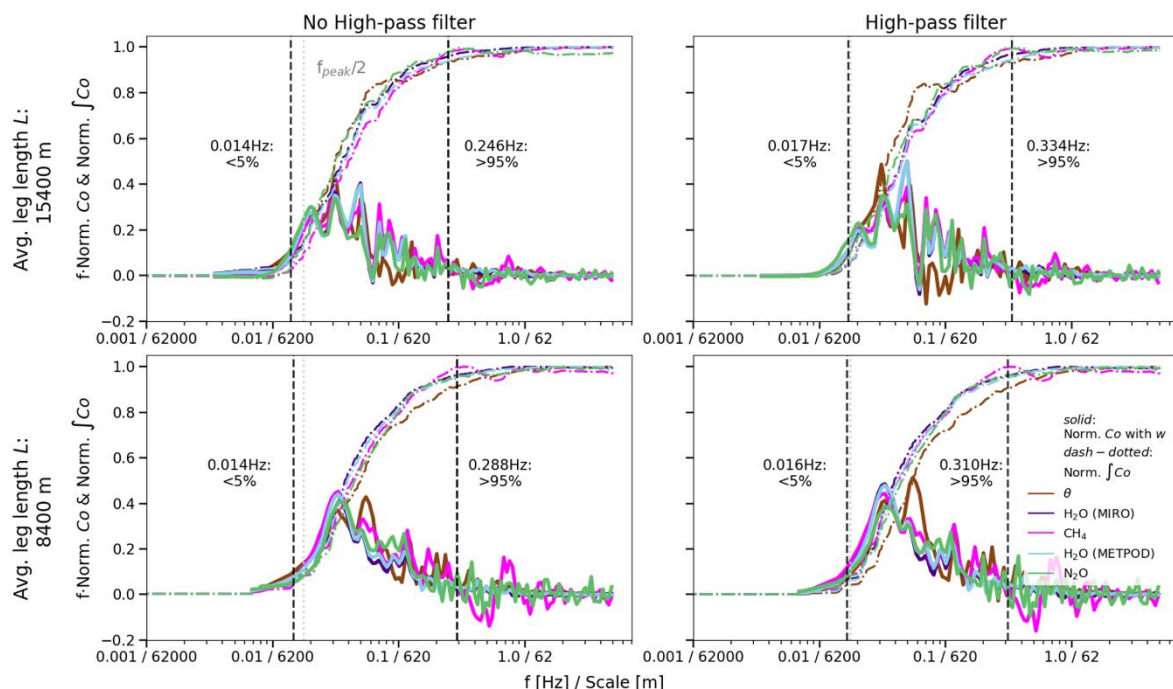
“The vertical wind component is reported with 100 Hz and an uncertainty below 0.2 m s^{-1} . Spectral analysis revealed, that up to 10 Hz wind measurements, no noise contributions or system-specific resonance frequencies affect the data quality.”

Line 407: If I understand correctly, this comparison is a pseudo-test of stationarity, but I don’t see you mention that directly.

Yes, it is, we adapted the paragraph accordingly. See also answer to comment in Line 135 above.

Figure 4: It would be nicer to the eye for the x scales to be the same for each panel.

Thank you for pointing this out. We changed the x axis tick arrangement to allow a direct comparison of the different panels.



Line 421: 100 Hz is the data rate but not the time response of METPOD (see comment above). I couldn't find in the reference the exact time response for the METPOD TAT measurement (I only saw a power spectrum for the wind components), but I've never seen a PT100 TAT sensor with a faster response than 7-8 Hz. Acknowledged that this does not affect the point you are making with the converging ogives by 0.3 Hz.

We apologize for the same erroneous phrasing similar to the comment to Line 334. We removed the statement of a 100 Hz measurement frequency. Instead, we use the phrasing "fast" and refer to Mallaun et al. (2015), where it is shown, that the wind measurement (including vertical wind speed) is at least reliable up to undisturbed measurement frequencies of 10 Hz. We have proven in our work, that this is clearly fast enough for the computation of the eddy covariance fluxes. Regarding the TAT measurement, we agree with the referee that 100 Hz is not the time response of the sensor, but in analogy to the wind measurements, we can state that based on our spectral analysis, the TAT measurement has no white noise contribution down to the Nyquist frequency of 5 Hz, hence at least has an effective measurement frequency of 5 Hz - fast enough for the GHGMon fluxes.

Line 446: Can you tell if the GHG sensor or wind sensor are the limiting noise source, or if they both contribute similarly?

Thanks for this question. We investigated the contributions to the instrumental noise error upon your question, by separately computing the terms including the precision of the GHG analyzer and the vertical wind measurement, since there is no interplay between those variables in the instrumental noise error term $\sigma_{instr.noise}$ (Appendix Equation A3). We found that for N_2O , the GHG analyzers contribution to the instrumental noise error is 85% compared to 15% of the vertical wind measurement (average over the flights presented in our work). For CH_4 , the GHG analyzers contribution is 69% on average, and the one of the vertical wind measurements 31%. We

included this information in the uncertainty discussion (Section 3.2), since it might be of interest when thinking about technical optimization of our or comparable systems.

“We notice that instrumental noise contributions from the GHG analyzer are approximately five to six times larger than those from the vertical wind measurements in the case of N₂O, and approximately two to three times larger for the CH₄ flux error (averaged across all four flights).”

Figure 7 caption: forenoon -> morning

The word “forenoon” was replaced with “morning” in all parts of the manuscript.

Line 530: Figur -> Figure

Corrected!

References

Mallaun, C., Giez, A., and Baumann, R.: Calibration of 3-D wind measurements on a single-engine research aircraft, Atmospheric Measurement Techniques, 8, 3177–3196, <https://doi.org/10.5194/amt-8-3177-2015>, 2015.