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Interactive comment on "Correcting high-frequency losses of reactive nitrogen flux measurements" *by* Pascal Wintjen et al.

Anonymous Referee #2

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GENERAL COMMENTS

The manuscript "Correcting high-frequency losses of reactive nitrogen flux measurements" by Wintjen et al. represents an important study which investigates the performance and applicability of different methods to correct for the high frequency attenuation of reactive nitrogen eddy covariance fluxes. This topic is very relevant since observed losses of reactive nitrogen fluxes are substantial and parametrizations that correct them are difficult to obtain due to the complexity of chemical and physical interactions of the reactive nitrogen species within the inlet system and instrumental setup.

Wintjen et al. compare five different methods that are commonly used for the flux correction of typically less reactive gases such as H_2O and CO_2 and apply them to the reactive nitrogen flux measurements at a peatland and a forest site. Since they

performed flux measurements over several years, the authors have an extraordinary dataset to conduct this analysis covering a wide range of environmental conditions. Although they do not find significant dependencies of the flux loss with environmental conditions, they are able to propose that using an empirical co-spectral approach is most the most suited of the five methods they investigated.

While the scientific analysis and conclusions are mostly sound (see for more details the specific comment below), I found the manuscript very difficult to read and it should be improved from a reader's perspective. At many points, sentences lack clear links to each other, making it difficult to follow the line of arguments. In some sections, this creates the first impression that they are placed quite arbitrarily, which is enhanced by that fact that some statements are restated at multiple times within one paragraph. Furthermore, often it is not clear what is referred to (at least it can be ambiguous), interrupting the reading flow. To improve the readability, I strongly recommend to better link individual sentences, to remove redundant statements and to make referrals more precise. I believe this will significantly improve the quality of the manuscript, making for a stronger case for the suitability of each flux correction method.

In the following, scientific comments are listed that should be addressed. Furthermore, I provided additional comments where the manuscripts needs further clarifications. These do not need to be answered individually but should be considered in the revised version.

SPECIFIC COMMENTS

[1] L. 47-50: Since they used a different approach to correct NH3 fluxes, I suggest to include the recent study by Moravek et al. (2019) here.

[2] L. 173-174, Figure 1 + Figure2: The authors use averaged co/power-spectra of several hours measured on one specific day for each site. How representative are the chosen spectra for (daytime) conditions during the entire measurement period? While it is clear that the shown spectra are exemplary, it would be beneficial to state on how

many days spectra with the same described features were observed. Furthermore, is there a reason why different days were chosen for the co-spectra in Figure 1 and 2? If so, these should be highlighted.

[3] L. 182-193: How does this approach compare to the approach by Aubinet et al. (2000), which uses a normalization factor instead of normalizing by the total covariances? I find it important to highlight the methodological differences here as the method by Aubinet et al. (2000) is also referred to widely in literature (Foken et al., 2012). Furthermore, it should be shown here how Eq. (2) was derived from Eq (1). Co(. . .) and TF_exp are functions of the frequency, but then solving for alpha is not straight forward, unless I miss something here. In general, the description of how the fits to obtain Tau_exp were performed can be improved. For example it should be mentioned what kind of least-square fit was performed, linear, non-linear?

[4] L. 200-207: To support the descriptive text, the Ogive equation should be included here. Also, the "optimization factor" (L. 207) has not been defined yet.

[5] L. 223-224: It should be explained here briefly why this parameterization of alpha is used, which uses the horizontal wind speed. While it provides the opportunity to apply the correction to a large dataset (once fc is known), the methodology is different from the other approaches used here, that determine alpha more directly. The mean horizontal wind speed (u) should be defined in the text.

[6] L. 241: It should be stated that the slopes in the inertial subrange are meant here. In addition, shouldn't a weaker slope (-0.62 and -0.63 compared to -2/3) result an increased flux contribution in the high frequency range, which is the opposite as stated here? Was the entire inertial subrange used to derive the slopes? This is probably problematic since both positive and negative slopes are observed in the same power spectra.

[7] L. 254-255: Was the precipitation filter for the Li-7500 data only used for the evaluation of the presented slopes? If so, shouldn't it have been applied for filtering the power

spectra and cospectra as well?

[8] L. 370-373: A stronger white noise of Ps(CO2) at BOG than FOR is not visible to me in the power spectra presented in Figure 2. Which frequency range the authors referring to? May the slight increase in the very high frequencies be due to the aliasing effect?

[9] L. 350-410: The power spectra used for the IPS method do not follow the expected shape and the authors relate the increase of power densities at high frequency to white noise. This is a good demonstration of the shortcomings of the IPS method, for conditions where instrumental noise impacts over a certain frequency range. As the instrumental noise is uncorrelated with the vertical wind speed, it is not detected in the co-spectra. Still, it has to be discussed how the instrumental noise impacts the detection of small mixing ratio fluctuations that relate to the trace gas flux, which then would also impact the Co-spectra. In my view this is not clearly discussed in the manuscript. While the authors state that the instrument was probably unable to resolve small concentration differences, the power spectra at the BOG site show a steep decline in the inertial subrange that is similar to the one from the temperature time series. This would suggest that the instrument was capable of capturing the concentration differences in the high frequency range. Also, it would be useful to include in the discussion under which conditions the IPS method can be used.

[10] L. 397-398: The authors state that under conditions with "less variability in concentrations and depositions fluxes" the IPS method fails. Shouldn't it be just under low flux conditions (regardless whether deposition or emission fluxes prevail)?

[11] L. 434-437: The authors list here parameters that could affect the time response. However they do not discuss which component of the TRANC-CLD setup they expect to have the largest impact on the time response. Since all Nr compounds are converted to NO by the TRANC, interactions with the tubing wall may be less important than for example interactions of NH3 at the inlet, which is much "stickier" than NO. Therefore,

adding a short discussion on the expected high frequency attenuation processes would help to better understand the observed/non-observed dependencies of alpha with environmental and instrumental parameters. I think this is important to discuss since it would show whether the system's time response was more similar to that of a NO (i.e. time response of tubing and CLD) or to that of a NH3 analyzer (as a sticky compound with potentially large flux contribution).

[12] L. 450: The authors state that a general parameterization of alpha was not possible. Still, there are some dependencies of the empirical method with stability and wind speed. While it is difficult to derive a robust parameterization, shouldn't at least be distinguished between night time and daytime alpha values?

[13] L. 493-494: This sentence is misleading as the phase shift is obviously not the only cause for steep decay in the high frequency range. Rather it would be important to state here what the percentage contributions of both transfer functions are to the overall alpha values.

[14] L. 516-518: From the results shown in Figure 6, there seems to be clear differences between stable/unstable conditions for both sites, as well as a dependency with wind speed at a BOG site. To me, these differences/trends/dependencies – despite some uncertainties - should be mentioned in the conclusions.

ADDITIONAL COMMENTS

L. 34-35: Add that these measurements were mainly for NH3.

L. 36: "...which have...": from the sentence structure it is not clear that the authors are referring to the QCL and TRANC analyzers. I suggest making a new sentence here.

L. 79-83: What were the concentration ranges of Nr species observed at the site? They should be mentioned as well as it was done for the forest site.

L. 84-86: To make it easier to follow, I would to state which of the previously mentioned compounds is converted at each step.

L. 89: Since the sensor separation distance is very critical for the presented study, I suggest referring already here to Table 1.

L. 119-124: Since the time lag determination is influenced by the damping of high frequencies, it would be important to mention here what the observed variation of calculated time lags were.

L. 124: Did the authors use the range of the time lag computation as filtering criteria? If so, this should be stated here clearly since it is not stated in Sect. 3.1.

L. 140-141: Use "see above" instead of Sect. 2.2.

L. 164: Since the authors describe the response of a first-order system, better describe tau_r as the "time constant", since the "response time" can be interpreted as a multiple of that.

L. 184-185: I suggest to denote TF_R and TF_deltaR in text and use ":" at the end of sentence.

Figure 1-6: I suggest adding the measurement site (BOR, FOR) in each subplot. This will provide easier readability without having always to refer to the Figure captions. Furthermore use either the full or abbreviated site names consistently in all Figure caption.

L. 218-220: These sentences sound misleading. If the authors used CO2 and H2O data from another eddy covariance setup that was installed in a certain distance from the reactive nitrogen flux measurements, then they did apply IPS to the BOG date, just with an additional uncertainty.

L. 230/Figure 2: It should be stated that the periods used to calculate the spectra in Figure 2 are different from the periods in Figure 1.

L. 245-246: The sentences are confusing as it sounds that another analysis was performed and it is not clear on which data set. Instead of using "We further estimated..."

I suggest to connect both sentences, for example like "at measurement sites, for which we estimated the slope...".

L. 250-260: I find this description of Figure 3 difficult to read since it seems to "jump" between positive and negative slopes, different scalars and sites. To make the description more concise one could for example speak of a "bi-modal distribution" of the CO_2 (for both sites) and Nr (in case of BOG) slopes.

L. 268: For alpha, either use "%" or ratio (as shown in Figure 5).

L. 303-304: This seems repetitive information as the IQR ranges of ICO and sICO were already described in L. 275-277.

L. 323-325: Do the authors mean here a differences compared to THEO or a bias "between" IOG, ICO and sICO?

L. 333: Do the authors mean no significant trend of the IQR? Alpha for sICO is decreasing with increasing stability.

L. 350- 410: As this subsection discusses several aspects of the noise effect, it should be divided into several paragraphs.

L. 377-378: "It may be caused...": This sounds like as if the drop in the high-frequency range was caused by white noise, but it is rather that the white noise (occurring at about 1 Hz as stated later).

L. 438-458: This paragraph discusses the general applicability of the correction methods for Nr fluxes, and less the differences between approaches. I suggest therefore to move this discussion to section at the end as it also relates to the Sections 4.3.2 and 4.3.3.

L. 471-473: In this sentence it is not clear whether alpha values from ICO or sICO were overestimated.

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