Response to reviewers‘ comments – manuscript BG-2020-364 Forest-atmosphere exchange of reactive nitrogen in a low polluted area – Part I: Measuring temporal dynamics

We thank again the anonymous referees for their comments to revised version. We recognized that the discussion about (effective) canopy resistances ($R_{c,eff}$) and deposition velocities (v_d) was too speculative and shortened it substantially. We added results on wet deposition measurements and clarified misleading statements to the DELTA denuders. Finally, we improved the discussion about the deposition occurred in February 2018 with own measurements on particulate nitrogen.

Comments of Referee 1 range from R1.1 to R1.26, Comments of Referee 2 range from R2.1 to R2. 73. Line numbers in the answers, where new information was added to the manuscript, refer to the original submitted version. The text which is enclosed by "…" is implemented in the manuscript.

Response to Referee 1

General Comment: *Based on initial reviews, the authors have eliminated the modeling component, which will be covered in a forthcoming separate manuscript, to focus only on the measurement component of the study. Many of initial review comments regarding the measurements were addressed but some technical weaknesses remain, as outlined below. Second, the overall writing of the manuscript, specifically the grammar and sentence structure, has not significantly improved. While some examples for improvement of the writing are included below, there are many more instances, the comprehensive correction of which is outside the scope of a scientific technical review. While I believe this dataset and its analysis can make a contribution to the literature, and is appropriate for Biogeosciences, treatment of the remaining technical issues (as outlined below) and readability must be addressed before the paper is suitable for publication.*

We thank the Reviewer for his/her comments on the revised version. We addressed all of your remarks and implemented your suggestions in the manuscript

Comment R1.1: *Section 2.2: The authors note different heights for the various measurements: 30m for ∑Nr and Delta, various heights for passive NH3, 30m for QCL NH3, and 50m for NO and NO2. The NO and NO2 measurements were used for assessment of Nr speciation. Did the authors investigate the potential magnitude of the NOx gradients between 30m and 50m? Since NOx appears to be the primary Nr component, some mention of the importance of differences in measurement height is warranted.*

Response to R1.1: Since no NO_2 and NO measurements were conducted at 30 m, no concentration gradient was calculated between 50 m and 30 m. "Seok et al. (2013) found highest NO_x concentrations above the canopy but their concentrations differences were negligible. Since both measurement heights were above the canopy, no correction was applied to NO_2 and NO concentration measurements." We added these sentences to line to 195 and discussed the influence of the different heights with regard to NO_2 and NO at line 527ff.

Comment R1.2: *Line 155: Can the authors give an explanation for the NH4NO3 efficiency of 142%?*

Response to R1.2: Marx et al. (2012) provided reasons for their results. NH_4NO_3 is semi-volatile under ambient conditions and can evaporate to NH_3 and HNO_3. Both gases are detected by the TRANC and influence the conversion efficiency of NH_4NO_3. Ambient air was used to clear the analysis chamber of the CLD and to transport the aerosol, resulting in a N_r background ranging from 20 to 39 ppb. The latter was corrected by the authors. For further details we refer to the publication of Marx et al. (2012).

Comment R1.3: *Line 182: Change "exposition" to "exposure".*

Response to R1.3: Done.

Comment R1.4: *Line 198: A brief description of the methods for wet and bulk deposition is needed, including organic nitrogen. This can be added to Supplemental.*

Response to R1.4: "Wet-only and bulk deposition were collected by four samplers, one wet-only and three bulk samplers, at an open site. The measurements took place in southwest direction of the tower (approx. 1.3 km). Bulk samplers had a funnel opening of 321 cm^2 at 1.25 m above ground. The automatic wet-only sampler (NSA 181K – cooled, Eigenbrodt, Königsmoor, Germany) had a funnel opening of 500 cm^2 at 2 m above ground. During the weekly sampling intervals, precipitation samples were kept dark and cool (<4°C). After sampling they were filtered (< 0.45 µm, Whatman) and cooled at 2 to 4°C without chemical preservation/treatment until analysis. No biocides were used during sampling because denitrification was unlikely due to the short exposure time and permanent cooling. In fact, we found very low carbon concentrations and no nitrite as an intermediate product of denitrification in the precipitation samples.

NH_4^+ and NO_3^- were analyzed following DIN EN ISO 10304-1. Determination of total wet N was done according to DIN 38409-27 and EN 12260. Dissolved organic nitrogen is calculated by subtracting NH_4^+-N and NO_3^--N from total wet N." We provided this description as a supplement and named it A1.

Comment R1.5: *Line 238: "Pump efficiency was controlled…". Do the authors mean "Pump efficiency was assessed.."? What does it mean to control the efficiency monthly?*

Response to R1.5: "We checked the pressure in the sample cell of the CLD during each, at least monthly, site visit. If the sample cell pressure was outside the allowed range, tip seals of the pump were replaced." We rephrased the sentence according to this response.

Response to R1.6: *Lines 308-320: Consider combining into a single paragraph.*

Comment R1.6: We decided to delete those lines since they were no longer needed for the discussion.

Comment R1.7: *Line 339: Change the phrasing of "almost similar". "Almost" appears in other places in the manuscript and should be avoided.*

Response to R1.7: We deleted the word "almost" at several places or changed the phrasing.

Comment R1.8: *Caption Figure 3: Change "exposition" to "exposure". Consider indicating the year along the x-axis.*

Response to R1.8: Agreed. We changed the word and labels of the x-axis for better readability.

Comment R1.9: Line 379: *I think this sentence is not necessary.*

Response to R1.9: Agreed. We deleted this sentence.

Comment R1.10: *General comment: Words such as "mostly", "almost", "slightly", "mainly" are used throughout the manuscript. In general, they should be used very infrequently in the context of data reporting/interpretation.*

Response to R1.10: Agreed. We reduced the usage of these words.

Comment R1.11: *Figure S8: The caption says the period is May-September. Please correct.*

Response to R1.11: Corrected. It should be December to February.

Comment R1.12: *Line 411: "The analysis of vd and corresponding fluxes show that their diurnal pattern was characterized by lower deposition during the night and highest values around noon, in particular from May to September (Fig. 6 and Fig. S6)" But this was not the pattern during winter, which showed low vd at mid-day (Fig. S8). Please clarify.*

Response to R1.12: "From May to September, a clear diurnal pattern was found for v_d and their corresponding fluxes (Fig. 6 and Fig. S6). It was characterized by lower deposition during the night and highest values around noon (Fig. S9). During winter, deposition fluxes were close to zero and showed no diurnal variation leading to a constantly low v_d during the day (Fig. S10)." We changed the sentence according to this response.

Comment R1.13: *Line 438: "vd and Rc,eff determined during rain were treated separately." Reviewers 1 and 2 both questioned the quality of the TRANC flux measurements during rain and the suitability of the EC method (or any micromet flux method) during rain as the washout process introduces an additional sink below the EC measurement height and therefor a height dependent flux. I believe the*

EC measurements conducted during rain should not be included in this analysis. If the authors retain them, the validity of the EC method during rain must be justified.

Response to R1.13: We agree that a height dependent flux could be induced due to precipitation since many N_r compounds are water soluble. In order to show the impact of precipitation on measured ΣN_r deposition velocities, we made the analysis of controlling factors for deposition velocities and effective canopy resistances during active and no precipitation separately. Possibly, there was a misunderstanding in treating the suggestions to precipitation in the first review round. We only applied a precipitation filter on Figs. 8 and 9 in order to strengthen process understanding of ΣN_r flux measurements.

"In order to avoid uncertainties due to the washout process as it introduces an additional sink below the measurement height leading to a height dependent flux, we applied a precipitation filter on ΣN_r flux measurements (IV)." We deleted discussions and subplots related to the impact of precipitation on the ΣN_r exchange. Since a precipitation filter was introduced (line 232), annual budgets were higher since lower ΣN_r fluxes were excluded from analysis.

Comment R1.14: *Figure 7 caption: "Dependency of measured concentrations on corresponding ΣNr fluxes…". Consider changing to "Relationships between measured concentrations and corresponding ΣNr fluxes…".*

Response to R1.14: We applied your suggestion to Figs. 4, 5, and 6. Please note that Fig. 7 of the original submitted manuscript was deleted.

Comment R1.15: *Table 2 and associated paragraphs. Because the TRANC is not Nr species specific, its primary strength is in quantifying the total Nr dry deposition to facilitate a total deposition calculation when combined with wet deposition. The authors have done this but some additional detail on the variability of the relative fractions of wet versus dry deposition would be helpful. Can the authors add a table or pie charts showing the total wet and dry fluxes and fractional contributions of wet versus dry deposition by year and summarized by overall annual and seasonal periods?*

Response R1.15: We appreciate the Reviewers suggestion. Figure 1 shows relative fractions of wet and dry deposition to total deposition for each season and for both measurement years.

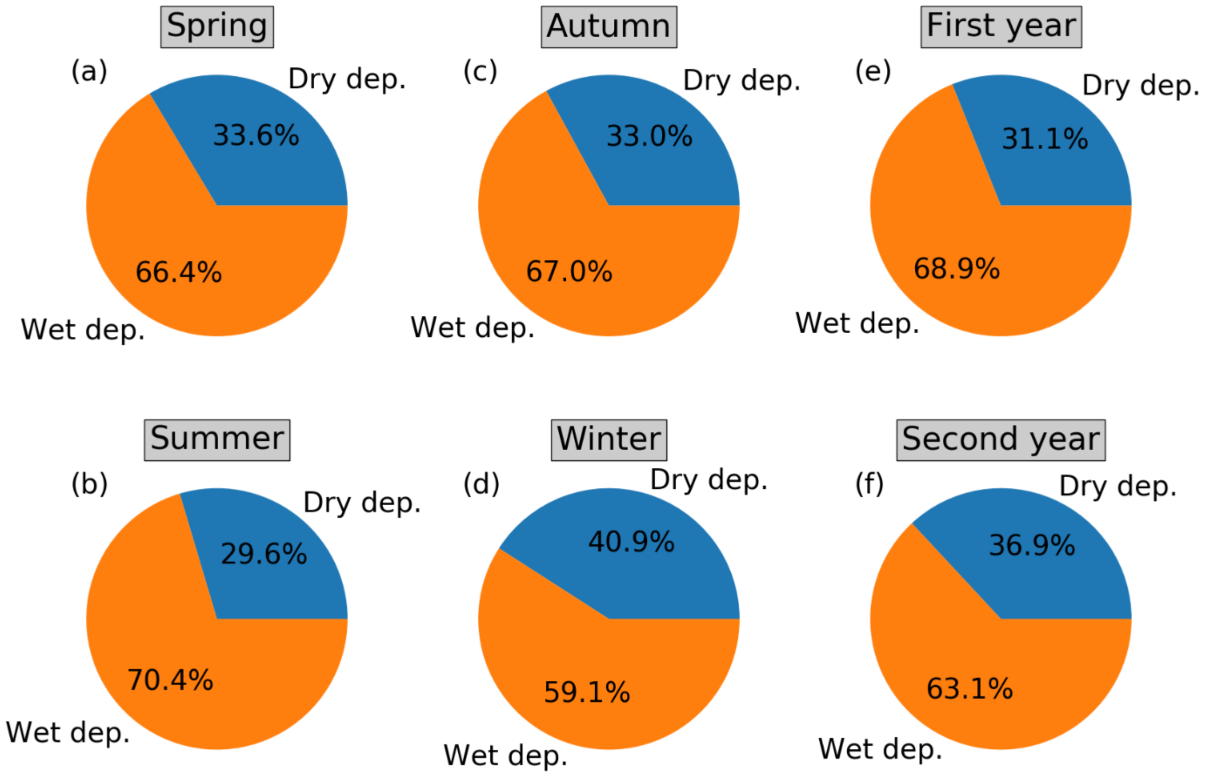

Figure 1 Contribution of dry and wet deposition to total deposition for each season and both measurement years labeled from (a) to (f).

In case of seasonal contributions, dry deposition contributed approximately one third to total deposition except for winter. In the second year, contribution of dry deposition was higher than in the first year. Higher fractions of dry deposition were related to the large dry deposition occurring in late February 2018. Table 2 shows seasonal and yearly deposition sums of dry and wet deposition measurements. Please note that the sum of all seasons corresponds to the sum of both measurement years.

Table 1 Annual and seasonal sums of dry deposition estimates (DD) and NH$_4^+$-N, NO$_3^-$-N, dissolved organic nitrogen (DON), and the resulting total wet deposition (TWD) from wet deposition samplers (bulk (BD) and wet-only (WD)) in kg N ha^{-1} period^{-1}. Uncertainties in dry deposition estimates are related to applied gap-filling technique.

Time	DD [kg N ha^{-1} period^{-1}]	WD [kg N ha^{-1} period^{-1}]				BD [kg N ha^{-1} period^{-1}]			
		NO$_3^-$-N	NH$_4^+$-N	DON	TWD	NO$_3^-$-N	NH$_4^+$-N	DON	TWD
Winter	2.0	1.5	0.9	0.4	2.8	1.7	1.3	0.5	3.5
Spring	2.2	1.8	2.3	0.1	4.2	1.9	2.4	0.1	4.4
Summer	2.0	1.9	2.6	0.2	4.7	1.6	2.2	0.6	4.4
Autumn	1.7	1.5	1.4	0.6	3.5	1.4	1.4	0.6	3.4
June 16 – May 17	3.8	3.8	4.2	0.4	8.4	3.5	4.2	1.0	8.7
June 17 – May 18	4.0	2.9	3.1	0.9	6.9	3.0	3.1	0.9	7.0

Small seasonal and annual differences in dry deposition were determined (approx. 200 g N ha^{-1} period^{-1}). Total seasonal and annual uncertainties related to gap-filling (Eq. (3)) were between 7 and 21 g N ha^{-1} period^{-1}. Due to the large fluxes in late February 2018, dry deposition and its uncertainty were remarkably high during winter. Total wet deposition (TWD) was highest in spring and summer. During those periods, NH_4^+-N contributed most to TWD which was probably related to high NH_3 concentrations. Interseasonal differences for NO_3^--N were found but were lower compared to changes in NH_4^+-N. DON deposition was lowest and was between 0.1 and 0.6 kg N ha^{-1} a^{-1}. Overall, differences in TWD for both sampler types were less than 300 g N ha^{-1} a^{-1} except for winter.

We added Fig. 1 to the Supplemental Material, replaced Table 2 of the manuscript by Table 1 of the response, and rephrased the description beginning at line 500.

Comment R1.15: *Line 507: "In total, we got a total nitrogen deposition of…" Please consider changing to "Total wet + dry deposition was equivalent to…".*

Response to R1.15: Done.

Comment R1.16: *Line 510: Change the section title to reflect the structure of the section, which is ordered as concentrations, fluxes, then deposition velocity.*

Response to R1.16: Agreed.

Comment R1.17: *Line 523: Please consider using a word other than "neutral" in this sentence, as neutral is commonly used to characterize atmospheric stability and is confusing as currently used.*

Comment R1.17: Supposedly, you refer to line 533. We replaced "closer to neutral conditions" by "closer to zero".

Comment R1.18: *Line 547: The explanation of high deposition in Feb 2018 being driven by NH4+ is not convincing (i.e., high SO2 corresponding to high ammonium sulfate/bisulfate concentrations). In other winter periods, NH4+ makes a relatively small contribution to Nr (Figure 3). Additionally, given the lower deposition velocity of particles a very large concentration of NH4+ would be needed to explain this much larger deposition flux. Such a large increase in NH4+ should have a regional signal. Can data from other monitoring sites be used to assess larger scale patterns in atmospheric chemistry during this period that could shed light on what could be driving the large increase in Nr at the authors study site? Some additional analysis may be possible here.*

Response to R1.18: After carefully reflecting the reviewer's comments to that aspect, we agree the explanation of the deposition in February 2018 is insufficient. For January 2017 (only NH_3) December 2017, March 2018, and April 2018 no DELTA measurements were available since the denuder pump was not working properly. For those months, averages from previous years were used to fill the gaps. Actually, we had measured delta concentrations for February 2018. Thus, Figures 2, 3, S1 and their corresponding descriptions were adjusted. We searched for air pollution stations, which are in close proximity to measurement station and are exposed to a similar pollution climate. Our measurement site is located in a remote area and represents a rural background station in the air pollution network hosted by the German Environment Agency. However, there is no nearby measurement site in a radius of 50 km representing rural background and conducting NH_4^+ measurements in the network. In 20 km distance, a station integrated in the Czech Air pollution network is measuring NH_4^+. Unfortunately, they provide no measurements of NH_4^+ during February 2018.

"During the exposure period of the DELTA samplers, we found 0.96, 0.17, 0.37, 0.27, and 1.70 µg N m^{-3} for NH_4^+, NH_3, NO_3^-, HNO_3, and NO_x, respectively. The aerosol concentrations were exceptionally large in February 2018, which have affected these averages considerably. Average NH_4^+ concentration during winter excluding February 2018 was only 0.38 µg N m^{-3} in comparison to 0.96 µg N m^{-3} for February 2018. The concentration in this month results in a NH_4^+ concentration 2.5 times higher than the average. Also, SO_2 was much larger concentrations (1.54 µg m^{-3}) in this month compared to the other winter month (0.37 µg m^{-3}). Figure 2 shows the relative contributions of each N_r compound for February 2018 compared to averaged fractions during winter excluding February 2018.

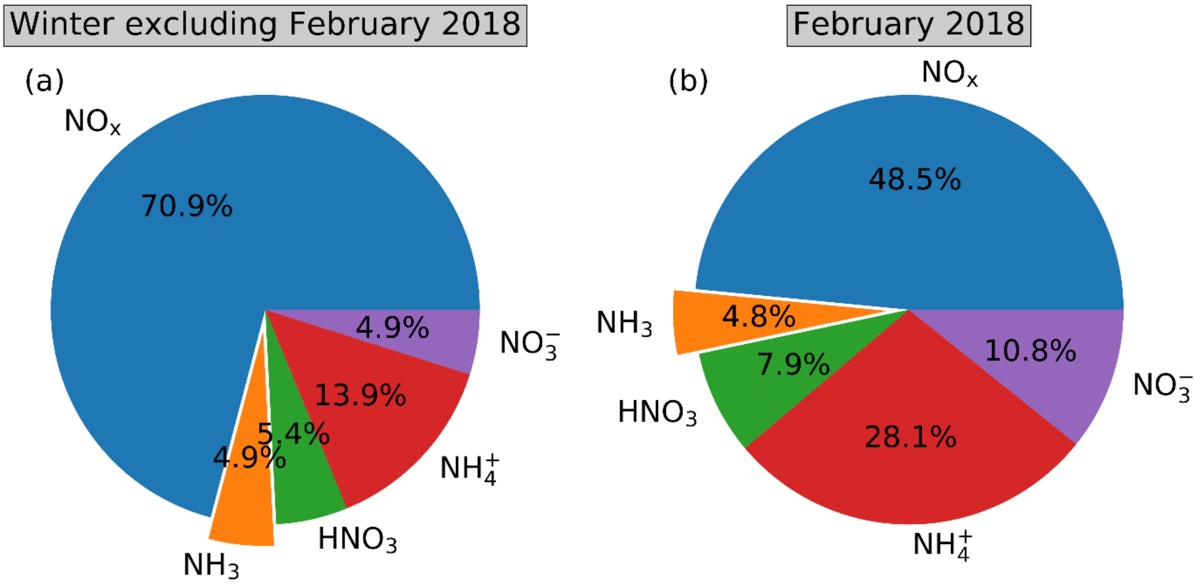

Figure 2 Relative contribution of concentrations for NO_x, NH_3, HNO_3, NO_3^-, and NH_4^+ to ΣN_r estimated from DELTA and NO_x measurements for winter and separately for February 2018. NO_x measurements are averaged to exposure periods of the DELTA samplers.

During February 2018, NH_4^+ made a significant contribution to the ΣN_r concentration. The measured NH_4^+ value is an integrated value over approximately one month. Thus, daily contributions of NH_4^+ could have been even higher. Earlier studies by e.g. Wolff et al. (2010) report events with large aerosol deposition. During their campaign, wind speeds were relatively high. Largest aerosol deposition occurred during dry conditions, e.g. low *RH*, no rain, and high visibility. Figure 3 shows micrometeorological parameters, deposition velocities, and gap-filled ΣN_r fluxes from the 12 February to 6 March. Large deposition fluxes were accompanied by high *wsp* (wind speed) and u_* values, high R_g indicating high visibility, and low *RH*. The observed conditions are typical for cold air streams with high aerosol loads coming from North east and led to a reduction in turbulent resistances resulting in a high v_d, which is allowed by turbulence. Hence, at low concentrations of NH_4^+ significant aerosol deposition is possible if R_a and R_b are reduced.

In conclusion, NH_4^+ aerosols, ammonium sulfate and nitrate, were most responsible for ΣN_r deposition due to their excess over NO_3^-. Since we had no high-resolution flux measurements of any ΣN_r compound during that time, we have no evidence which aerosol predominated the ΣN_r flux. "

We rephrased the discussion about the deposition event from lines 548 to 565 by this response. Figure 2 was implemented in Sec 4.1 and Figure 3 was added to the Supplemental Material.

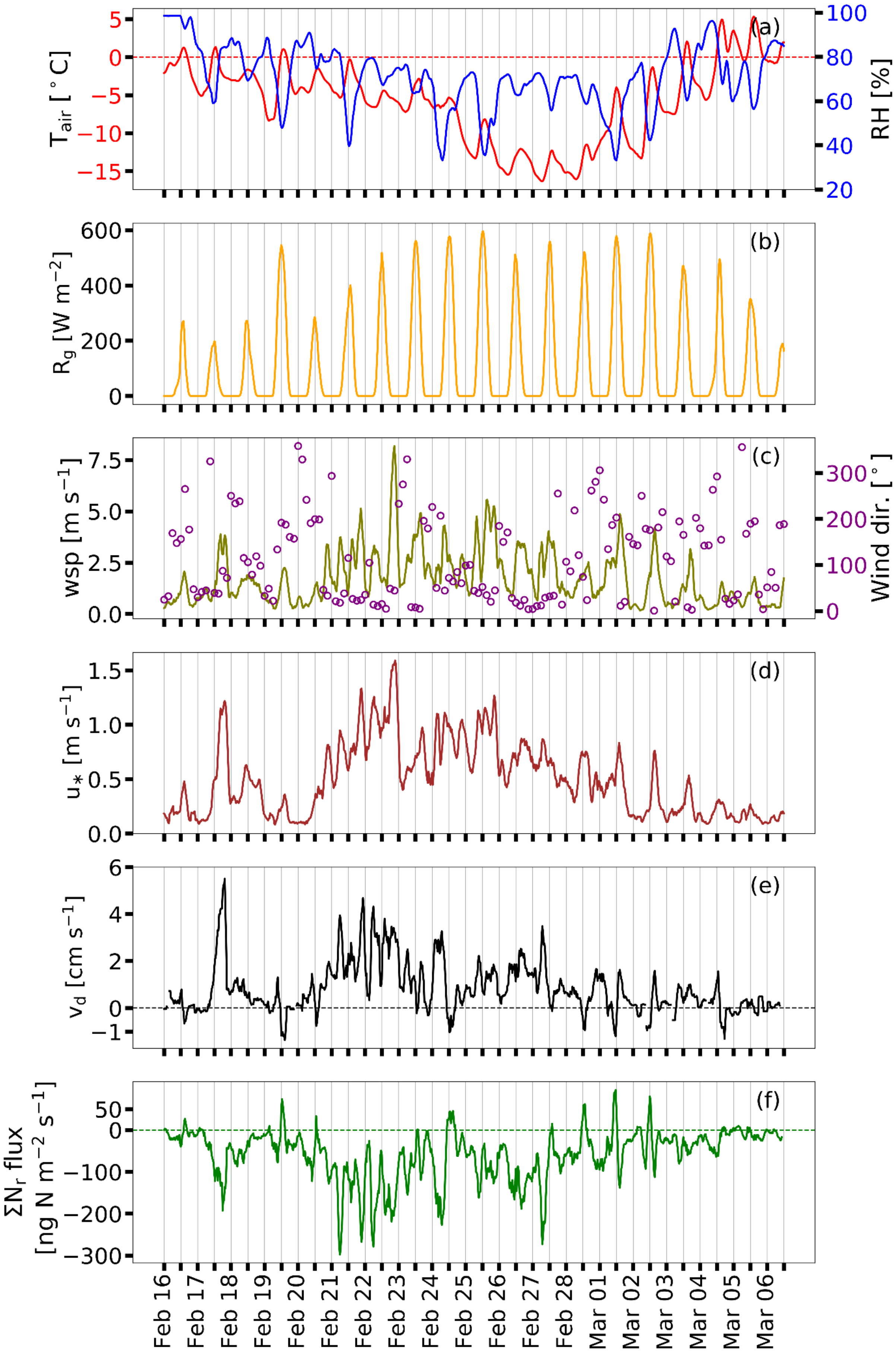
(a)
T_{air} [°C]
5
0
−5
−10
−15
RH [%]
100
80
60
40
20
(b)
R_g [W m^{-2}]
600
400
200
0
(c)
wsp [m s^{-1}]
7.5
5.0
2.5
0.0
Wind dir. [°]
300
200
100
0
(d)
u_* [m s^{-1}]
1.5
1.0
0.5
0.0
(e)
v_d [cm s^{-1}]
6
4
2
0
−1
(f)
ΣN_r flux [ng N m^{-2} s^{-1}]
50
0
−100
−200
−300
Feb 16
Feb 17
Feb 18
Feb 19
Feb 20
Feb 21
Feb 22
Feb 23
Feb 24
Feb 25
Feb 26
Feb 27
Feb 28
Mar 01
Mar 02
Mar 03
Mar 04
Mar 05
Mar 06

Figure 3 Recorded air temperature (T_{air}.), relative humidity (RH), global radiation (R_g), wind speed (*wsp*), friction velocity (u_*), v_d, and gap-filled ΣN_r flux as 3-h running mean from 16 February to 6 March 2018. Wind direction corresponds to values measured at 3-h time stamps.

Comment R1.19: *Line 578: Change "been happened" to "been occurring".*

Response to R1.19: Agreed.

Comment R1.20: *Line 599: Section 4.1 is structured as a summary of concentrations, fluxes, then deposition velocity. The paragraph beginning on line 599 describes concentrations and should be moved to follow (or integrated with) the first paragraph in the section (beginning at line 511).*

Comment R1.20: We agree. We integrated the corresponding lines 599-616 to the first paragraph of Section 4.1 (line 527).

Comment R1.21: *Line 614: Regarding the DELTA, the authors include these statements: "Concentration peaks could not be collected sufficiently by the coated surfaces. The latter are exposed to environmental influences like temperature and moisture, and their sensitivity may reduce over time." The authors appear to be saying that the DELTA measurements are biased low because short term peaks in concentration are under sampled (inefficiently collected by denuder coatings) and due to loss of denuder collection efficiency over time. I have not read this previously about the DELTA denuders. The authors need to support these statements with evidence or remove them.*

Response to R1.21: In this paragraph, we mentioned possible reasons for differences between denuder and TRANC measurements. "In addition, higher oxidized compounds like N_2O_5 or peroxy acetyl nitrates could not be collected by DELTA, but probably converted by the TRANC. Issues in the temperature stability or CO supply leading to instabilities in the conversion efficiency of the TRANC may be responsible for disagreements to the collection efficiency of the denuders. The difference between TRANC and DELTA concentrations is also related to their aerosol cut-off sizes (R2.13 and R2.18). A key uncertainty was the data coverage of the TRANC, which was at 78% on average during the exposure periods."

Still, the coating of denuders can be washed off if water is sucked in by the pump. In order to avoid that, we mounted a funnel at the opening of the denuder. During the measurement campaign, no water was found in the denuders. Thus, a loss of collection efficiency due to water can be excluded. Following the Reviewers suggestion, we decided to delete these lines (614-616) and added the information given in this response to that line 614. Please note that the discussion was shifted to line 527.

Comment R1.22: *Line 631: Change "not considered" to "not be considered".*

Response to R1.22: Done.

Comment R1.23: *Line 639: "Consequently, a second mechanism, the stomatal compensation point firstly proposed by Farquhar et al. (1980) likely controls the uptake of the Nr compounds." This statement is false as written. The stomatal compensation point does not control the exchange of all Nr compounds. The authors should be specific about which Nr compounds they are referring to here (i.e., NO2 and NH3?). If the authors are suggesting a compensation point driven exchange of NO2 then an appropriate citation is needed.*

Response to R1.23: We agree that the stomatal compensation point is not responsible for all N_r compounds and further clarification is needed. Zöll et al. (2019) examined ΣN_r fluxes at the same site during summer and found a high contribution of NH_3 to the ΣN_r concentration. Since v_d of NH_3 is higher than NO_2 above forest, the saturation point in the light response curve of ΣN_r is probably caused by the stomatal compensation point of NH_3.

Please note that line 639 was deleted.

Comment R1.24: *Line 652: "Micrometeorological parameters were controlled by natural processes." This statement is unnecessary.*

Response to R1.24: Agreed. We deleted the sentence.

Comment R1.25: *Line 709: A paragraph should not begin with "However".*

Response to R1.25: Agreed.

Comment R1.26: *Line 803: "It shows that deposition of sedimenting organic and inorganic particles is not relevant at the site." What does this statement mean? It is unclear as written.*

Response to R1.26: We found that differences in wet deposition estimated from bulk and wet-only samplers were negligible. Small differences could be induced by the sedimentation of organic and inorganic dusts or by the dry deposition of gases like NH_3 or HNO_3 (Staelens et al. 2005). We rephrased the statement according to this response.

Response to Referee 2

General Comment: *The separation of the paper into a paper mainly reporting the measurements and a paper on modelling of the exchange has made this paper more accessible. However, the revised paper now covers much of the same ground in the discussion as the paper by Zoll et al. (2019). I found it rather disingenuous that the paper does not point out right at the beginning that it presents a continuation of the time-series of Zoll et al. (2019), taken at the same site, which is not spelt out until line 651. There is little point in re-iterating the findings of Zoll et al. to the extent done here and much of the Discussions section is rather speculative. I suggest that the analysis and discussion gets more focussed on the presentation of the annual budgets, cutting the paper by about 1/3. This is particularly the case as I do not believe that either the present paper nor the analysis by Zoll et al. provides insights into the mechanistic controls. Whilst the neutral network analysis (and the correlation analysis of the present papers) can identify associations, they cannot identify causality.*

We wrote in the introduction that Zöll et al. (2019) conducted flux measurement of ΣN_r with the same instrumentation at the measurement site. They selected a short time period from the 2.5 measurement campaign (14 July to 30 September 2016) for identifying links ΣN_r between CO_2 using artificial neural network approach. In addition, their post-processing and quality-selection was different. Thus, the term continuation may be misleading. Still, we agree that arguments in Sec 4.2 are similar to Zöll et al. (2019) and the subsequent discussion on canopy resistances, stomatal, and non-stomatal deposition is rather speculative. Thus, we shortened Sec. 4.2 substantially according to your comments.

We agree that the analysis of mechanistic controls is done for individual compounds and compared to results of resistance models. Based on the reviewers' suggestions, we decided to remove the discussion about the resistance analysis, integrated your suggestions to the dependence of deposition velocities on their concentrations to the manuscript, and shifted the focus of this manuscript on the determination of annual budgets.

We addressed all of your remarks and implemented your suggestions to scientific and technical issues in the manuscript.

Major comments

Comment R2.1: *At several places (starting with the opening two sentences and final sentence of the abstract) the paper overstates the utility of the Nr measurements for the development of model improvements / parameterisations. Because it is unknown which Nr compounds dominate the flux during any particular 30-minute flux period, any parameterisation of the net exchange is not transferable to other situations subject to another compound mix. I am not saying the analysis of the Vd and Rc of the net exchange is not worth doing but the paper should point out more clearly (e.g. lines 60-66) that the main utility of the TRANC is to quantify net dry deposition inputs of Nr with a single instrument (rather than a suite of instruments for each compound individually) and that the analysis of the net exchange parameter is a by-product.*

Response to R2.1: We agree that measuring only the ΣN_r exchange provides no information about the actual flux contribution of each compound – a main drawback of the TRANC. Thus, a parametrization of the net exchange is not very meaningful. However, a comparison of modeled and measured deposition velocities of ΣN_r could hint on deficits in deposition modeling. For example, it is possible to compare Fig 9. to ΣN_r deposition velocities determined with inferential modeling. Thus, the analysis of the net exchange should be a point of interest but treated as an additional outcome. We agree that the corresponding lines in the abstract provide a false impression about the utility of TRANC measurements.

We added the main utility of the TRANC, which is the determination of the annual dry deposition and temporal dynamics of ΣN_r with one instrument, to lines 60-66 and rephrased corresponding lines in the abstract (lines 1-3 and 32-33). We further focused the evaluation of the TRANC measurements on annual budgets and removed the discussion on effective canopy resistances ($R_{c,eff}$). The latter was removed since the quasi-laminar resistance (R_b) is not known (see R2.4). Still, we found that v_d was influenced by micrometeorological parameters but not driven by the overall concentration (see R2.3).

Comment R2.2. *As mentioned above, the paper does not sufficiently clearly distinguish between associations and correlations on the one hand and causes / drivers on the other. Radiation, turbulence and temperature (and sensible heat flux) are highly correlated with each other (Figure S7) and it is impossible to decide which is the mechanistic driver and I am not convinced that vd is controlled by the plant activity rather than u* (Line 435-436) and other drivers that correlate with Rg. From what we know of the exchange of the individual Nr compounds, stomatal exchange will be important for NO2 and its importance is highly variable for NH3 as pointed out in the paper. However, it is not so important for HNO3 and NH4+/NO3-. However, Rg will change Nr composition over the day with HNO3, a particularly fast depositing compound, typically peaking at midday (again related to Rg, this time via photochemical production), and it will produce a diurnal pattern on the effect of NH4NO3 volatilisation, which deposits fast only during daytime when temperature gradients are large. I therefore can't see that the measurements prove that stomatal conductance is the main controller of the Nr flux (Section 4.2.1). Stating that u* does not affect the flux would be saying that Ra and Rb do not exist. If this analysis were done on Rc or 1/Rc at least the influence of turbulence would have been removed.*

Response to R2.2. We agree that the differentiation in associations and correlations was not done clearly. We totally agree that it is impossible to state global radiation, turbulence or temperature as the mechanistic driver of the ΣN_r flux due to their high correlation with each other. We carefully checked the manuscript for corresponding cases and corrected them. We further agree that the role of u_* as a control for v_d and the flux has to be rephrased. The analysis of v_d vs concentration showed that ΣN_r concentrations did not correlate with their v_d (see R2.3). We corrected the corresponding lines about plant activity as control (line 435-436). From an analysis of the net flux, we cannot examine stomatal or non-stomatal controls since fluxes of individual compounds are not known and highly variable during the day. Thus, we removed sections (Sec. 4.2.1) and corresponding lines about stomatal control and non-stomatal controls (lines 687-747) on the ΣN_r flux. As written in R2.1 and R2.4, the discussion about $R_{c,eff}$ was removed.

Comment to R2.3: *In this context I do not find the analysis of the controlling factors for the flux very helpful. The flux would be expected to be affected by u* (Fig. 7, Table 1 and associated text) as it still* *contains the control via Ra and Rb. Also, the authors seem to try to convey that the slope changes with u*. Except for the possibility of a non-zero intercept, the slope is actually vd. Thus, the authors should either plot vd vs Nr concentration or the ratio Vd/u* against Nr concentration. They might also want to consider binning data according to y-values rather than showing raw data to convey a clearer message. This has implications for the discussion in Section 4.2.1. I fail to see why the lack of correlation between flux and concentration within a u* class suggests that u* is not a driver (line 645ff). Surely, it would suggest that the concentration is not the driver.*

Response to R2.3: We highly appreciate the Reviewers' suggestions to Fig. 7. Actually, the plot flux vs concentration was requested by Reviewer 1 during the first review round. Still, we note that the conclusions drawn from Fig. 7 were incorrect and we agree that a plot of v_d vs concentration or the ratio v_d/u_* vs concentration surely shows if the concentration is a driver of the ΣN_r exchange. We removed lines 417-430 and replaced them by the text and figures given below. Figures 4 and 6 were provided as supplemental material. Figure 5 was added to the manuscript.

"In order to investigate the influence of u_* on the ΣN_r exchange, Fig. 4 illustrates the dependency of v_d on u_* for deposition and emission fluxes during day and night. The R_g threshold for day and nighttime fluxes was set to 10 W m^{-2}. For better visibility, we binned data in 0.1 m s^{-1} increments of u_*. Since bins are not equal in size, we added corresponding half-hourly fluxes to the plots. Red dots represent averages of each bin and error bars correspond to their standard error.

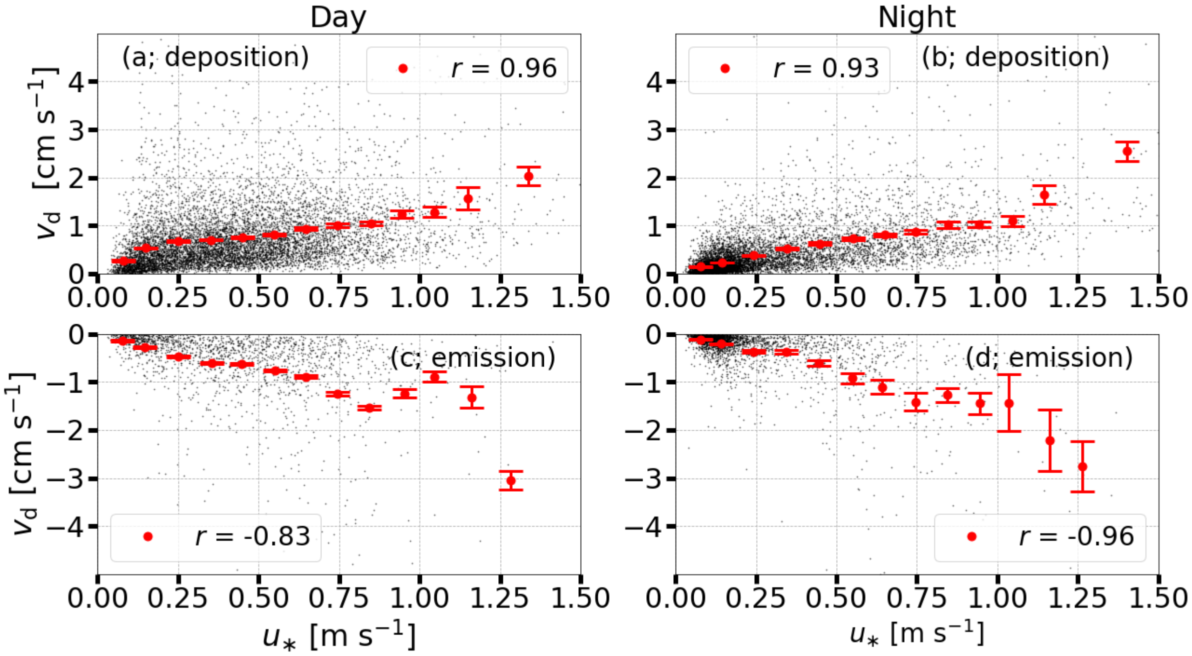

Figure 4 Relationships between measured u_* and corresponding ΣN_r v_d separated in emission and deposition during day ((a) and (c)) and night ((b) and (d)). Half-hourly data is displayed in black, red dots represents averages binned in increments of 0.1 m s^{-1}. Error bars indicate the standard error of the averages. The threshold for identifying day and nighttime v_d was set to 10 W m^{-2}. r represents the measure of correlation evaluated for the binned data.

We found that v_d increased slightly with u_* due to dependency of v_d on R_a and R_b. The latter are proportional to the inverse of u_* suggesting that the increase with u_* should follow a power law. In case of particles, linear relationships between u_* and v_d were found by Gallagher et al. (1997); Lavi et al. (2013); Donateo and Contini (2014). A relationship between v_d and u_* seems to exist as suggested by the correlations (r), but no clear functional relationship could be identified due to the large scattering of half-hourly v_d.

For visualizing the impact of concentration on v_d (Fig. 5), we plotted ΣN_r concentration against the ratio v_d/u_* in order to reduce the influence of R_a and R_b on v_d. The threshold for R_g was set to 10 W m^{-2}, and we binned data in 0.5 µg N m^{-3} increments of ΣN_r concentration.

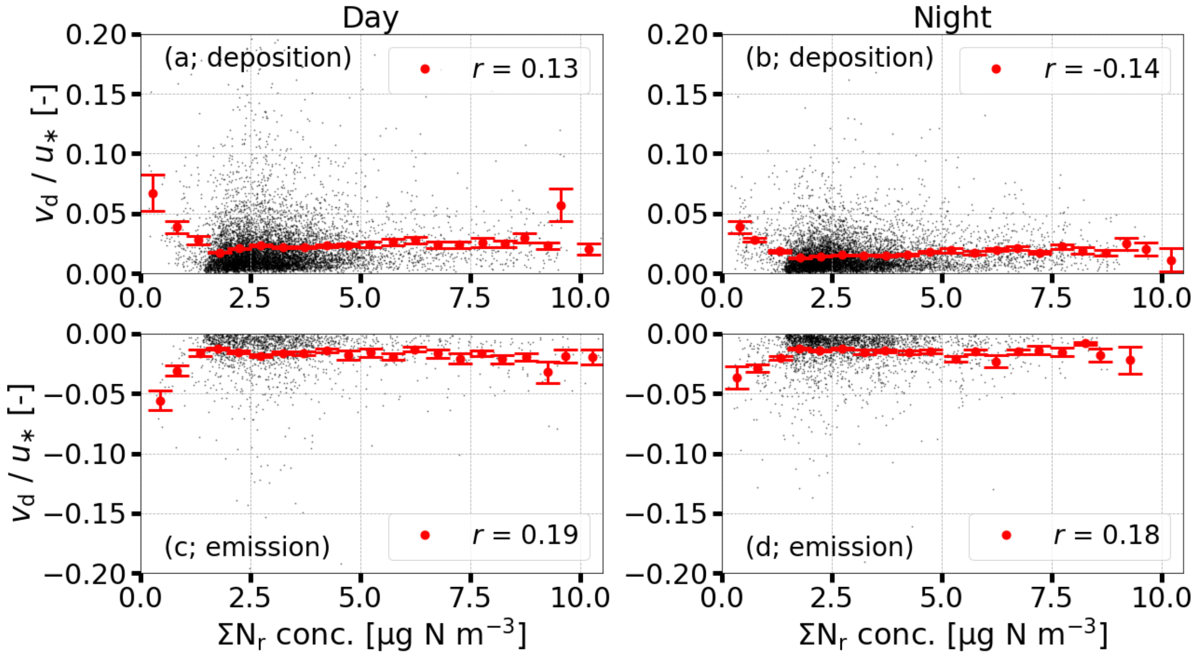

Figure 5 Relationships between measured ΣN_r concentrations and corresponding ratios v_d/u_* separated in emission and deposition during day ((a) and (c)) and night ((b) and (d)). Half-hourly data is displayed in black, red dots represents averages binned in increments of 0.5 µg N m^{-3}. Error bars indicate the standard error of the averages. The threshold for identifying day and nighttime v_d was set to 10 W m^{-2}. r represents the measure of correlation evaluated for the binned data.

It is obvious that v_d/u_* exhibited no significant dependence on ΣN_r concentration as shown by the low values for r. The ratio appeared to be constant across the (entire) concentration range. It demonstrates that ΣN_r concentration had no significant influence on their v_d. In case of particles, the ratio v_d/u_* depends on Obukov-Length (L) and particle size according to Gallagher et al. (1997) and Lavi et al. (2013). In case of deposition fluxes measured during daytime, we found that the ratio decreased for $-0.2 > L^{-1} < 0$ up to a minimum if L^{-1} reaches zero (neutral stratification) (Fig. 6).

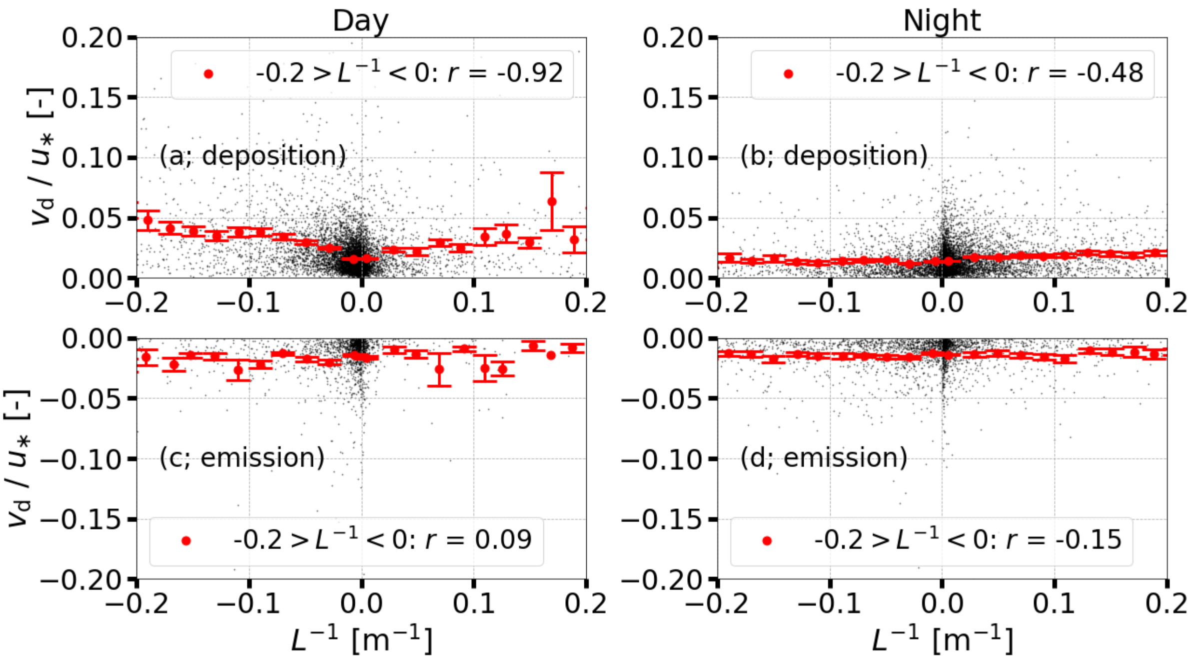

Figure 6 Relationships between L^{-1} and corresponding ratios v_d/u_* separated in emission and deposition during day ((a) and (c)) and night ((b) and (d)). Half-hourly data is displayed in black, red dots represents averages binned in increments of 0.02 m^{-1}. Error bars indicate the standard error of the averages. The threshold for identifying day and nighttime v_d was set to 10 W m^{-2}. *r* represents the measure of correlation evaluated for the binned data.

This relationship was observed by Gallagher et al. (1997) and Lavi et al. (2013). Although the scattering of half-hourly ratio is large, the decrease of the ratio with increasing L^{-1} as well as the dependence of v_d on u_* demonstrate that v_d had a higher affinity to micrometeorological parameters than to the ΣN_r concentration.

From the analysis of the figures 4, 5, and 6, it is impossible to state u_* or *L* as the controlling variable of the ΣN_r exchange since turbulence, stratification, R_g, sensible heat flux, air temperature, and relative humidity are highly correlated with each other" as visualized by Fig S7. Thus, the dependence on u_* could also be related to effects of the sensible heat flux, *T*, or R_g and it is impossible to decide which is the mechanistic micrometeorological driver of the ΣN_r flux.

According to these results, we removed the entire discussion of Sec 4.2.

Comment R2.4. *Although it theoretically provides more insights, the Rc analysis is quite uncertain due to the calculation of Rb. On the one hand the authors attempt to calculate an Rb that is weighted by the different compounds, on the other hand they set Rb for particles to 0 so that their full interaction with the canopy enters Rc. This is a crude approximation because the authors only have long-term information on composition (rather than half-hourly) and, numerically, the weighting should be done according to the compound contribution to the flux rather than the concentration. Moreover, the statement that particles are not subject to an Rb term (line 285) is incorrect. Rb describes the resistance posed by the laminar sublayer resistance and this is in fact larger for particles than it is for gases. However, the concept behind the terminology of Rb is that of Brownian diffusion, whilst particles have other mechanisms (interception, impaction, gravitation settling) to overcome this boundary layer in addition to diffusion (which is very ineffective for all but the smallest particles). Thus, for particles, the concept of Rb is usually replaced with that of Vds = Vd(z0) = 1/(1/Vd-Ra). The current approach followed* *in the paper therefore derives an Rc that is a combination of different elements that mean different things for different compounds. This highlights again the limitations of the total Nr flux for mechanistic analysis (point 1 above).*

Response to R2.4: We thank the Reviewer for his/her remarks to the implementation of R_b in the resistance analysis. Based on the analysis of the DELTA denuders, we set R_b for particles to zero since generally their contribution to ΣN_r concentrations was relatively low compared to gases. We agree that setting R_b to zero for particles is not very meaningful based on observations on aerosol deposition and its implementation in current resistance models. We agree that the weighting should be done for the contribution of each compound to the ΣN_r flux. Due to these limitations, we decided to remove the entire analysis and discussion on R_c (Sec 4.2.1 to Sec. 4.2.3) and related paragraphs in the introduction (lines 66-91) and theoretical background (lines 272-320). As written in R2.1, the analysis of the net exchange of ΣN_r is an additional product. Therefore, we shifted the focus towards the annual budgets and shortened the discussion about micrometeorological influences on v_d.

Comment R2.5. *It is similarly incorrect that the flux pattern of "nitrogen aerosols … is driven by Ra" (line 317). In fact vd for particles tends to be more reduced compared with 1/Ra than that the vd of gases.*

Response to R2.5: We agree that the statement is incorrect since it ignores the contribution of the surface to v_d of aerosols. We deleted the sentence.

Other scientific comments

Comment R2.6: *Throughout the manuscript it is not clear to me which results/ figures are based on u* filtered data and which not. Please clarify throughout. For deposition u* filtering introduces a bias to the remaining data, removing preferentially small fluxes.*

Response to R2.6: In line 231, we wrote that a u_*-filter was applied to the measured fluxes. Thus, figures 5, 6, 8, 9, S5, S6, S9, S10, S12, S13, and associated text are based on u_* filtered data. In Section 3.3, we discussed the effect of a turbulence filter on nitrogen dry deposition estimates. In the first review round, it was mentioned that a u_*-filter will remove preferentially small fluxes. We calculated annual dry deposition for two flux data sets with and without u_*-filter. On both datasets, the Mean-Diurnal-Variation (MDV) technique was applied as gap-filling approach. As visualized in Fig. 10, the effect of the u_*-filter is present but within the uncertainty range of the gap-filled fluxes. The comparison of different MDV approaches was done for both flux data sets as written in the caption of Fig. 11. We clarified the description of the flux filtering (line 233 and 251).

Comment R2.7: *I cannot fully follow the alternative implementation of the MDV in which you consider temperature, humidity and precipitation (lines 485ff). The introduction of the approach is not very clear and should probably be moved the methods section anyway. Could you please go through the English. "Dry deposition without restriction" (line 485) is not very meaningful. You probably mean "agreed within +/- 3C" rather than "varied by". It would probably make the section more readable if you gave this implementation a name. What about "stratified MDV" or "conditional MDV". Overall, I wonder* *whether it would make more sense to apply the MDV gap filling to vd rather than fluxes as it is the exchange mechanism that is impacted by the meteorology rather than the concentration. Clearly, this would only work for periods for which you have concentration data.*

Response to R2.7: We agree that the description of the alternative MDV approach needs to be clarified. Yes, the wording "agreed within +/- 3°C" is meant here. In the revised version, we moved the description to the method section (line 251), improved its readability, and entitled the MDV approach with additional micrometeorological criteria as "conditional MDV" (CMDV) according to your suggestion.

Since we had substantial gaps of different sizes in the ΣN_r concentration time series, an application of the MDV method to v_d seems to be less useful.

Comment R2.8: *Line 17ff and line 826. Ra and Rb do not make a contribution to vd, but to Rt=1/vd. Alternative reword to say that Ra and Rb make a negligible contribution to limiting vd.*

Response to R2.8: We agree. Please note that the discussion related to Figure 9 was deleted. Thus, we removed corresponding sentences in the abstract and conclusion.

Comment R2.9: *It is well established that closed path sensors lead to a dampening of the fluctuations and thus the fluctuations induce the artificial flux due to quantum mechanical quenching are reduced compared with the true latent heat flux and as a result Eq. (1) will overestimate the correction by analogy to the impact on the density correction (e.g. Ibrom et al., 2007). Because the relative correction is small this is not a major issue, but the authors should acknowledge the uncertainty and clarify that the correction is an upper estimate.*

Response to R2.9: We agree. "Since we measured H_2O fluxes with an open-path system and used them for correcting ΣN_r fluxes, density corrections following the Webb-Pearman-Leuning correction for H_2O fluxes measured with closed-path systems (Ibrom et al. 2007) were not accounted for. The impact on the correction is likely small, but the determined interference flux correction should be seen as an upper estimate." We added these details to line 229.

Comment R2.10: *The paper incorrectly states that the aerosol detected by the TRANC is NH4NO3 (line 46; line 144). In fact it detects the sum of NH4+ and NO3-, with the former also representing ammonium sulfates and the latter also sodium and calcium nitrate. Figure 4 very clearly demonstrates the presence of excess NH4+ over NO3- at this site.*

Response to R2.10: We replaced "particulate ammonium nitrate (NH_4NO_3)" by particulate ammonium (NH_4^+) and nitrate (NO_3^-) (line 46). Yes, different NH_4^+ and NO_3^- aerosols are converted by the TRANC. We rephrased line 144 as follows: [..] "leading to a split up of NH_4^+ and NO_3^- aerosols such as ammonium sulfate, ammonium nitrate, sodium and calcium nitrate into their subcomponents. In case of NH_4NO_3, it is thermally converted to NH_3 and HNO_3 (Marx et al., 2012)".

Comment R2.11: *The review of previous studies (lines 45 to 91) is incomplete and inconsistent. Firstly, it is worth mentioning that other micrometeorological methods do exist, beyond EC. In fact the references in line 69 refer partly to flux gradient measurements although the paragraph starts with "Prior EC studies of …". Secondly, there are not as few flux studies of Nr compounds to remote sites as stated. I could probably easily list 30, but many only cover short campaign periods. I therefore suggest starting the sentence in Line 67 with "Only a few long-term studies have been conducted to derive annual inputs at remote locations." and then focus on listing the long-term studies which can be done more exhaustively. This is also consistent with the true benefit of the TRANC system and this dataset as outlined above.*

Response to R2.11: We appreciate your suggestion to the introduction. Yes, there are other micrometeorological methods for estimating biosphere-atmosphere exchange of reactive nitrogen, for example the flux gradient method. Here, the focus is on eddy-covariance since it is the "common method for estimating greenhouse gas fluxes (Aubinet et al., 1999; Baldocchi, 2003) in flux monitoring networks (FLUXNET (Baldocchi et al., 2001), ICOS (Heiskanen et al., 2021)) and also suitable for reactive nitrogen compounds" as shown by the listed references. "However, the EC method requires fast-response analyzers." These sentences were added to line 48.

From line 45 to 59, we wanted to show that eddy covariance method has been applied to several N_r compounds, but a simultaneous operation of individual devices using EC is challenging. We focused on listing EC studies in these lines since the coupling of the TRANC to a fast-response detector for NO allows the application of the EC method for total reactive nitrogen.

We appreciate your suggestions to the second paragraph since it strengthens the significance of TRANC measurements for the derivation of annual nitrogen dry deposition. We rephrased lines 67-91 as follows:

"Only a few long-term studies have been conducted to derive annual inputs with micrometeorological methods at (remote) forest ecosystems. Munger et al. (1996) conducted EC measurements of NO_y, which refers to the sum of all oxidized N_r compounds, e.g., NO, NO_2, HNO_3, dinitrogen pentoxide (N_2O_5), peroxyacyl nitrates (PAN), aerosol nitrates, above a mixed deciduous forest for five years. Averaged NO_x concentrations were at 0.62 and 4.26 ppb (0.36 and 2.44 µg N m^{-3}) during summer and winter, respectively, if wind was blowing from Northwest. During southwesterly winds, mean NO_x concentrations were 1.25 and 9.48 ppb (0.72 and 5.43 µg N m^{-3}) during summer and winter, respectively, indicating a varying pollution climate. The authors reported an annual net dry deposition of NO_y covering 1990 to 1994 of 2.49 kg N ha^{-1} a^{-1}. Munger et al. (1998) reported an annual reactive N deposition of wet + dry deposition measurements of 6.4 kg N ha^{-1} a^{-1} for the period 1990 to 1996 at the same site. Dry deposition of NO_y contributed 34% to total deposition. Wet deposition of NH_4^+ was comparatively low estimated to 1.1 kg N ha^{-1} a^{-1}.

Neiryck et al. (2007) and Erisman et al. (1996) conducted GM measurements in order to estimate dry deposition of NO_x and NH_3. Neiryck et al. (2007) published GM measurements from July 1999 to November 2001 above mixed coniferous/deciduous forest, which was in close proximity of a highway and the city of Antwerp leading to mean NO_2 and NH_3 concentrations of 8.7 and 3.0 µg N m^{-3}, respectively. The authors determined an annual NH_3 dry deposition of 19.6 kg N ha^{-1} a^{-1} and NO_x emission of 2.7 kg N ha^{-1} a^{-1}. NO_x emissions were probably related to a strong contribution of soil-emitted NO. Erisman et al. (1996) reported NO_x and NH_3 fluxes above a Douglas Fir stand of 2.5 ha surrounded by a larger forested area of 50 km^2 for 1995. Mean NH_3 concentration was 4.5 µg N m^{-3}

possibly related to livestock farming in the surroundings of the site. They estimated annual dry depositions of 17.9 kg N ha^{-1} a^{-1} and 2.8 kg N ha^{-1} a^{-1} for NH_3 and NO_x, respectively.

These were the few micrometeorological measurements of N_r species above forests. No recent reports on long-term flux measurements of N_r were found. Since several N_r compounds contribute to ΣN_r each with different chemical and physical properties, a complex arrangement of different, highly specialized measurement devices would be needed for quantifying ΣN_r exchange. To our knowledge, there is no publication available reporting annual ΣN_r deposition at (remote) forest ecosystems using micrometeorological methods. As stated above, the true benefit of the TRANC is that the most relevant N_r species are converted, and a single instrument is sufficient for determining dry nitrogen deposition. Therewith, we were able to determine annual dry deposition and show seasonal changes in the ΣN_r flux pattern."

Comment R2.12 *Section 2.2: Please add horizontal and vertical displacement between TRANC inlet and anemometer, as well as the pressure downstream of the critical orifice and the turbulent Reynolds number in this low pressure region.*

Response to R2.12: The horizontal and vertical displacement heights were 32 and 20 cm, respectively (Wintjen et al., 2020). "The pressure gradient from the critical orifice to the CLD was not measured. Thus, only assumptions about the turbulent flow regime can be made. Considering tube length and lag time minus residence time in the converter, the latter assumed to 2 sec at maximum due to tube length and platin mesh as an additional flow resistance, flow speed was at 2.7 ms^{-1} at maximum. Using an inner diameter of 4.4 mm and a kinematic viscosity at 15°C ($1.485*10^{-5}$ m^2/s), we calculated a Reynolds number of 800 indicating an overall laminar flow. We cannot provide a reasonable explanation to the low Reynolds number since pressure gradient was not measured.

Generally, the flow type inside the tube affects high-frequency attenuation (Massman, 1991; Lenshow and Raupach, 1991; Moncrieff et al., 1997). High-frequency attenuation was corrected with an empirical method based fully on measured cospectra (Wintjen et al., 2020). Since an empirical approach was used to estimate the high-frequency damping, effects originating from the low Reynolds number and from physical and chemical processes occurred after the critical orifice were considered in the flux analysis."

We replaced line 149-152 by this response.

Comment R2.13: *Some more information on how the DELTA denuders were operated would be helpful without having to look up the quoted references. What filters were used and which coating for the denuders? The use of paper filters has been found to result in an aerosol underestimation of about 30%, which is not an issue for PTFE filters. K2CO3 coating results in a positive artefact on HNO3 from other NOy compounds, while NaCl coating is more selective. It is only in Section 4.1 that the paper seems to imply that K2CO3 coating was used. It is also worth stating that the cut-off of the DELTA denuder is approximately PM4.5 (see Tang et al., 2015; https://uk-air.defra.gov.uk/library/reports?report_id=861). The implications should be discussed also when comparing the TRANC and the sum of the Nr compounds (line 353). Mention also that the APNA-360 NO2 measurement was (presumably) made with a thermal converter and is therefore cross-sensitive to other oxidised nitrogen compounds.*

Response to R2.13: We thank the Reviewer for hinting on the publication of Tang et al. (2015). The positive artifact of carbonate coated denuders on HNO_3, the consequence of using paper filters, and the DELTA denuder cut-off size of 4.5 µm were added to line 187.

"Basic denuders were coated with sodium carbonate to collect HNO_3, SO_2, and HCl. Citric acid was applied to acid denuders for removing NH_3. Two cellulose filter papers (Whatman No. 1, 25mm diameter) were used for collecting aerosols. The first filter was prepared with potassium carbonate in glycerol, the second filter with citric acid."

For measuring NO and NO_2, the used APNA-360 was equipped with a thermal NO_x converter resulting in cross-sensitivity to higher oxidized nitrogen compounds (line 194).

We considered these implications of the chosen coating, the aerosol cut-off size of DELTA, and the thermal NOx converter in the discussion (line 599 to 616, shifted to line 527). In comparison to the DELTA denuders cut-off size, we assume that TRANC cut-off size is higher. Due to the high temperatures in the TRANC (≥ 870°C), coarse NO_3^- aerosols are probably decomposed. For example, sodium nitrate originating from sea salt is converted by the TRANC as shown by Marx et al. (2012). Thus, coarse fractions of nitrogen aerosols were converted in the TRANC implicating a higher cut-off size than the DELTA samplers. We added the discussion about the different cut-off sizes to line 527.

Comment R2.14. *Lines 189-192. I suggest you state already in this context that you were not able to calculate NH3 fluxes.*

Response to R2.14: We added the following sentence to line 191. "In contrast to Zöll et al. (2016), we were not able to calculate NH_3 fluxes with the QCL using the EC method (see Sec. 2.2)".

Comment R2.15: *Please add some details or reference with respect to the wet/bulk deposition measurements. Was a biocide used to avoid denitrification?*

Response to R2.15: Please see the response to R1.4.

Comment R2.16: *Line 305ff. Strictly speaking a "compensation point" is defined at the concentration at which (biological) consumption equals production. Thus, when talking about compensation points in a context other than "stomatal compensation point" it may be better to use the term "emission potential" or "equilibrium concentration", depending on context.*

Response to R2.16: We agree. Please note that lines 298 to 320 were removed as the discussion about R_c was deleted.

Comment R2.17: *Line 315. It is worth noting that the evaporation of NH4NO3 during the deposition process also implies that some of the NH4 and NO3 measured as aerosol does not reach the surface as aerosol but as NH3 and HNO3 and can therefore deposition faster than particles.*

Response to R2.17: We agree. Please note that lines 315ff were deleted.

Comment R2.18: *Section 3.1. Figure 3 actually conveys the relative contribution of NH3 and NOx to total Nr more clearly than Figure 2. Maybe refer forward to Figure 3 when you discuss Fig. 2. In my mind Figure 3 does two things: (a) it shows the best estimate of the relative breakdown of Nr into the different species and (b) it acts as a quality control of the total Nr measurement. However, to interpret the figure in terms of (b), the reader would need to know which stacked bars are fully based on real data and which rely on gap-filling and also the % coverage of the Nr measurement for each data period. Could both pieces of information be added to the figure? With this additional information February 2018 could then be re-added to the figure: it reflects real data, but the gap filling does not work well on this data point. It would also indicate the years to which the sampling periods refer.*

Response to R2.18: We appreciate your suggestions to the discussion of figures 2 and 3. We added the following sentence after line 325. "A breakdown of ΣN_r in compounds contributing most to its concentration pattern is shown in Fig. 3 illustrating a comparison of ΣN_r concentrations with DELTA denuder and NO_x measurements on monthly basis."

We added coverage of valid ΣN_r concentration measurements in % for each exposure period. After carefully reviewing DELTA concentrations and replacing missing NH_3 values by passive samplers, we had data gaps for January 2017 (only NH_3), December 2017, March 2018, and April 2018. Remaining gaps averages were replaced by monthly averages estimated from other years. We rephrased the caption of Fig. 3 and indicate gap-filled bars as hatched. We added the year to the x-axis labels. Coverages of TRANC measurements during each exposure period were added if TRANC data was available. In the previous version of Fig. 3, TRANC concentrations were shown for June 2017. During the exposure period of the denuders, we had less than 1% coverage of TRANC measurements. We decided to remove that bar.

Comment 2.19: *Line 335. No systematic difference in NH3 between 20 and 30 m would indicate that NH3 showed no flux. Or is the uncertainty just too large to resolve gradients?*

Response to 2.19: For the entire campaign, "we found no systematic difference between NH_3 concentrations within the canopy and just above the canopy. Only for short time periods, for example summer 2016 and 2017, differences in passive samplers were found indicating a small NH_3 flux. Considering the LOD for IVL passive samplers for NH_3 of 0.4 µg N m^{-3} determined by Dämmgen et al. (2010), shows that passive sampler measurements were conducted close to their LOD. It suggests that the uncertainty of the passive samplers was too large to resolve flux gradients." We added these details to the discussion (line 527).

Comment R2.20: *I have some comments regarding the assessment of the limit of detection and positive and negative fluxes (lines 371 to 390). The Finkelstein and Sims (2001) algorithm returns a different random error (and hence detection limit) for each 30-minute flux value. It is fine to state the average / median of this detection limit, but does it not make more sense to evaluate the fraction of data points for which the LOD is exceeded against individual LODs rather than the average LOD. For a near-zero* *flux below the LOD one would expect about half of the flux values to be positive and half to be negative, but this does not really carry much information on the actual contribution of emission events as many of the positive fluxes would not be significantly different from zero. It would therefore be useful to add what fraction of the flux values above the LOD shows emission and deposition. The LOD is a function of instrument signal-to-noise, but also of turbulence and would be expected to be larger over forest. This needs to be taken into account when comparing LODs between studies (lines 538ff). As with other parts of the manuscript it is not very clear whether the median deposition figures (lines 381ff) refer to the filtered or the gap-filled data.*

Response to R2.20: According to your suggestion, we did the analysis of the LOD on half-hourly basis. "The comparison of half-hourly fluxes with their individual LOD revealed that 79% of the measured fluxes were above their detection limits. Deposition fluxes contributed with 84% to fluxes above the LOD. The fraction of emission was estimated to 16%. The relative contribution of emission fluxes to measured fluxes decreased under the consideration of the LOD. It shows that emission fluxes were closer to the flux detection limit of the instrument." We added these sentences to line 374ff.

We agree that micrometeorological factors like turbulence influence the flux detection limit, too. We rephrased the statements in line 538ff. As noted in comment R2.6, measured fluxes refer to filtered fluxes.

Comment R2.21: *Figure 9: Fluxes scale with gc = 1/Rc rather than with Rc and thus mean values of Rc should be calculated by averaging 1/Rc values and then turning back into Rc (or presenting as gc). The resulting pattern can look quite different. Was a filter applied for maximum that was allowed for Ra+Rb? At large values of Ra+Rb, Rc potentially becomes a small difference of two large numbers and thus quite uncertain.*

Response to R2.21: We applied filters for R_a and R_b. Possibly, they were not strict enough as seen by the outliers in Fig. 9. As outlined in R2.1 to R2.4, we removed the discussion on R_c since an accurate calculation of R_b was not possible for ΣN_r.

Comment R2.22: *It would be worth discussing the annual N input (line 507) in relation to the Critical Loads for woodland.*

Response to R2.22: As written in lines 114 to 115, the discussion about critical loads will included in part II. However, we agree that critical loads for woodland should be mentioned. Published critical loads for *Picea abies* and *Fagus sylvatica* ranged from 10 to 15 kg N ha^{-1} a^{-1} and 10 to 20 kg N ha^{-1} a^{-1}, respectively (Bobbink and Hettelingh, 2011). Since the forest stand consists to approximately 80% of Norway spruce in the footprint and the surrounding forest stand is predominated by Norway spruce, the critical load for the forest stand is probably closer to the limits of *Picea abies.* Estimated annual N input was 12.2 and 10.9 kg N ha^{-1} a^{-1} for the measurement years and were found to be at lower estimate of critical loads. It suggests that the forest is currently not in a critical state in relation atmospheric N input. We integrated details of this response to line 842.

Comment R2.23: *Line 549. Did February 2018 stand out in any other way? Was the wind direction unusual? Do the reports of the federal and state measurement networks report anything unusual?*

Response to R2.23: Please note the response to R1.14.

Comment R2.24: *Lines 562-565. I am not convinced there is a threshold NH3 concentration for ammonium sulfate formation. I thought any free NH3 would be pulled into the aerosol phase by the presence of sulphuric acid or bisulfate. Also, I am not sure this analysis works well with monthly data. The high concentrations could have been due to a short event during which no NH3 was present.*

Comment R2.24: As written above, please note the response to R1.14. We agree that a threshold analysis did not work well with monthly data. Therefore, we decided to delete these lines.

Comment R2.25: *Lines 566 to 598. It is fine to point out the difference in flux during the winter periods 2016 and 2017, and their relationship to snow cover. However, it would be prudent to show in Fig. 12a the fluxes during periods with snowfall as dotted lines as they are highly uncertain (it could be argued they should be removed completely) and I find the section overly lengthy and speculative. I am highly sceptical that NH3 would be able to diffuse through a 60 cm snow layer without being re-captured. Is there literature evidence that this might be possible? Also, please select colours to be readable by people with red/green blindness (Figure 12).*

Response to R2.25: In Fig. 12, we showed fluxes smoothed with a 3-h running mean. In case of averaging the uncertainty of the fluxes is significantly reduced. Still, uncertainties are not reduced completely. Due to visibility reasons, we decided to show fluxes and concentrations as solid lines and changed the colors according to your suggestion.

We decided to remove the discussion about NH_3 since the diffusion of NH_3 through a snow layer of a large depth (60 cm) seems improbable. We found no literature evidence for diffusion of NH_3 through a snow/ice layer. NO could be responsible for the observed ΣN_r emission fluxes but different observations were made about correlations of NO with snow cover, micrometeorological parameters, and about sources of NO emissions as stated in the manuscript. Since we had no measurements of NO close to the forest floor or measurements of the mass loss rate of litter under snow cover, we can only made assumptions about the origin of the ΣN_r emission fluxes. As mentioned in lines 596 zo 598, NO emitted from the forest soil is rapidly converted to NO_2 (Rummel et al., 2002). Thus, the measured ΣN_r flux probably had a high NO_2 contribution during that time. In the revised version of the manuscript, we shortened the discussion of Fig. 12.

Comment R2.26: *Line 604f. I don't understand this sentence. Are you trying to say that the DELTA measurements suggested that gaseous compounds made a significant contribution to the Nr concentration?*

Response to R2.26: Yes. We rephrased the sentence.

Comment R2.27*: Line 605. Which slight increase in HNO3 and decrease in NH4+?*

Response to R2.27: The increase in the relative contributions of HNO_3 from spring to summer compared to the decrease of NH_4^+ and NO_3^- (Fig. 4) can be related to the evaporation of NH_4NO_3. We rephrased the sentence according to this response.

Comment R2.28: *Line 610ff. The NOx analyser was likely a thermal analyser and cross-sensitive to* *other NOy compounds? Worth mentioning here also the likely difference in cut-off diameters between DELTA and TRUNC for aerosol.*

Response to R2.28: As written in R2.13, the NO_x analyzer was a thermal analyzer and likely cross-sensitive to other NO_y compounds. "However, measured concentrations of HNO_3 or NO_3^- were comparatively low as seen in Fig. 3. Thus, their influence on NO_x measurements appeared to be negligible." As written in R2.13, we integrated the possible difference in cut-off sizes of TRANC and DELTA in the discussion (line 527ff).

Comment R2.29. *Line 614f. What is your evidence that the DELTA suffered break-through at high concentration peaks? Or are you just speculating that this might be a possibility. Maybe the use of the word "could" is not quite right? Also, a key uncertainty originates from the TRUNC measurement likely not covering 100% of the DELTA sampling time. See comment 18 above.*

Response to R2.29: Reasons for differences between TRANC and DELTA are the cut-off diameters, issues in the conversion efficiency, and the data coverage of the TRANC, which was 78% on average for the exposure periods. In addition, N_2O_5 or peroxy acetyl nitrates are not collected by denuders, but probably converted by the TRANC. We rephrased line 614f accordingly.

Comment R2.30: *Lines 617-624. Deposition velocities of NH3 are highly variable and would be expected to decrease for semi-natural forests that are subject to high Nr input (because the stomatal compensation points would go up; see Massad et al., 2010) and with decreasing ambient concentration (away from sources). The importance of the NH4NO3 evaporation effect that likely affected the summer measurements of Wolff et al. (2010) would likely be much smaller during cooler periods resulting in smaller deposition rates at other times of the year. So I am not sure the conclusions hold.*

Response to R2.30: After carefully reflecting your comment, we found a comparison of a v_d for ΣN_r with individually measured v_d as less useful since we have no information about their relative contribution to the v_d of ΣNr. Thus, we decided to remove the lines 617 to 624.

Comment R2.31: *Line 639. A stomatal compensation point has only been shown to exist for NH3 and has in some studies been indicated for NO2. There is no such thing as a stomatal compensation point for total Nr. And there is also no canopy compensation point for Nr (Line 685). The concept of a canopy compensation point has not been introduced in the paper anyway.*

Response to R2.31: We agree that the sentence is incorrect (line 639) and the canopy compensation point was not introduced. Please note the lines were removed and the substantial changes to Sec 4.2.

Comment R2.32: *Line 645. HNO3 is formed by reaction of NOx with OH not O3.*

Response to R2.32: Corrected.

Comment R2.33: *Lines 664 to 669. This paragraph seems to mix up the effects of concentration on the flux and vd. Clearly, for a depositing compound, the flux increases with concentration. For vd this may or may not be the case.*

Response to R2.33: We agree that in these lines the effect of concentration on flux and v_d are mixed up. The analysis of Fig 5 has shown that v_d was not dependent on concentration. We corrected the statements accordingly.

Comment R2.34: *Lines 729 to 738. I am not sure the water holding capacity of leaves at intermediate relative humidity is governed only by NO3-/NH4- in air. Any hygropscopic aerosol from dry, wet and fog deposition could contribute to this. As the authors show their measurement site is by no means pristine. See also Sutton et al. (1998) and Flechard et al. (1999).*

Response to R.34: These lines belong to the Fig 9 and v_d measured during precipitation. The discussion about Fig. 9 was removed and fluxes determined during rain were filtered out from analysis. Thus, we deleted lines 729 to 738.

Comment R2.35: Line 801. What do you mean by ‘canopy outflow’? Do you mean “catchment outflow” or “throughfall”? How can those measurements distinguish between dry and wet deposition?

Response to R2.35: We meant here the canopy budget technique (Beudert et al. 2014). We corrected the sentence.

Comment R2.36: *Line 803. Which two sampler types? Positive artefacts on bulk deposition gauges (if this is what you are referring to here) can also originate from dry deposition of gas phase NH3 and HNO3.*

Response to R2.36: We referred here wet deposition from wet-only and bulk samplers. As shown in Table 1, differences between bulk and wet-only deposition were negligible. Small differences could be induced by organic and inorganic dusts or related to dry deposition of NH_3 and HNO_3 as you mentioned. The effects were not relevant for the annual nitrogen deposition at the measurement site. We rephrased line 803 according to the Reviewers’ comments.

Comment R2.37: *Line 834. I can see that inferential modelling would extrapolate fluxes mechanistically to low turbulence conditions, however I fail to see how this is possible with neutral flux networks if they are trained with u* filtered data.*

Response to R2.37: If neural networks are trained with u_* filtered data and subsequently used for gap-filling, biases to the u_* threshold are introduced. We decided to remove neural networks from the sentence.

Technical comments

Comment R2.38: *The papers uses different units in different places. When discussing the Nr components these are in µg-N m-3, which is logical, but when referring to previous measurements (e.g. line 121, 512, 521, …) they change to ppb. I suggest adding values in µg-N m-3 in brackets here so the reader can compare more easily.*

Response to R2.38: We thank the Reviewer for his suggestion. We changed the unit to ppb because the cited references provided their results in ppb. We adapted your suggestion and added values in µg N m^{-3} in brackets.

Comment R2.39: *Line 10. "was observed for the contribution of NH3 …"*

Response to R2.39: Changed.

Comment R2.40: *Line 15. "changes in composition of ΣNr and radiation"*

Response to R2.40: Changed.

Comment R2.41: *Line 23. "During these periods, cuticular or soil …"*

Response to R2.41: Sentence was deleted.

Comment R2.42: *Line 38. Correct subscript on PM2.5.*

Response to R2.42: Corrected.

Comment R2.43: *Line 45. The community tends to use "nitric oxide" over "nitrogen monoxide" for NO.*

Response to R2.43: We changed the order.

Comment R2.44: *Line 74. "… radiation as the primary driver for …"*

Response to R2.44: Please note that lines 66-91 were removed.

Comment R2.45: *Line 76. "… as a secondary driver …"*

Response to R2.45: see above.

Comment R2.46: *Line 134. "the dominating Norway spruce is recovering"*

Response to R2.46: Corrected.

Comment R2.47: *Line 149. The sentence starting "The mass flow rate …" seems redundant.*

Response to R2.47: We agree. The sentence was removed.

Comment R2.48: *Line 208. The sentence "Figures with the notation …" seems over the top. I suggest you just write "(see Fig. S1 of the Supplementary Material)"*

Response to R2.48: We deleted the sentence and adapted your suggestion.

Comment R2.49: *Line 266. Missing parenthesis "2010))."*

Response to R2.49: Corrected.

Comment R2.50: *Figure 2. I think the figures would be more readable if they all used the same y-scale, possibly capped at 15 ug N m-3.*

Response to R2.50: We adjusted the y-limit of all subplots to 15 µg N $m^{-3.}$

Comment R2.51: *Figures 2 and 5. The whiskers do not look like they scale with the IQR. Please state sampling intervals for both figures as the statistics depend on it.*

Response to R2.51: In Fig. 2, the boxplots cover the entire measurement campaign. As noted in Fig. 5, boxplots refer to monthly intervals. The whiskers extend to the outermost points within the Q1 - 1.5*IQR and Q3 + 1.5*IQR range. Thus, whiskers have different lengths.

Comment R2.52: *Line 329. "reached values of up to"*

Response to R2.52: Corrected.

Comment R2.53: *Figure 5. Rather than the last sentence in the caption, the authors could use arrows with values to indicate the magnitude of the three points that fall outside the y-range.*

Response to R2.53: "The whiskers in February 2018 cover the range from -191 to 105 ng N m^{-2} s^{-1}, the upper whisker of December 2017 was at 69 ng N m^{-2} s^{-1}." We rephrased the sentence according to this response.

Comment R2.54: *Line 405. "shows the median vd for the corresponding fluxes."*

Response to R2.54: Corrected.

Comment R2.55: *Line 416. Should this more accurately read "the deposition of total Nr."?*

Response to R2.55: Corrected.

Comment R2.56: *Figures 8 and 9. I would find these easier to grasp if the plots of the first row were labelled (a), (b), (c) etc.*

Response to R2.56: We changed the labelling order.

Comment R2.57: *Line 443. "dry conditions (no precipitation) are associated with enhanced deposition"*

Response to R2.57: Corrected.

Comment R2.58: *Line 507. "In total, we derived a total"*

Response to R2.58: Corrected.

Comment R2.59: *Line 521. The word "who" cannot refer to authors in brackets.*

Response to R2.59: We removed "who" and started a new sentence beginning with "The authors …"

Comment R2.60: *Line 524. Change "expectable" to "to be expected" – it is highly unusual and not in all dictionaries.*

Response to R2.60: Done.

Comment R2.61: *Line 535. Change "phase" to "period".*

Response to R2.61: Done.

Comment R2.62: *Line 542. "that the flux magnitude"*

Response to R2.62: Corrected.

Comment R2.63: *Line 559. Correct "SO42-"*

Response to R2.63: Please note that the sentence was deleted.

Comment R2.64: *Line 560. "is the dominant aerosol form."*

Response to R2.64: Please note that the sentence was deleted.

Comment R2.65: *Line 599. "that NOx made the largest contribution"*

Response to R2.65: Changed.

Comment R2.66: *Line 603. "NH3 had a strong presence"*

Response to R2.66: Corrected.

Comment R2.67: *Line 653. "may also be related"*

Response to R2.67: Please note that the sentence was deleted.

Comment R2.68: *Line 657. "as a primary controlling"*

Response to R2.68: Please note that the sentence was deleted.

Comment R2.69: *Line 705. "During periods of"*

Response to R2.69: Please note that the sentence was deleted.

Comment R2.70: *Line 723. "When the canopy gets drier"*

Response to R2.70: Please note that the sentence was deleted.

Comment R2.71: Line 726. "than stomatal deposition"

Response to R2.71: Please note that the sentence was deleted.

Comment R2.72: Line 788. *This acronyms were introduced in line 95 and not used since! Probably worth spelling out in full here.*

Response to R2.72: Agreed. We wrote their full form here.

Comment R2.73: *Line 834. "to the use of friction"*

Response to R2.73: Please note that the sentence was deleted.

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