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Interactive comment on "Measurements and quality control of ammonia eddy covariance fluxes: A new strategy for high frequency attenuation correction" *by* Alexander Moravek et al.

Anonymous Referee #1

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In the present study, the authors used an eddy covariance system to measure ammonia fluxes during 5 months over a corn crop field. The main objective of the manuscript is to propose and demonstrate a new method for the quantification and correction of the high frequency attenuation (HFA) of the flux measurements. The method is based on subjecting the NH3 analyzer (incl. inlet system for EC) periodically to a step change in concentration by switching to zero air. From the temporal response in the detected concentration signal (described by the sum of two exponential decay functions) the spectral transfer function for turbulent fluctuations was derived. This was then applied

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[Discussion paper]

to the cospectrum of the undamped sensible heat flux to determine the integral atten-uation factor (relative flux loss) for the measured NH3 flux. The new proposed HFA determination method is compared to the ogive method that was previously proposed in the literature. The ogive method (like similar cospectral method) compares the shape of the damped NH3 cospectrum to the undamped sensible heat cospectrum and de-rives the integral damping factor by assuming full spectral similarity in the lower half of the turbulent frequency range. Beside the methodological aspects of the proposed HFA determination method, the manuscript also presents mean diurnal cycles of the corrected NH3 fluxes exhibiting mainly emissions in spring/summer and mainly depo-sition in autumn.

I consider this study as a very interesting and important contribution to the problem of HFA in eddy covariance fluxes of reactive and 'sticky' (highly water soluble) trace gases. The manuscript is written mostly in a clear and concise way and discusses many important aspects of the problem and the proposed method. Yet, there are some issues that need to be addressed before the paper can be published. They are listed in the following.

MAJOR COMMENTS

[1] page 6, line 34: The details of the zero air inlet design are quite crucial for the success of the proposed HFA method. However, unlike stated in this sentence, no corresponding details are visible in Fig. 1. The authors should give a close-up view of the gas inlet and the attached inlet for the zero gas. In addition it needs to be specified, how the zero gas unit (heating catalyst) was operated during periods of normal EC measurements (outside the zeroing periods).

[2] page 11, Eq. 8: The presented correction of the cospectra using sqrt[T(f)] only accounts for the damping of the fluctuation amplitudes. However a first order filter as characterised in Eqs. 5 and 6 also leads to a phase shift in the damped frequencies (e.g. Horst, 1997; Massman and Ibrom, 2008). Because the constant phase angle

shift corresponds to different lag times for different frequencies, this effect is only partly compensated by the empirical lag time correction. Thus especially for HFA over a large frequency range, as observed here, the unaccounted phase shift can be important. This effect should be addressed in the manuscript.

[3] Page 19, line 5-6: Concerning the results of the ogive method, it is stated here that the results from least-squares regression (corresponding to an arithmetic averaging) strongly deviated from the median. This is not surprising because the ogive results represent ratios of (very noisy) half-hourly cospectra. Such data usually have a non-Gaussian (non-symmetric) distribution. Therefore the application of a simple least-squares regression in Fig. 6 is not adequate. Instead, the calculation of binned medians (as e.g. used in Fig. 8) or a median regression would be much more suitable (eventually with separation of stable and unstable cases). In that way, a dependence on windspeed possibly could be detected. Also for the analysis of the temporal behavior (before/after inlet cleaning) a running median filter should be applied to the ogive data. Generally: if two approaches for HFA are inter-compared and rated, they should both be evaluated in an adequate and careful way. Thus the evaluation of the ogive method needs to be improved as suggested.

MINOR COMMENTS

[4] page 8, line 3-6: With respect to its potential influence on the HFA, it would be of interest to show (or quantitatively describe) the temporal variations of observed lag times.

[5] page 9, line 21: How stable was the 'background' signal (y0) of the QCL during the study? Its variability would be an indication for the systematic uncertainty and the total accuracy of the QCL measurements.

[6] page 10, Eq. 7: The representation of the two different response times (exp. functions) by the combined transfer function in Eq. 7 yields an adequate result in practice for the present study. However, the additive combination of individual transfer functions

is conceptually problematic. It e.g. implies that the fast tau1 filter only acts on a fraction (1-D) of the concentration fluctuations. However, if this filter (as suggested by the authors) can be attributed to the air mixing in the analyser detection cell, it physically acts on all fluctuations. Therefore, a multiplicative combination of the transfer functions would be conceptually more adequate in my view, and it would be compatible to the classical transfer function method (Moore, 1986; Moncrieff et al., 1997). At least the relation and differences between additive vs. multiplicative combination of transfer functions should be discussed.

[7] Although the focus of this study is the HFA method, it would be suitable to give information about the possible sources of NH3 in May and August (Fig. 3) as a difference to other months showing deposition. Therefore, the timing, type and amount of fertilizer applications to the corn field (before or during measurement period) should be reported.

[8] In order to decide whether the estimated flux detection limit (closely related to the flux uncertainty) is plausible, some exemplary time series with measured half-hourly fluxes should be presented. These would be more informative in the present context than the statistical diurnal cycles in Fig. 3.

[9] page 11, line 24: This precision only represents the precision at zero concentration while the precision at higher ambient concentrations could be larger. This should be specified.

[10] A separate quantification of the HFA factor with the tau1 transfer function alone would be of interest in order to partition the total damping into 'classical' damping effects applicable to all trace gases and wall sorption effects only applicable to NH3.

[11] page 13, lines 6-7: Were the distinct temporal variations in D found only after fixing tau1 and tau2 to overall constant values, or were they equally found with variable tau1 and tau2?

[12] page 13, lines 17-18: In my view the dependence of the D value from the NH3 concentration in Figure 8 is not so clear as stated in this sentence. To get a better impression from Fig. 8, it would be useful to indicate the number of data (n) in each bin.

[13] page 13, line 26: "time constants were fairly constant over time" is a strange statement. I suggest to replace "time constants" by "response times".

[14] Figure 7: What was the reason for the decrease and subsequent increase of D at the end of August?

Additional References:

Horst T.W., 1997. A simple formula for attenuation of eddy fluxes measured with first-order-response scalar sensors. Boundary-Layer Meteorology, 82(2), 219-233.

Moncrieff, J. B., Massheder, J. M., deBruin, H., Elbers, J., Friborg, T., Heusinkveld, B., Kabat, P., Scott, S., Soegaard, H., and Verhoef, A.: A system to measure surface fluxes of momentum, sensible heat, water vapour and carbon dioxide, Journal of Hydrology, 188, 589–611, https://doi.org/10.1016/S0022-1694(96)03194-0, 1997.

Moore, C. J.: Frequency response corrections for eddy correlation systems, Boundary-Layer Meteorology, 37, 17–35, https://doi.org/10.1007/BF00122754, 1986.