Response to reviewers' comments - manuscript *AMT-2019-375* "Correcting high-frequency losses of reactive nitrogen flux measurements"

We thank the reviewers for their constructive comments. We have addressed all of them and modified the manuscript accordingly. Our detailed answers are given below. Referee comments are given in italic, the answers in standard font. The comments by Reviewer 1 were numbered from R1.1 to R1.27 titled as other minor comments, and the specific comments of Reviewer 2 range from R2.1 to R2.14. The additional comments start at R2.15 and end at R2.37. The line and figure numbers in the answers, where we will add the new information into the manuscript, refer in this document to the originally submitted version. The text which is enclosed by "..." will be implemented in the revised manuscript.

Response to Reviewer 1

General Comments

The manuscript outlined that high-frequency measurements of reactive nitrogen species – required for eddy covariance observations of reactive nitrogen fluxes - are subject to high-frequency attenuation due to chemical reactions and adsorption/desorption along the intake tubing. The authors investigated five methodologies for correcting these losses, and applied them in a critical manner for two datasets taken over different surfaces. They were able to show that theoretical spectral corrections were lacking in characterizing the chemical losses in the eddy covariance system, and hence concluded that experimentally-derived corrections in the high-frequency range – in particular in-situ cospectral corrections – were the most appropriate method for their dataset.
The manuscript presents a good overview of the usual methods to correct for inlet attenuation with a closed path flux sensor. The conclusion that corrections based on spectral similarity with an in-situ (non attenuated) scalar cospectrum like sensible heat work best, is very reasonable and undoubtedly correct.
Overall, methods and conclusions seem appropriate for terrestrial flux measurements where you have big signals, but will be tricky or impossible to apply when signals are weak. While my work overlaps with eddy covariance flux research, I lack the depth of expertise to thoroughly evaluate the applied methods and their evaluation. The manuscript is overall very well organized and written. It considers and nicely builds on previous literature. Without doubt, the authors are leading experts in this field. This gives me confidence that this part of the paper is of high quality.
While overall the English language is easily understandable and pretty good, the paper would benefit from careful editing by a native English speaker. I believe the Copernicus staff will do that during the proofsetting, which will likely cover this need.

Thank you for your compliments on this work. Your comments and recommendations are answered below.

Other minor comments:

Comment R1.1 *Here are a few pointers for a start: Commas should probably be inserted in lines 2, 9, 12, 21, 22, 36, 42, 59, 61, 77, 116, 119, 119, 129, 129, 131, 131, 132, 135, 138, 140, 146, 148, 180, 187, 190, 196, 197, 202, 215, 220, 242, 245, 247, 247, 252, 254, 256, 269, 273, 273, 277, 279, 284, 284, 284, 287, 289, 290, 296, 314, 316, 317, 337, 330, 337, 340, 341, 360, 366, 382, 396, 403, 439, 440, 444, 451, 452, 463, 498. Please review and consider placing commas according to 'Oxford comma' rule*
Response to R1.1 We added commas in the mentioned lines after the Oxford comma rule.

Comment R1.2 *Abstract: I suggest adding a sentence or two that summarize the value of this work to a general audience*
Response to R1.2 We appended the following sentences at the end of the abstract:
"Flux measurements of reactive nitrogen compounds are of increasing importance to assess the impact of unintended emissions and on sensitive ecosystems and to evaluate the efficiency of mitigation strategies. Therefore, it is necessary to determine the exchange of reactive nitrogen gases with the highest possible accuracy. This study gives insight in the performance of flux correction methods and their usability for reactive nitrogen gases."

Comment R1.3 *Line 13. I recommend using past tense (underestimated) here and in similar situations (Line 101, ...)*
Response to R1.3 Changed.

Comment R1.4 *Line 75: Please provide detail about the sampling inlet.*
Response to R1.4 We added the following sentences to the corresponding section (Line 83):
"The sampling inlet was designed after Marx et al. (2012) and Ammann et al. (2012). The inlet tube is 15 cm long, consists of FeNiCr, has an outer diameter of 1/4", and is actively heated from the edge of the tube. Inner temperatures are higher than 100°C".
Further details about the sampling inlet are given in the cited publications.

Comment R1.5 *Line 92: This study may also be of relevance: "Characterization and mitigation of water vapor effects in the measurement of ozone by chemiluminescence with nitric oxide", by Boylan, P.; Helmig, D.; Park, J. -H., ATMOSPHERIC MEASUREMENT TECHNIQUES Volume: 7 Issue: 5 Pages: 1231-1244 Published: 2014*
Response to R1.5 We thank the reviewer for the literature advice. The instrument presented by Boylan et al. (2014) is a custom-built chemical luminescene analyzer and suited for the detection of ozone. They determined a reduction in sensitivity of 4.15% in ozone signal per 10 mmol mol^{-1} water vapor. The reduction in sensitivity of our CLD was determined to 0.19% per 1 mmol mol^{-1} water vapor increase by Marx (2004). Both values are comparable. Since the instrument is not the same as the one we used, a comparison would be of low value and we decided not to mention the ozone analyzer. Brümmer et al. (2013) and Ammann et al. (2012) discussed the impact of water vapor on nitrogen concentrations measured with the CLD TR780 and give an equation for the correction of estimated fluxes.

Comment R1.6 *Lines 204, 213, 221: Can the authors provide reasons for choosing their frequency cutoff limits?*
Response to R1.6 The limits were chosen with regard to former studies and subjective decisions explained in the following lines. The limits of the fitting range for IOG were based on the values and suggestions of Ammann et al. (2006) and Ferrara et al. (2012). The missing information about the examination of the power spectral cut-off limit has been added to Sec. 2.3.5.
"The value for the 'lowest noise frequency', which was set in EddyPro for running IPS, was a subjective decision based on visual screening through power spectra. Therefore, we calculated slopes of ΣN_r power spectra in the inertial subrange and estimated the frequency, at which noise started to increase and slopes got positive."
Zöll et al. (2016) conducted NH_3 flux measurements based on the EC method in close proximity to our tower. The cut-off limits of their high-frequency damping analysis were similar to our values (personal communication), which is probably related to the high amount of NH_3 in the ΣN_r signal (Hurkuck et al., 2014; Zöll et al., 2016). Zöll et al. (2016) estimated the attenuation of their EC system with the ogive method.
In case of cospectral approach, the following lines were added to Sec. 2.3.5.
"The decision of the lower frequency limits were further proven by the examination of the ogives ratio, which shows constant values in a certain frequency range. The position exhibits the frequency, at which high-frequency attenuation mostly starts to increase."

Comment R1.7 *Figs. 4+5: Values for n are in awkward location - perhaps at the very top or bottom of figure?*
Response to R1.7 We changed the position of the values and placed them at the top of the figures.

Comment R1.8 *Table 1: Why would be the lag time be different by a factor of 8, when the tubing length is different by a factor of 4, at similar flow rates?*
Response to R1.8 The dimensions of the critical orifice were different causing the different lag times and therefore mostly responsible for the differences. The calculation through the equation for volumetric flow rate and uniform movement is not possible, because the equations do not consider the pressure reduction induced by the critical orifice. The pressure reduction in the tube results in a higher gas flow.

Comment R1.9 *L. 125: Isn't the time lag actually zero given the nature of the measurements?*
Response to R1.9 A time lag for the open-path CO_2/H_2O analyzer arised due to separation distance between the sonic and the gas analyzer. Thus, the time lag was less than a second, but usually greater than zero.

Comment R1.10 *L. 145: So, do I understand this right that actually only ∼10% of the observations were retained for the data analyses after all filters had been applied?*
Response to R1.10 Yes, about 10% of the cospectra were used for the damping analysis. These cospectra passed different filtering criteria ensuring a high quality for the attenuation analysis. We confirm that the number of valid cospectra is not quite high. This is mostly related to physical and chemical properties of ΣN_r (lines 438-443). Due to the variability in concentrations and generally a low concentration level of ΣN_r, the influence of noise on cospectra can vary significantly. Consequently, instruments need to detect very low fluctuations of ΣN_r precisely, which is probably not possible for the instrument (lines 388-393). Inert gases like CO_2 or H_2O have much higher concentrations and distinctive daily cycles. Therefore, the impact of noise on cospectra is much lower, and thus the amount of high-quality cospectra is much higher. Consequentially, the low amount of valid cospectra is related to the complexity of compounds in ΣN_r and to current limitations in the detection limit of the devices.

Comment R1.11 *L 147: I suggest to delete 'as well as'.*
Response to R1.11 Accepted and replaced with 'and'. We further rearranged the sentence and slightly modified it.
"The theoretical slope for power spectra of temperature and inert trace gas concentrations is -2/3."

Comment R1.12 *Figure 4,5: I found it hard to keep track of the many abbreviations used in the text and figures. Maybe the authors could provide a table that lists them all in one place? It would also help to explain/spell out abbreviations again in the Figure caption.*
Response to R1.12 We included a table with all necessary abbreviations at the end of Sec 2.3.
"Table R1 gives an overview about abbreviations used in this study."

Parameter or Term	Abbreviation
Theoretical damping calculation	THEO
In-situ cospectral method	ICO
Semi in-situ cospectral method	sICO
In-situ ogive method	IOG
In-situ power-spectral method	IPS
(Power) spectrum	Ps(..)
Cospectrum	Co(..)
Ogive	Og(..)
Transfer function	TF
response time	τ_{r}
damping factor	α
Bourtanger Moor (semi-natural peatland)	BOG
Bavarian Forest (mixed forest)	FOR
Total Reactive Atmospheric Nitrogen Converter	TRANC
Chemiluminescence dectector	CLD

Table R1: Important terms and corresponding shortcuts used in this study.

Comment R1.13 *Line 241: What are the likely reasons for dampening of temperature power spectrum?*
Response to R1.13 A slight high-frequency damping of Ps(T) can be caused by the path averaging of the sonic anemometer (e.g. Moore, 1986). In addition, the observed shape of the spectrum (slope) can deviate from the theoretical shape due to non-ideal environmental conditions (e.g. non-homogeneous turbulence, influence of roughness sublayer). We added this information at line 241.

Comment R1.14 *Line 265: I would not call these graphics time series plots. They show statistical analyses of monthly data.*
Response to R1.14 Revised. We corrected it according to the reviewer's suggestion. Terms were exchanged to "statistical analyses" at further locations, e.g. line 340, 341, and the corresponding figure captions.

Comment R1.15 *Line 268: "platinum gaze" = "platinum gauze"?*
Response to R1.15 It is platinum gauze.

Comment R1.16 *Line 287: It is not entirely clear in this sentence whether these periods of poor instrument performance were removed or included in the analysis. Perhaps a minor reword to make this clear.*
Response to R1.16 We thank the reviewer for his/her advice. We added some information to line 111 and rephrased the corresponding sentence:
"Periods of maintenance and insufficient instrument performance were removed from damping analysis by manual screening and monitoring performance parameters such as TRANC heating temperature or flow rate."
As a matter of fact, not all affected fluxes can be excluded by the selection criteria. Thus, an

influence on the damping analysis can't be excluded.

Comment R1.17 *Table 2: Consider swapping location of FOR and BOG for consistency with other figures.*
Response to R1.17 Revised.

Comment R1.18 *Line 303 to 305: Appears to repeat already-presented information. Consider beginning paragraph at "For investigating..."*
Response to R1.18 We deleted the corresponding lines.

Comment R1.19 *Fig. 7: Swapped ordering of FOR and BOG again, as opposed to order Fig. 6.*
Response to R1.19 We changed the position of the subplots.

Comment R1.20 *Line 330: Can you explain what the response time actually means? It's obiously not the time from the entry of the sample in the inlet to when the instrument gave a signal?*
Response to R1.20 We agree that the term 'response time' in the manuscript needs further clarification. We added a corresponding paragraph at the end of Sec. 2.3.1:
"Physically, the analyser response time $\tau_{r,a}$ represents the time, at which the difference between the measured signal and the measured quantity is reduced by 1/e after a step change. Thus, it is also called e-folding time. If it is zero, changes will be recognized instantaneously. This is mostly not possible for common gas analysers. Our TRANC-CLD system, which has proven to be suitable for EC measurements (Marx et al., 2012; Brümmer et al., 2013), has an e-folding time of about 0.3-0.35 s. $\tau_{r,a}$ is used for the first-order filter transfer function (Table A1) in the THEO approach. In this manuscript τ_r, which is also called response time, is a fitting parameter used in equation (2). It is linked to the cut-off frequency $f_c = 1/2\pi\tau_r$, at which the cospectrum is damped to $1/\sqrt{2} \approx 0.71$ or the power spectrum to 50%."

Comment R1.21 *Sec 3.3: Could the analysis given between meteorological variables and alpha be repeated for tau? It seems that alpha and tau have an inverse relationship (covered briefly in the conclusions and therefore might it be possible to include this in Sec. 3.2.*
Response to R1.21 According to the reviewers suggestion, we did a statistical analysis of τ_r classified by meteorological variables. We further determined the correlation between monthly averaged τ_r and α. Correlations of -0.83 for BOG and -0.72 for FOR show that there is significant inverse relation between both parameters, which is expected due to the inverse dependency of τ_r in the empirical transfer function. The analysis of τ_r stratified by meteorological variables can be useful in order to investigate, if the scattering in α is related either to the variability in cospectra or to the instrument performance. τ_r is mostly a device-specific parameter. It should have a higher affinity to instrument or measurement setup parameters such as measurement height, pump and heating efficiency, altering of the inlet, and sensitivity of the analyser than to atmospheric, turbulent variations. Changes in gas concentrations may also affect τ_r. Therefore, we classified the meteorological parameters into bins, calculated τ_r for each bin and display them as box plots. The box plot is shown below.

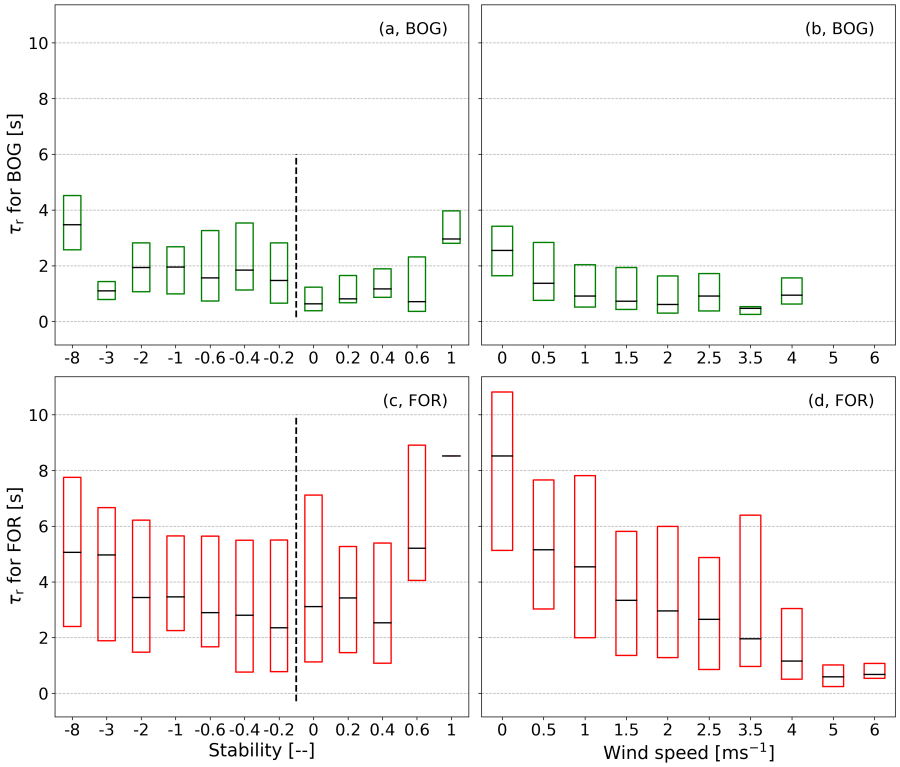

Figure R1: Dependency of the response time (τ_r) on stability and wind speed classes as box plots without whiskers and outliers (box frame = 25 % to 75 % interquartile range (IQR), bold line = median). (a) and (b) refer to the BOG site and (c) and (d) to the FOR site.

τ_r is mostly constant for medium and high wind speed at BOG and exhibit slightly higher values at low wind speeds (0-0.5 m/s). During highly stable and unstable conditions τ_r reaches up to 3.5 s. It seems rather constant during medium, unstable conditions, but increases under stable conditions. The same is valid for τ_r at FOR. τ_r exhibits highest values under both highly unstable and stable conditions. However, τ_r is strongly affected by wind speed at FOR. It decreases with wind speed and seems to follow a non-linear relationship. The analysis confirms the statements of our conclusions that the usage of constant τ_r or α after meteorologically classified parameters is problematic. Generally, it is not known how much the variability in τ_r contributes to the scattering in α for certain wind speeds or stability values. Thus, usage of τ_r classified by meteorological parameters is only recommended for medium or high wind speeds at BOG or near-neutral and unstable atmospheric conditions at both sites. As mentioned in the conclusion, using a constant τ_r is problematic due to its variation with time, which is probably related to the instrument performance or changes in the composition of ΣN_r.
For covering the additional aspects, we extended the results Sec. 3.3 and introduce a new section in the discussion. In Sec. 4.3 we give recommendations for correcting high-frequency flux losses of ΣN_r, for example, which correction factor seem to be most suitable to correct our flux data. Additionally, we moved parts of line 438 to 458 to this subsection (see comment of Reviewer 2, R2.36).

Comment R1.22 *Line 364: Correct to 'ppb'.*
Response to R1.22 Corrected.

Comment R1.23 *Line 371: Can a similar analysis to that in the later section of Sec. 3.2*

be used to investigate the influence of humidity on CO2?
Response to R1.23 In principle, it is possible for CO_2 or other trace gases, too. However, such an analysis is beyond the scope of this study, which is focused on ΣN_r rather than on CO_2. The determined slopes of Ps(CO_2) can be separated into different humidity classes and displayed as box plots. A similar analysis could be done for the CO_2 damping factors. A comparison may help to investigate the impact of white noise on the damping under wet or humid conditions.

Comment R1.24 *Line 379: How can a logger contribute to noise?*
Response to R1.24 We noticed the mistake, deleted the logger from the enumeration, and changed the enumeration as follows: "[..] components of the setup like pump, air-conditioning system or electrical components."

Comment R1.25 *Line 486: Correct to 'low-frequency'.*
Response to R1.25 Revised.

Comment R1.26 *Line 499: Based on your results and experience, can you comment on the possible complication from water vapor and NO having different lag times in the converter and sampling system, and how that then affects the interference from quenching of the NO signal?*
Response to R1.26 The effect of water vapor quenching on the NO signal is very small, which is caused by the low sensitivity reduction of 0.19% per $1\,\mathrm{mmol\,mol^{-1}}$ water vapor. The impact on determined fluxes is corrected by the procedure given in Brümmer et al. (2013) and Ammann et al. (2012). Brümmer et al. (2013) found only $25\,\mathrm{g\ N\ ha^{-1}}$ higher deposition during 11-month field campaign. We measured ΣN_r exchange above a remote, mixed forest for 2.5 years. According to Brümmer et al. (2013), we got a shift towards less deposition by approximately $100\,\mathrm{g\ N\ ha^{-1}}$ on the remaining quality-controlled fluxes. Total dry nitrogen deposition was around $12\,\mathrm{kg\ N\ ha^{-1}}$ during the 2.5 year field campaign. The correction is only applied to the determined fluxes.

Comment R1.27 *Conclusion section: It was not clear to me whether the damping factor values transform directly into flux value reductions. Therefore, as the results/discussion was related largely to damping factor values, the introduction of flux value reductions within in the conclusions seemed somewhat new. I would also suggest here to place again these flux reduction values within the previous literature, as cited earlier in the paragraph beginning in line 45.*
Response to R1.27 We agree with the reviewers comment and added the presented lines to the results to differentiate between damping factors and flux loss values (line 300). Furthermore, we put the citations of the previous literature in the conclusion (line 510):
"By subtracting the damping factor from an ideal, unattenuated system, which has a damping factor of one, the result will be the flux loss value (=1-α). This loss value shows how much of the signal is lost from the inlet to the analysis of the signal by the instrument. Thus, flux losses calculated by IPS for our TRANC-CLD setup are around 6% at BOG and around 5% at FOR. The flux loss after THEO was approximately 12% at BOG and about 5% at FOR. The methods using measured cospectra or ogives (ICO, sICO and IOG) showed a flux loss of roughly 16-22% for FOR and around 26-38% for BOG. ICO shows the strongest damping at both sites."

Response to Reviewer 2

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 H2O and CO2 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.

We thank the Reviewer for his/her comments and criticism on this work. We addressed all mentioned points and implemented most of the suggestions in the revised manuscript. A detailed reply to your comments is given below.

Specific comments

Comment R2.1 *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.*
Response to R2.1 We added it to the corresponding chapter:
"Moravek et al. (2019) proposed a new approach for correcting high-frequency flux losses of NH_3 measured by a QCL. The method is based on frequently measuring the analyzer's time response. The application of this method resulted in 46% flux loss."

Comment R2.2 *L. 173-174, Figure 1+Figure 2: 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 with the same described features were observed. Furthermore, is there a reason why different days were chosen for the cospectra in Figure 1 and 2? If so, these should be highlighted.

Response to R2.2 On average, wind speed and stability were approximately 1.65 ms^{-1} and -0.22 at BOG during daytime. At FOR, the average wind speed and stability were 1.91 ms^{-1} and -0.44 during daytime. Wind speed conditions of the averaged power spectra and cospectra are almost similar to the average values during daytime for the entire period. Stability values of the displayed case are in agreement with daytime average for BOG. At FOR, the shown example refers to stable conditions whereas an unstable average is exhibited during daytime. In general, daytime stability values of both sites are rather low and close to neutral conditions. At both sites, approximately 10% of the analyzed cospectra were in the range of $\pm 0.5\,\mathrm{ms}^{-1}$ to the average wind speed and ± 0.15 to the average stability. Using only the wind speed restriction resulted in 40% agreement at FOR and 55% at BOG. It seems that the stability is more diverse and not correlated to wind speed. The correlation between wind speed and stability for the analyzed cospectra used for the damping analysis is rather low for both sites (0.26 for BOG and 0.15 for FOR). The choice of different days was caused by data gaps in the measurements.

The discussion of the long-term wind speed and stability averages compared to the exemplary case were added to the manuscript beginning at line 178.

Comment R2.3 *L. 182-193: How does this approach compare to the approach by Aubinet et al. (2000), which uses a normalisation 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 were performed can be improved. For example, it should be mentioned what kind of of a least-square fit was performed, linear, non-linear?*

Response to R2.3 We improved the description of the ICO method and the estimation of α according to the suggestion of Reviewer 2. The following sentences were added after line 185.

"The approach used in this study is somewhat different to other methods that are also based on using measured cospectra of heat and gas flux, for example the method of Aubinet et al. (1999). The latter uses a normalisation factor, which corresponds to the ratio of the heat flux cospectrum to gas flux cospectrum. Both cospectra are integrated until frequency f_o, which should not be affected by high-frequency damping, but high enough to allow an accurate calculation of the normalisation factor. However, the definition of f_o is rather imprecise and thus, an incorrect setting of f_o can lead to significant uncertainties in the damping analysis. In our approach cospectra are normalized by their corresponding total covariance. In order to consider the damping of the gas flux cospectrum and its covariance, the damping factor is introduced in Eq. (2). Thus, we assume that both approaches give similar results, since both approaches cover the damping of the gas flux cospectrum."

We also expand Eq. (1) to clarify the definition of α (line 155). Furthermore, we add the following flow chart to support the descriptive text about the procedure for determining α after ICO.

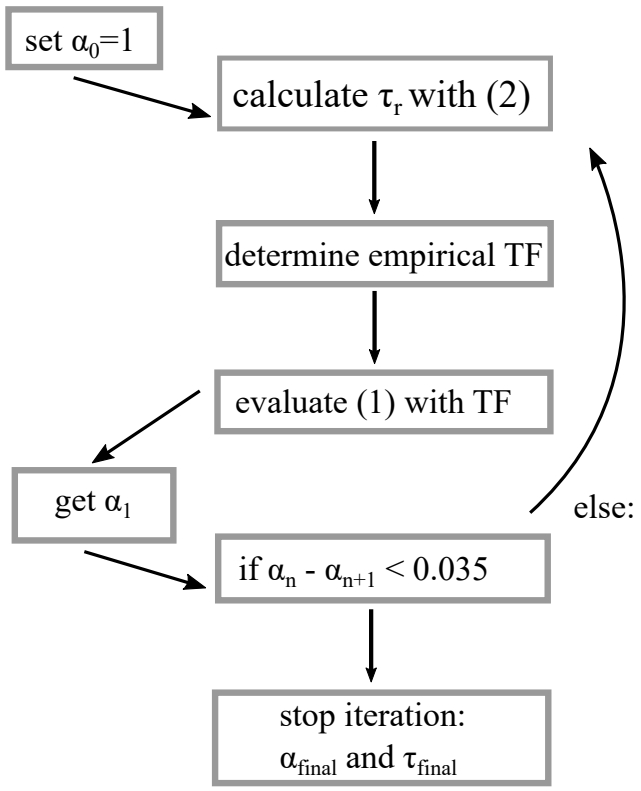

Figure R2: Illustration of the calculation of α and τ_r by ICO.

Comment R2.4 *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.*
Response to R2.4 We add the mathematical definition of the ogive to the corresponding section (line 200). We rewrite line 207 to further specify the optimization factor.
"Finally, the optimization factor, which minimizes the difference between $\mathrm{Og}(w, \Sigma \mathrm{N_r})$ against $\mathrm{Og}(w, T)$, is the result of the least-squares problem and corresponds to the damping factor."

Comment R2.5 *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 (one fc is known), the methodology is different fro the other approaches used here, that determine alpha more directly. The mean horizontal wind speed (u) should be defined in the text.*
Response to R2.5 The following sentences were added to the Sec. 2.3.5 (after line 224).
"In general, the idea of IPS is that the EC system can be simulated by a recursive filter. Thereby, α is determined by the ratio of the unfiltered covariance $\overline{w'T'}$ to the filtered covariance $\overline{w'T'_f}$, and applying the recursive filter to degrade the time series of sonic temperature (Ibrom et al., 2007). However, Ibrom et al. (2007) argued that this ratio gives erroneous results for small fluxes. Therefore, they parameterized α by the mean horizontal wind speed ($\bar{u}$), stability, and f_c. Ibrom et al. (2007) investigated a proportionality between α^{-1} and $u \cdot f_c^{-1}$. By introducing a proportionality constant A_1 and a second constant A_2, which should consider for spectral properties of the time series, the following equation for calculating the correction factor was proposed (for details see Ibrom et al., 2007, Sec. 2.4):."

Comment R2.6 *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 in an increased flux contribution, 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*
Response to R2.6 We specified that the slope of Ps(T) was estimated for the inertial subrange (line 240) and corrected the sentence about the interpretation of the slope value (line 241). The fitting range used for the derivation of the slopes is smaller than the inertial subrange, for example, to exclude slightly positive slopes at the very high frequencies of the inert trace gases.

Comment R2.7 *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 and cospectra as well?*
Response to R2.7 The precipitation filter was also applied for filtering the lower quality cases of CO_2 and H_2O shown in Fig. 3. Since the TRANC-CLD setup is a closed-path system, the precipitation filter was not applied to ΣN_r measurements. Quality assurance of Ps(T) and Co(w, T) were made by the criteria of Mauder and Foken (2006), which already account for insufficient conditions compromising flux calculation. Due to the stricter criteria for ΣN_r filtering most of the less quality cases were rejected.

Comment R2.8 *L. 370-373: A stronger white noise of Ps(CO_2) at BOG than at FOR is not visible to me in the power spectra presented in Figure 2. Which frequency range are the authors referring to? May the slight increase in the very high frequencies be due to aliasing effect?*
Response to R2.8 We apologise for not including a figure reference here. In fact the text refers to data shown in Figure 3, not Figure 2. We improved the mentioned lines as shown below. The slight increase at very high-frequencies of Ps(CO_2) observed in Fig. 2 is mostly probably not induced by aliasing (of the real flux contributions above 5 Hz), because of the very steep slope and the lack of a similar increase effect in Ps(T) and Ps(H_2O).
"Before, we argued that concentration of ΣN_r leads to differences in the slope distribution. Concentrations of CO_2 were not significantly different between the sites. As a consequence, there has to be another parameter responsible for discrepancy in the contribution of positive Ps(CO_2) slopes at the measurement sites. We suppose that the discrepancy of positive Ps(CO_2) slopes corresponds to different levels of humidity at the measurement sites."

Comment R2.9 *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.*

Response to R2.9 Since the question is quite long and treats several aspects of Sec. 4.1, we divided the answer into several paragraphs. At first, we complemented the lines 390-393 by the discussion about the impact of the instrumental noise:
"Instrumental noise affects the shape of the covariance function. It can lead to a broadening of the covariance peak and generally enhances the scattering of the covariance values. Both effects are already enlarged in case of small mixing ratio fluctuations. Thus, instrumental noise further compromises the time lag estimation and leads to additional noise in cospectra. Due to the applied time lag criterion the effect of instrumental noise is mostly cancelled out. The position of the cospectral peak is less impacted, and thus instrumental noise can only lead to an enhancement of scattering of cospectral values, preferentially in the low-frequency range of the cospectrum. In other words, instrumental noise mostly contributes to low-frequent noise, the red noise."
Based on the reviewer's advice we improved our suggestion using IPS for reactive gases. We added the following aspects to the section about the impact of noise on IPS beginning at line 398:
"However, Fig. 2 reveals that Ps(ΣN_r) shows a steep decline in the high-frequency range after the peak at BOG, which is similar to the decline of Ps(T). ΣN_r concentration was 24.4 ppb on average and exhibits a standard deviation of 9.6 ppb for the averaging period in Fig. 2 suggesting significant differences in concentration levels. It confirms the statement that the concentration is an important driver for the quality of Ps(ΣN_r). This leads us to the assumption that the instrument was in principle able to capture differences in concentration levels in the high-frequency range if mixing ratio fluctuations are relatively high."
Basically, for time periods with remarkable differences in concentration levels IPS should perform well. Yet, the opposite is the case which is probably related to the negligible difference in the slopes of Ps(ΣN_r) and Ps(T), and to the violation of the critical assumption requesting spectral similarity of the power spectra (Fig. 2). This assumption is clearly not valid for Ps(ΣN_r) due to the low-frequent noise.
"Consequently, for estimating damping factors with IPS certain conditions seem to be fulfilled. For example, instruments need a low detection limit for detecting low turbulent fluctuations, sources and impact of noise and strategies for the elimination of noise should be known, gases should be rather inert or have little interaction with surfaces or other chemical compounds, and, in case of IPS, show a wind speed dependency on damping factors. Similar to cospectral methods, IPS will also benefit from a well-defined footprint, equal canopy height, and sufficient turbulence. Satisfying these aspects is quite difficult for a custom-built EC system, which is rather new and thus not all attenuation processes are identified, and designed for measuring a trace gas, which consists of several compounds with unknown contribution, complex reaction pathways, and generally low fluctuations." Our conclusion is that IPS seems not be the optimal choice for correcting high-frequency flux losses of ΣN_r due to large red and white noise.

Comment R2.10 *L. 397-398: The authors state that under conditions with "less variability in concentrations and deposition" fluxes the IPS method fails. Shouldn't it be just under low flux conditions (regardless whether deposition or emission fluxes prevail)?*
Response to R2.10 We agree with the reviewer's comment and correct the sentence as follows:
"Therefore, we recommend using cospectra to estimate τ_r and α of reactive gases, since these gases exhibit normally low density fluctuations."

Comment R2.11 *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 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 walls 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 it is important to discuss since it would show whether the system's time response was more similar to that of 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).

Response to R2.11 We agree with the reviewer that such a discussion is helpful for understanding, which part of the TRANC-CLD setup has the strongest impact on the system's time response and damping. We rephrased the mentioned lines and consider the new aspects in the revised manuscript as follows:

"There could be other effects which superpose the wind speed and stability dependencies, for example, (chemical) damping processes occurring inside the TRANC-CLD system. Humidity and ΣN_r could affect the aging of the tube and consequentially the adsorption at inner tube walls. However, we found no dependency of these parameters on damping factor and time response. Interactions with tube walls is probably less important, especially for the tube connecting the end of the TRANC to the CLD, because the main trace gas within the line is NO, which acts rather inert in the absence of ozone and NO_2. Because NO_2 and O_3 are converted in the TRANC, it can be assumed that the influence of interaction with tube walls on time response and high-frequency flux losses is mostly negligible compared to effects, which happen in the CLD and TRANC. The CLD contributes more to the total attenuation than the tubing, but supposedly not as much as the TRANC. Rummel et al. (2002) also used the CLD 780 TR as device for measuring NO fluxes. High-frequency flux losses were rather low and ranged between 21% (close to the ground) and 5% (11 m above ground). Also, Wang et al. (2020) observed low flux losses of NO by approximately 12% by measuring with a QCL (1.7 m above ground). Consequently, the strongest contributor to the overall damping has to be the TRANC. NH_3 is, considering all possible convertible compounds, the most abundant in certain ecosystems, highly reactive, and rather "sticky". In absolute terms it has the highest influence on the damping of ΣN_r. QCL devices, which may be used for the detection of NH_3 (Ferrara et al., 2012; Zöll et al., 2016; Moravek et al., 2019), were equipped with a special designed, heated, and opaque inlet to avoid sticking of NH_3 at tube walls, water molecules, and preventing unwanted molecules entering the analyser cell. Thus, NH_3 has high flux loss factors ranging from 33 to 46% (Ferrara et al., 2012; Zöll et al., 2016; Moravek et al., 2019). These damping factors are closer to the damping factors of ΣN_r, in particular for BOG, at which high NH_3 concentrations were measured and most of ΣN_r can be attributed to NH_3 (Hurkuck et al., 2014; Zöll et al., 2016). At FOR, flux losses were lower due to physical reasons (Sec. 4.1, lines 393-394) and due to lower contribution of NH_3 to ΣN_r at FOR (Sec. 4.2.1, lines 447-449). NH_3 is converted inside the TRANC at the platinum gauze after passing through the actively heated inlet and iron-nickel-chrome (FeNiCr) alloy tube. Since the main part of the pathway is heated and isolated against environmental impacts, the inlet of the TRANC and the distance to the sonic seem to be critical for the detection and attenuation of NH_3. Finally, we suppose that the response time and attenuation of our TRANC-CLD system is more similar to that of an NH_3 analyzer under a high ambient NH_3 load."

Comment R2.12 *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 day time alpha values?*

Response to R2.12 The motivation behind this formulation was that there was no explicit dependency on the investigated parameters. Dependencies on wind speed and stability were only valid for certain ranges and under certain site conditions. For example, a parameterization can be performed for unstable conditions and for wind speeds above $1.5\,\mathrm{ms}^{-1}$ at BOG. As mentioned in the manuscript, other parameters showed no clear dependency on α. We classified α in different radiation classes, but found no significant difference between day and night time values. The exchange pattern of ΣN_r is rather bi-directional during the entire day. The exchange pattern of inert gases like CO_2 is largely related to photosynthesis and respiration. During daytime CO_2 exhibits also bi-directional exchange characteristics. During the night the exchange of CO_2 is mostly unidirectional. Thus, we would expect a diurnal variation of the CO_2 attenuation. The influence of global radiation on the biosphere-atmosphere exchange of ΣN_r and CO_2 was explicitly shown by (Zöll et al., 2019) for FOR. They also investigated drivers of ΣN_r. However, global radiation explained only 22% of the variability in ΣN_r fluxes, whereas 66% of the variability in CO_2 fluxes were related to global radiation. ΣN_r had the concentration as a second driver, which was approximately 24%. Consequently, there are additional factors controlling the biosphere-atmosphere exchange of total reactive nitrogen, which may be of chemical nature and challenging to quantify. We revised the mentioned sentence and expanded the discussion with certain aspects written here.

Comment R2.13 *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.*
Response to R2.13 We rephrased the sentence to clarify the statement: "The inclusion of the shift mismatch in Eq. (3) leads to a steeper slope in the empirical transfer function and variations around zero at higher frequencies (see Fig. 1) compared to a first-order function alone (not shown). If α is calculated without phase-shift effect, we get an overestimation of the damping up to 10% for both sites. This could be expected and indicates that most of the damping is related to a time shift."

Comment R2.14 *L. 516-518: From the results shown in Fig. 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.*
Response to R2.14 We agree that the mentioned lines sound quite general (lines 518-520) and therefore, added some details to the description.
"In case of the empirical methods, we found a wind speed dependency on damping factors (α), apparently a linear decrease in α with increasing wind speed at BOG. However, the trend is limited to wind speeds higher than $1.5\,\mathrm{ms}^{-1}$. At FOR, α of IOG, sICO, and ICO seem to be invariant to changes in wind speed. For unstable cases α values are rather constant at FOR (~ 0.85). At BOG, α of IOG and ICO were similar and vary between 0.6 and 0.8 at unstable conditions, whereas sICO values were higher by approximately 0.05-0.15. The expected decline of α with increasing stability was only observed in sICO at both sites, probably related to the usage of Kaimal cospectra. IOG and ICO showed no clear trend for stable cases."

Additional comments

Comment R2.15 *L. 34-35: Add that these measurements were mainly for NH3.*

Response to R2.15 Revised.

Comment R2.16 *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.*
Response to R2.16 Revised and changed to: "Both measurement systems..."

Comment R2.17 *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.*
Response to R2.17 Information about the concentration levels were appended to the site characteristics of the semi-natural peatland site (line 81).
"Average NH_3 concentrations ranged from 8 to 22 ppb, HONO was mostly below 0.1 ppb, HNO_3 had an average concentration of 0.04 ppb, NO was approximately 3.6 ppb, and NO_2 showed 8.6 ppb on average (Hurkuck et al., 2014; Zöll et al., 2016). Concentrations of NO and NO_2 were provided by the 'Air Quality Monitoring Lower Saxony' (Lower Saxony Ministry of Environment, Energy and Climate Protection) (for data availability please see `https://www.umwelt.niedersachsen.de/startseite/themen/luftqualitat/lufthygienische_uberwachung_niedersachsen/aktuelle_messwerte_messwertarchiv/`)."

Comment R2.18 *L. 84-86: To make it easier to follow, I would state which of the previously mentioned compounds is converted at each step.*
Response to R2.18 We thank the reviewer for his/her advise and added the following information to the revised manuscript (Line 85 and Line 86):
"Inside the FeNiCr tube, NH_4NO_3 is thermally split up to gaseous NH_3 and HNO_3. HNO_3 is thermally converted to NO_2, H_2O, and O_2. NH_3 reacts at a platinum gauze with O_2 to NO and H_2O. HONO is thermally split up to NO and a hydroxyl radical. [...] resulting in a reduction of the remaining nitrogen compounds, NO_2 and other higher nitrogen oxides, to NO inside the gold tube."

Comment R2.19 *L. 89: Since the sensor separation distance is very critical for the presented study, I suggest referring already here to Table 1.*
Response to R2.19 Revised.

Comment R2.20 *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 time lags were.*
Response to R2.20 According to the reviewer's suggestion, we appended the additional information written here to line 124.
"The chosen range for the time lag computation coincides with range of the highest time lag density. The variation of time lags around the physical lag were almost constant for both measurement campaigns and not correlated to the temporal variation of the damping factors. The difference in ranges may be related to different site characteristics, different mixing ratio fluctuations of ΣN_r compounds at the sites, and performance of the TRANC-CLD setup."

Comment R2.21 *L. 124: Did the authors use the range of time lag computation as filtering criteria? If so, this should be stated here clearly since is it not stated in Sect. 3.1.*
Response to R2.21 The reference to Sec. 3.1 in the discussion manuscript is misleading and was deleted. Of course, the time lag window was used for the whole analysis. In the revised manuscript the time lag criterion used for the slope distribution is identical to the one used for

the determination of α.

Comment R2.22 *L. 140-141: Use "see above" instead of Sect. 2.2.*
Response to R2.22 Revised.

Comment R2.23 *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.*
Response to R2.23 Based on the comments of Reviewer 1 and Reviewer 2 to the same topic, we added some text in order to clarify the difference between the analyzer response time $\tau_{r,a}$ and the empirical cospectral response time τ_r used in this manuscript. Since the term "response time" is well established to characterize instrument performance, we consider it as better suited for the present purpose than the term "time constant", which is very unspecific. In addition, it could be misleading, since τ_r is not really constant, considering the results of Fig. 7. Also, Moravek et al. (2019) measured a significant variability in $\tau_{r,a}$ during their measurement campaign.

Comment R2.24 *L. 184-185: I suggest to denote TF_R and TF_deltaR in text and use ":" at the end of the sentence*
Response to R2.24 Revised.

Comment R2.25 *Figure 1-6: I suggest adding the measurement site (BOG, FOR) in each subplot. This will provide easier readability without having always to refer to Figure captions. Furthermore, use the full or abbreviated site names consistently in all Figure captions.*
Response to R2.25 We added the site names and improved the figure captions.

Comment R2.26 *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 BOG data, just with an additional uncertainty.*
Response to R2.26 We agree. We rephrased the mentioned lines according to the reviewer's recommendation:
"Using the IPS through EddyPro for ΣN_r at BOG requires CO_2 and H_2O measurements. Since both inert gases were not measured at ΣN_r tower, we used CO_2 and H_2O data from the EC setup described in Hurkuck et al. (2016), which was placed next to the ΣN_r setup. Then, the application of IPS to ΣN_r at BOG was done, thereby inducing additional uncertainty."

Comment R2.27 *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 Fig. 1.*
Response to R2.27 Revised.

Comment R2.28 *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.*
Response to R2.28 Revised. The analysis of the slope distribution was done after we had noticed that a significant amount of $Ps(\Sigma N_r)$ was affected by white noise. Thus, we applied the filtering criteria to exclude rather low fluxes, instrument performance issues, and conditions of insufficient

turbulence, which could have been responsible for the observed white noise.

Comment R2.29 *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.*
Response to R2.29 We rearranged and rephrased the mentioned lines.

Comment R2.30 *L.268: For alpha, either use "%" or ratio.*
Response to R2.30 Done.

Comment R2.31 *L. 303-304: This seems to be repetitive information as the IQR ranges of ICO and sICO were already described in L. 275-277.*
Response to R2.31 These sentences are deleted in the revised manuscript.

Comment R2.32 *L. 323-325: Do authors mean here a difference compared to THEO or a bias "between" IOG, ICO and sICO?*
Response to R2.32 Here, the bias with THEO is meant. We corrected the corresponding sentence and rearranged the description sightly.

Comment R2.33 *L. 333: Do the authors mean no significant trend of the IQR? Alpha for sICO is decreasing with increasing stability.*
Response to R2.33 We specified the description in the revised manuscript as follows:
"During stable situations ICO, IOG, and sICO exhibit no distinct trend through all positive stability classes. Only for stability values above 0.4 a decrease in α is visible. However, this decline in α is rather uncertain, since the IQR is relatively large compared to the unstable classes and the amount of cospectra, which are attributed to stable conditions, is relatively small."

Comment R2.34 *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 1Hz as stated later.*
Response to R2.34 We replaced "may" by "most likely". The words were chosen in such a way because we do not know, which component or combination is most responsible for the observed uncorrelated noise. For this reason, we mentioned several components, which may be sources of uncorrelated noise such as the pump or electrical components.

Comment R2.35 *L. 350-410: As this subjection discusses several aspects of the noise effect, it should be divided into several paragraphs.*
Response to R2.35 We agree with the reviewer's suggestion. For a better readability we separated Sec 4.1 into three subsections. The first paragraph is called "Sources of spectral noise (Lines 350-374), the second "Impact of noise on power spectra and cospectra" (Lines 374-395), and the third "Impact of noise on IPS" (Lines 395-411). The second and third paragraph were extended due to the reviewer comment R2.8.

Comment R2.36 *L. 438-458: This paragraph discusses the general applicability of the correction methods for Nr fluxes, and less the differences between the approaches. I suggest therefore to move this discussion to the section at the end as it also relates the sections 4.3.2 and 4.3.3.*

Response to R2.36 For implementing the discussion about the expected high-frequency damping processes (R2.8), some information of the mentioned lines are also needed, but in total we agree with the reviewer's comment. We moved parts of the text and introduced a section about the applicability of the correction methods for ΣN_r fluxes after Sec. 4.2.3.

Comment R2.37 *L. 471-473: In this sentence it is not clear whether alpha values from ICO or sICO were overestimated.*
Response to R2.37 We clarified to: "...if Kaimal cospectra (sICO) were used."

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