Response to reviewers' comments - manuscript *BG-2020-364* "Forest-atmosphere exchange of reactive nitrogen in a low polluted area – temporal dynamics and annual budgets"

We thank the reviewers for their constructive comments. As outlined in the author comment, comments to measurement part of the manuscript are answered in this review. Comments to the modeling part are highlighted in red. Referee comments are given in italic, the answers in standard font. The comments by Reviewer 1 are numbered from R1.1 to R1.81 titled as specific comments. The main scientific comments of Reviewer 2 range from R2.1 to R2.15, the additional comments start at R2.16 and end at R2.42, and the comment from R2.43 to 2.76 refer to technical corrections/suggestions. Comments of Reviewer 3 range from R3.1 to R3.5. The line and figure numbers in the answers, where we add the new information into the manuscript, refer in this document to the originally submitted version. The text which is enclosed by "..." is implemented in the revised manuscript.

Response to Reviewer 1

General Comments *This manuscript presents 2.5 years of measurements of total reactive nitrogen (Nr) fluxes above a mixed forest in Germany. The measurements are used to assess annual dry deposition budgets and are then compared to deposition estimates derived from a field scale model and a gridded chemical transport model. This study directly addresses the need for new Nr flux measurements to improve Nr deposition budgets, assess exceedances of critical loads of Nr, and improve models of reactive N deposition. The dataset developed is novel and should prove useful to the ecological and atmospheric chemistry communities interested in N deposition. Furthermore, the interpretation of the measurements in relation to micrometeorology and atmospheric chemistry sheds new light on the processes influencing air-surface exchange of Nr and the relative importance of Nr species to the dry Nr deposition budget. However, there are a number of technical details of the analysis and discussion, along with some organizational issues, that should be addressed before the paper is suitable for publication. In general, the paper would benefit from a more thorough quantitative analysis of the flux patterns and their relation to micrometeorology and atmospheric chemistry. Section 4.2 touches on these relationships but could be extended along the lines of several suggestions outlined below. As also suggested below, the current content of Section 4.2 could be reorganized and shortened by eliminating some redundancies, making it possible to expand the analysis without significantly lengthening the Section overall. Sections 4.3.2 and 4.3.3, which describe uncertainties in the modeling approaches, as well as the Conclusions section, could be significantly reduced in length. More specific comments are detailed below.*

We thank the Reviewer for his/her comments on this work. As mentioned in the author comment, we split the preprint into two parts. We appreciate your suggestions to the discussion of the measurements. We included a comparison of the TRANC measurements to results from DELTA samplers and investigated the effect of micrometeorology on the deposition velocity and resistances of ΣN_r. As outlined in the author comment, discussions on the modeling results will be shifted to the modeling manuscript. Questions related to the nitrogen modeling will not answered in this response in detail. Your suggestions will be included in the preparation of the modeling manuscript.

Specific comments

Comment R1.1 *Line 83: Change "pattern" to "patterns".*
Response to R1.1 Revised.

Comment R1.2 *Line 90: Should the first word be "methods"?*
Response to R1.2 Yes.

Comment R1.3 *Line 94: CTM should be plural.*
Response to R1.3 Revised.

Comment R1.4 *Line 117: "site located" should be "site is located"*
Response to R1.4 Done.

Comment R1.5 *Line 145: For clarity, consider rewording this sentence to something like: "In a 2nd step, a gold tube passively heated to 300C catalytically converts the remaining oxidized Nr species to NO."*

Response to R1.5 We reworded the sentence according to your suggestion.

Comment R1.6 *Line 148: Is 2.1 L/min the flow rate through the converters (atmospheric pressure) or through the reduced pressure portion of the tubing downstream of the orifice? If the latter, please indicate the flow rate through the converters.*
Response to R1.6 2.1 L min^{-1} was the flow rate after the critical orifice. The following lines were added to line 148: "The mass flow rate before the critical orifice was the same as after the critical orifice. Since mass flow was equal to both sides of the critical orifice, a difference in flow velocity was induced due to the reduction in pressure. Flow velocities were not measured for the different sections."

Comment R1.7 *Line 164: What type of passive sampler was used? What was the sampling duration?*
Response to R1.7 "Passive samplers of the IVL type (Ferm, 1991) were used for NH_3, and the exposition duration was approximately one month at a time". The information given here was added to line 164.

Comment R1.8 *Line 207: What was the typical magnitude of this correction to the total Nr flux?*
Response to R1.8 "The correction contributed approximately 132 g N ha^{-1} to two years of TRANC flux measurements if the Mean-Diurnal-Variation (MDV) approach was used as gap-filling approach. Half-hourly interference fluxes were between -3 and +0.3 ng N m^{-2} s^{-1}. Their random flux uncertainty ranged between 0.0 and 0.5 ng N m^{-2} s^{-1}" We added the information given in this response to line 209.

Comment R1.9 *Line 216: What caused the reduced sensitivity of the CLD and how was it identified?*
Response to R1.9 "The reduction in sensitivity may be caused by reduced pump performance leading to an increase in sample cell pressure. If pressure in the sampling cell is outside the regular operating range, low pressure conditions needed for the detection of photons emitted by excited NO_2 molecules may not hold. Pump efficiency was controlled at least monthly, and tip seals were replaced if necessary. The sensitivity of the CLD could also be reduced by changes in the O_2 supply from gas tanks to ambient, dried box air if O_2 gas tanks were empty. Issues in the air-conditioning system of the box could also affect the sensitivity of the CLD. An influence of aging on the inlet, tubes, and filters may also affect the measurements. In order to minimize an impact on the measurements, half-hourly raw concentrations were carefully checked for irregularities like spikes or drop-outs by visual screening." We added the information given in this response to line 216.

Comment R1.10 *Line 266: How was the quality of the DELTA measurements assessed?*
Response to R1.10 "The denuder preparation and subsequent analyzing of the probes was identical to the procedure for KAPS denuders (Kananaskis Atmospheric Pollutant Sampler, (Peake, 1985; Peake and Legge, 1987)) given in Dämmgen et al. (2010) and Hurkuck et al. (2014). We controlled the pump flow to keep it at a constant level and checked the pipes for contamination effects before analyzing. Blank values were used as additional quality control." We added the description of the DELTA measurements to line 165.

Comment R1.11 *Line 275: Has LAI been measured at this site? How variable is the LAI throughout the seasons, given the relative fractions of spruce and beech.*
Response to R1.11 The leaf area index (LAI) was not measured at the site. It was modeled after the same scheme used for DEPAC (see Appendix B of van Zanten et al., 2010). A linear increase of the LAI was modeled from mid of April to begin of May, a linear decrease from October to begin of November. Values ranged between 4.1 and 4.8. Fig. R1 shows the modeled LAI for measured land-use classes.

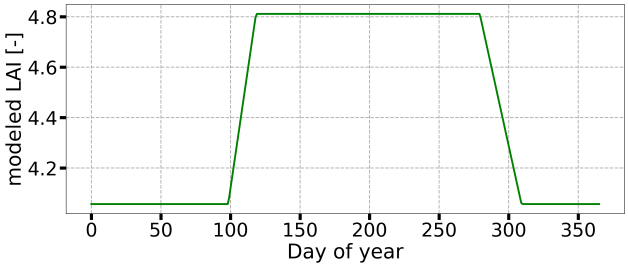

Figure R1: Modeled LAI following van Zanten et al. (2010) for measured fractions of coniferous forest (81.1%) and deciduous forest (18.9%) within the flux foot print for a year.

Figure R1 was added to Sec 2.4 "Determining deposition velocity and canopy resistance of ΣN_r from measurements".

Comment R1.12 *Line 285: Should be "gaps in micrometeorological".*
Response to R1.12 Yes, you are right.

Comment R1.13 *Figure 1: The blue bar in the lower plot is incorrectly labeled NH3.*
Response to R1.13 Corrected.

Comment R1.14 *Line 296: The findings here relative to concentrations of NOx > 20ppb make me question the description of this site as being situated in a "low pollution" area. Some additional justification of this site characterization is needed.*
Response to R1.14 We characterized the site as "low polluted" since average concentration level of NO_x was comparatively low (see Fig. 1). NO_x peaks above 10 μg N m^{-3} were observed only for short time periods during winter.

Comment R1.15 *Line 302: The figure numbering configuration for the Appendices (e.g., Figure B1) was not immediately clear to me. I believe the format for Biogeosciences is for such material to be included as "Supplemental Material".*
Response to R1.15 We agree that some figures are more suitable for "Supplemental Material". For the revised version, we prepared a supplemental file. Figure A1 of the appendix was moved to the supplemental file. Other figures of the appendix were deleted.

Comment R1.16 *Line 303: How does the sum of the concentrations measured by the DELTA compare to the TRANC Nr measurement?*
Response to R1.16 According to the suggestion of Reviewer 2, we added a stacked bar graph (Fig. R9) showing monthly concentrations of the DELTA measurements compared to the TRANC ΣN_r concentrations. Latter were averaged to the exposition period of the DELTA samplers. The

comparison revealed significant underestimations of TRANC ΣN_r from March to mid of May 2018 and from July to mid of August 2017. We found that the zero-air calibration value of the TRANC-CLD system was incorrect for the mentioned time periods by approximately 0.9 μg N m^{-3} compared to the uncorrected TRANC-CLD concentrations. Concentrations and fluxes were recalculated with the bias correction. Figures and evaluations shown in the response and in the revised manuscript were made with the bias-corrected data. Figures of the manuscript were updated accordingly. On average, the TRANC values were slightly higher by 0.3 μg N m^{-3} than DELTA+NO_x. The results of this comparison were added to line 302.

Comment R1.17 *Line 317: What fraction of the non gap-filled half-hourly fluxes exceeded the flux detection limit?*
Response to R1.17 The following sentences were added to line 318 "In total, 51% of the non gap-filled fluxes were higher than the flux detection limit. It shows that for large parts nitrogen dry deposition was close to detection limit of the used measuring device and that nitrogen exchange happened at a comparatively low level." Despite the low signal-to-noise ratio at the measurement site, we were able to investigate the exchange pattern of ΣN_r and could estimate reliable dry deposition sums (see R2.9).

Comment R1.18 *Line 350: Should "based on" be "are based on"?*
Response to R1.18 Yes, you are right. Please note that sentence was deleted.

Comment R1.19 *Line 352: I might expect the sensor nearest the ground to remain "wet" later into the morning than the sensor closest to the top of the canopy.*
Response to R1.19 The statement made in line 352 was misleading. As an example, Fig. R2 shows diurnal patterns of the leaf wetness for all sensors on monthly basis for 2017. Since no difference was found between the spruce and beech tree, colors were chosen to highlight a potential effect of the measurement heights on the leaf surface wetness.

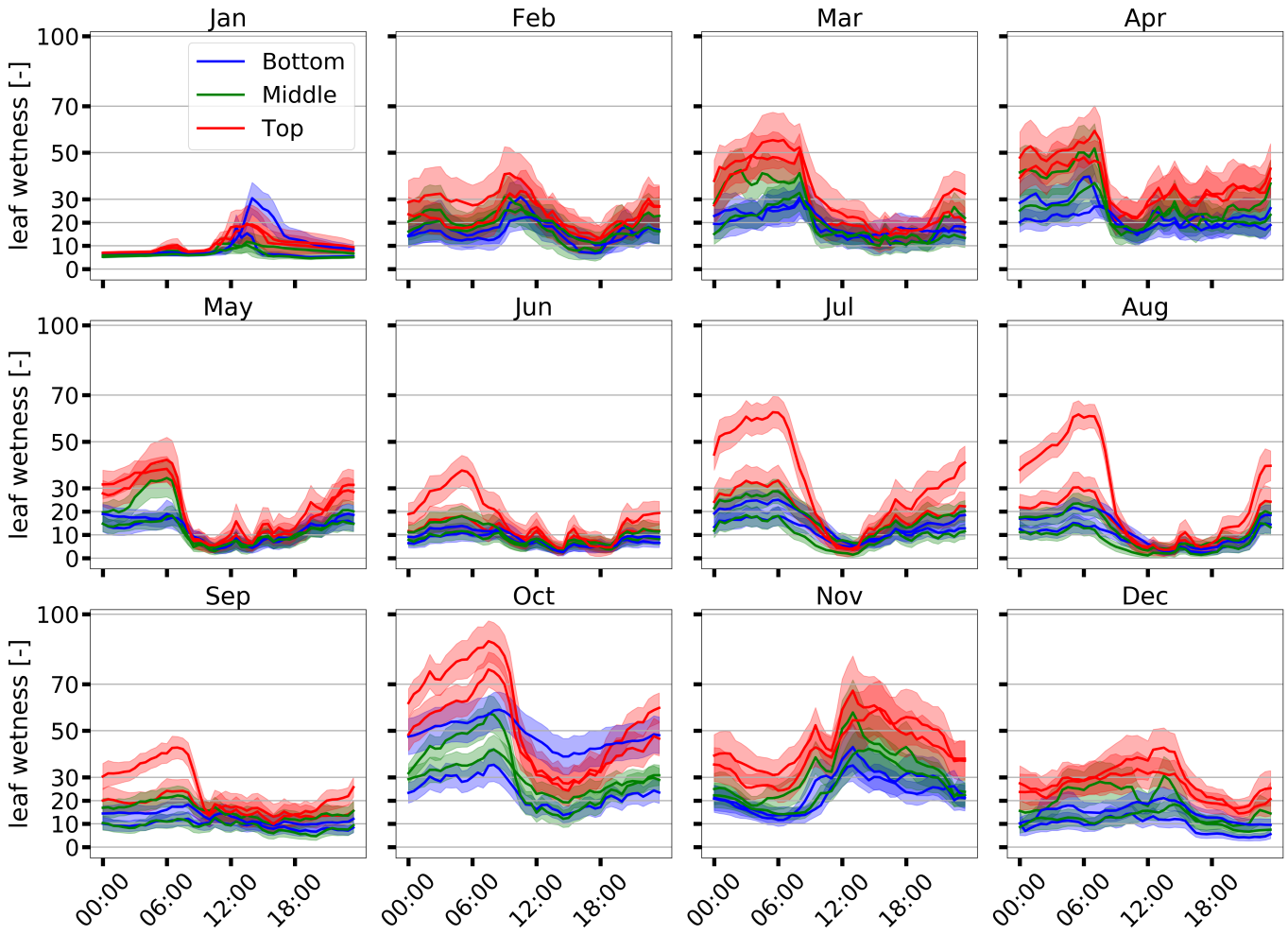

Figure R2: Daily cycles of the leaf wetness for 2017. Colors indicate installation heights of the sensors (red=top, green=middle, blue=bottom). Shaded areas represent the standard error of the mean.

Figure R2 shows diurnal patterns of the leaf wetness for all sensors on monthly basis for 2017. On monthly basis, the diurnal patterns of the sensors were almost the same for a season. From April, the start of the growing season, to September highest values were measured during dawn and lowest values during the day. During daylight, only slight differences in measurement height were visible. Considering the standard error, the differences in measurement heights diminished, especially between the lowest and middle sensor. Also, sensors from the mid and the top were within their uncertainty ranges. In conclusion, sensors at the lowest height seem to remain "wet" later during the morning, but effect is within the standard error range. Using only the top sensors for deriving the leaf wetness value, seems not to be appropriate with regard to the uncertainty ranges. Thus, we used all sensors for deriving a wetness boolean, which also lowered its uncertainty. Figure R2 and corresponding description were provided as supplemental material.

Comment R1.20 *Line 398: "It seems that....most likely driven by particulate Nr compounds." Is this supported by the particulate measurements? Do the DELTA measurements show relatively higher concentrations of particulate NH4NO3 during this period? Given the lower Vd of particles relative to gases, the concentrations would need to be much higher to drive the high total N*

deposition during this period, correct?
Response to R1.20 Unfortunately, we had no DELTA measurements during this period since the pump was not working properly. However, we found that SO_2 concentrations were remarkably high during the deposition period in February 2018. Passive sampler measurements showed a low NH_3 concentration level in February 2018. Presumably, ammonium sulfate or compounds formed at lower NH_3 concentrations, e.g., ammonium bisulfate were responsible for the observed ΣN_r deposition. Please see Sec. 4.1 of the revised version for further details.

Comment R1.21 *Lines 404 and 405: So a linear interpolation is used? Please clarify.*
Response to R1.21 The deposition of the first half of 2018 was linearly interpolated to the end of 2018. As a second approach, we calculated the average deposition from the second half of 2016 and 2017 and used their average as assumption for the second half of 2018. Due to the deposition event in February 2018, the TRANC deposition estimated with MDV was significantly higher in case of linear interpolation. We will add this aspect to the second part.

Comment R1.22 *Line 411: "..significantly higher lower and upper.." I understand what you mean here but it is a little confusing. Consider rewording for clarity.*
Response to R1.22 Agreed. We will reword it.

Comment R1.23 *Line 425: "4.6 kg N/ha/a are determined as a lower estimate." Please clarify how this estimate was determined.*
Response to R1.23 Probably, you are referring to line 415. The given values are averages of the lower and upper canopy budget technique (CBT) estimates from 2016 to 2018. We will add it as explanation and think about more proper names for the calculated values.

Comment R1.24 *Line 426: Should this section heading read "Sensitivity of deposition estimates to measured vs. modeled input parameters"?*
Response to R1.24 We agree. We will change the section header within the preparation of the modeling study.

Comment R1.25 *Line 428: Remove comma after "class" and add "the" after "considering".*
Response to R1.25 Revised.

Comment R1.26 *Line 432: Specify that you are referring to the apoplastic ratio of NH4+ to H+. I would also suggest you clarify that you are referring to the stomatal compensation point in the latter part of this sentence. Have any measurements of the soil and vegetation chemistry been conducted at this site such that compensation points could be estimated?*
Response to R1.26 We appreciate the Reviewer's suggestion. Unfortunately, no measurements of soil and vegetation chemistry had been conducted at the site.

Comment R1.27 *Line 435: "...global radiation enhances the opening of the width of the stomata" It may be more straightforward to say that the stomatal resistance is influenced by global radiation.*
Response to R1.27 Agreed.

Comment R1.28 *Line 439: I would suggest that reporting the bias (absolute percent) in the modeled values relative to the measured values is more informative than the correlation in this context.*

Response to R1.28 We agree and will modify the description of the figure within the modeling part.

Comment R1.29 *Lines 450 - 455: A table comparing the measured Nr species concentrations (Delta compounds, NOx, QCL NH3, passive NH3) to LOTOS-EUROS would help clarify this section.*
Response to R1.29 We will add a stacked bar graph to similar to Fig. R9 but with the ΣN_r concentrations from LOTOS-EUROS to the modeling manuscript. A time series showing monthly averages of NH_3 from the QCL, passive samplers, and LOTOS-EUROS is also planned. Please also note Fig. R13 showing a comparison of the different NH_3 measurement techniques.

Comment R1.30 *Line 458: "... are very low compared to other studies" This statement is true relative to the three references cited but perhaps not so for Nr flux studies in a global sense. Some additional context is required for this statement, e.g., low relative to sites influenced by agricultural activities, previous studies in European ecosystems, etc.*
Response to R1.30 We agree that the interpretation of measured concentrations and fluxes was misleading (see also R2.4). We modified deleted "are very low compared to other sites" and replaced it by "low relative to sites exposed to agricultural activities or urban environments."

Comment R1.31 *Line 464: Consider modifying sentence to "...higher ground-level concentrations...".*
Response to R1.31 Revised.

Comment R1.32 *Line 470: "Values..." This sentence seems incomplete.*
Response to R1.32 We changed it to "Concentration values of NH_3 and NO_x...".

Comment R1.33 *Line 472: "...confirm the seasonal pattern of Nr". Do you mean that those studies show patterns consistent with the current study?*
Response to R1.33 The corresponding lines were deleted in the revised version and were replaced by the following sentence "Studies like Wyers and Erisman (1998); Horii et al. (2004); Wolff et al. (2010) conducted measurements of NH_3 and NO_2 above remote (mixed) forests and reported similar concentrations for those gases."

Comment R1.34 *Line 473: "Obviously, measured concentration levels were significantly higher since the observed ecosystems were subject of agricultural management or in close proximity to industrial or agricultural emissions." Are the authors referring here to the studies listed in Line 471? At least for the Geddes study, NOx was lower than in the present study. Please clarify and correct this statement as needed.*
Response to R1.34 We thank the Reviewer for his/her recommendation. Yes, it should refer to publications listed in line 471. We modified the sentence accordingly.

Comment R1.35 *Line 477: This sentence should include references for the "few studies focusing on Nr".*
Response to R1.35 Agreed. Namely Ammann et al. (2012), Brümmer et al. (2013), Zöll et al. (2019), and Wintjen et al. (2020) measured ΣN_r fluxes with the eddy-covariance method. We added the references to the corresponding line.

Comment R1.36 *Line 483: Please consider changing "their flux pattern" to "the flux pattern observed by Ammann et al. (2012)...".*
Response to R1.36 Agreed.

Comment R1.37 *Lines 488 – 497: The discussion of the high emission fluxes observed in December requires some additional detail and clarification. The authors refer to decomposition of fallen leaves beneath a snow layer. Are the authors suggesting that the decomposition is enhancing emissions of NH3 or NO or both? Decomposition rates typically decrease at low temperatures. The authors mention that they "discovered an increase in nitrogen concentration in the investigated samples". Samples of what? Soil? How were these samples taken and analyzed and for which compounds? How frequently were they collected and at what depths? How much did the N concentrations increase and over what time period? The statement regarding the influence of the freeze-thaw cycle on the emission fluxes is interesting but very speculative. Can a soluble gas like NH3 diffuse through a partially wet snow layer to the atmosphere? Do the fluxes correlate with air temperature in a pattern that would support this statement? Looking more closely at the December diurnal profiles in Figure 3 it appears the emission fluxes were mostly observed in 2017, which also had much higher variability in general than 2016. Were there more periods of snow cover in 2017? Did the two years differ in other ways in terms of meteorology or air concentrations that might help explain the emissions observed in 2017?*
Response to R1.37 First of all, we thank the Reviewer for his/her suggestions to this paragraph and we agree that clarifications and more details are needed. We did not take leaf or soil samples at the site. "discovered an increase in nitrogen concentration in the investigated samples" referred to Taylor and Jones (1990). As suggested, we took a closer look in the temperature, concentration, and snow fall measurements during the emission period in December 2017 and compared them to the same period in December 2016. Figure R3 shows recorded temperature, snow fall, concentrations, and estimated fluxes of ΣN_r from 6 December to 15 December for 2016 and 2017. We deleted lines 488-497 and replaced the description as follows:

"In December 2017, large emission fluxes were measured. Compared to 2016, significant difference in temperature and snowdepth were observed. Figure R3 shows recorded temperature, snow fall, concentrations, and estimated fluxes of ΣN_r from 6 December to 15 December for 2016 and 2017. Here, ± 3 days were chosen for filling the gaps in order to keep the short-term variability of the fluxes.

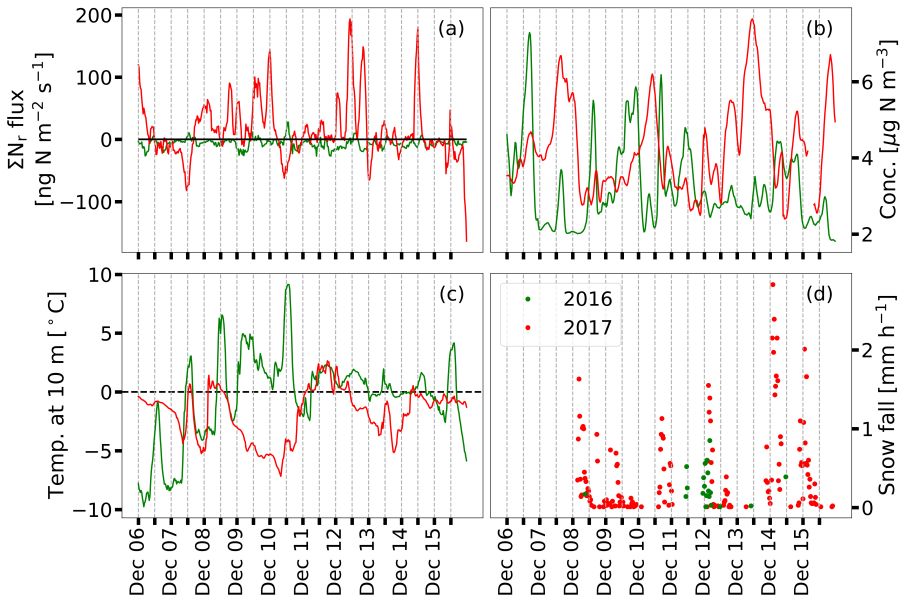

Figure R3: ΣN_r gap-filled fluxes (a), concentrations (b), air temperature at 10 m height above ground (c), and snow fall (d) from 6 December to 15 December for 2016 (green) and 2017 (red). Gaps are filled with the MDV approach with fluxes being in a range of ±3 days. Fluxes and concentrations of ΣN_r were smoothed with a 3-h-running mean for better visualization.

In 2017, we observed substantial snow fall and a slower varying temperature compared to 2016 leading to significant snow depths compared to 2016. On the 1st of December, 1 cm and 20 cm snow depth were measured in the fetch of the tower for 2016 and 2017, respectively. Two weeks later, snow depth increased to 5 cm and 60 cm, respectively. In addition, temperatures were mostly higher than 0°C in December 2016. In 2017, temperatures were mostly below 0°C and only for one day above 0°C, and global radiation was mostly below 100 W m^{-2}.

Hansen et al. (2013) reported a change in the NH_3 flux pattern from deposition to emission due to the senescing of fallen leaves. The decomposition of litter leading to NH_3 emissions from the forest ground could be responsible for the observed emission fluxes of ΣN_r although the decomposition rate of litter is reduced at lower temperatures. However, the snow pack could act as an insulator and inhibited soil frost penetration. Therefore, decomposition of litter could have been happened under the snow pack. Kreyling et al. (2013) compared different snow treatments and their effect on decomposition. The authors observed nearly no soil frost penetration under snow insulation. The annual cellulose decomposition was greatly reduced for the snow removal treatment ($\sim$ 46%). An increasing mass loss rate was found under a deeper snow pack (Saccone et al., 2013) depending on the type and age of litter (Bokhorst et al., 2013). Due to a small snow depth in 2016, soil frost penetration had a higher potential to reduce the decomposition rate. In addition, temperatures were mostly above the freezing point leading to partial melting of the snow layer, which probably inhibited the release of hygroscopic N_r species such as NH_3. Thus, emission of nitrogen from the soil or the decomposition of leaves was probably reduced compared to 2017. The deeper snow layer promoted microbial activity, and the generally lower temperatures and radiation inhibited a melting of the upper snow layers. Thus, leakage of N_r species like NH_3 could have happened in December 2017.

NO seems to be less responsible for the observed emission pattern following the findings of Medinets et al. (2016). They measured soil NO, N_2O, and CO_2 fluxes at a spruce forest during

the 'cold' season (daily average temperature < 3°C). They found that NO fluxes were positively correlated to air and soil temperature. Snow cover was not identified as a determining factor for the NO fluxes by the authors, since NO efflux during snow cover and snow free periods were similar. However, the reported snow depth was only 4.6 cm on average. Soil frost penetration could have happened in the topsoil and lowered the NO emissions leading to lower correlation between NO and snow cover. As stated by the authors, different results had been published about the origin of NO emissions from snow covered soils (see Medinets et al., 2016, and references therein). An influence of NO either emitted from the snow pack or the soil cannot be fully excluded. A correlation of the measured fluxes with temperature was not found. This could be related to a time-shift between emission and dropping temperature. It has also to be considered that we measure approximately 30 m above the forest soil, and not only NO contributes ΣN_r. In addition, NO emitted from the forest floor can be converted to NO_2. Thus, low correlations were expected."

Comment R1.38 *Line 508: Change "proposed" to "reported".*
Response to R1.38 Please note that the sentence was deleted.

Comment R1.39 *Line 511: Change "by DELTA" to "by the DELTA".*
Response to R1.39 Please note that the sentence was deleted.

Comment R1.40 *Line 528: Change "high" to "large".*
Response to R1.40 Please note that the sentence was deleted.

Comment R1.41 *Line 529: Change "at less" to "at a less".*
Response to R1.41 Please note that the sentence was deleted.

Comment R1.42 *Line 530: Change "It shows" to "These studies indicate"*
Response to R1.42 Please note that the sentence was deleted.

Comment R1.43 *Line 539: Please consider splitting up this long sentence for clarity.*
Response to R1.43 Please note that the sentence was deleted.

Comment R1.44 *Lines 544 – 547: The last two sentences of this paragraph seem more appropriate for the conclusions section.*
Response to R1.44 In principle, we agree that those lines would be more suitable for a conclusion section. However, we decided to leave out from the conclusions of this study.

Comment R1.45 *Section 4.2: In general this discussion would benefit from some reorganization and a more thorough quantitative evaluation of relationships between flux, micrometeorology, and air concentrations. The authors discuss radiation/photosynthesis, air concentration, dryness/RH/temperature, and precipitation as important variables. Perhaps these can be discussed in sequence, rather than jumping back and forth among them throughout the section, to make the discussion read more smoothly and to eliminate redundancies. For example, the role of air concentration is mentioned in numerous places, as are relative humidity and temperature. Some care should be given to revising this section as it will be of particular interest to readers seeking a better understanding of the processes driving Nr fluxes above forests.*
Response to R1.45 We appreciate the Reviewers' suggestions to Sec. 4.2. After carefully reflecting the Reviewer comments related to that section, we agree that this section needs to be

improved in readability and content. As outlined in the author comment, a discussion of the deposition velocity (v_d), the aerodynamic resistance (R_a), the boundary-layer resistance (R_b), and the effective canopy resistance ($R_{c,eff}$) of ΣN_r was added. The discussion related to the effect of precipitation on the ΣN_r exchange was deleted. For better readability, we separated the section in three subsections in which we discussed the effect of (global) radiation on ΣN_r exchange (Sec. 4.2.1), the effect of concentration v_d (Sec. 4.2.2), and the seasonal changes in the uptake capacity of ΣN_r (Sec. 4.2.3)

Comment R1.46 *Line 548: Section heading 4.2 only mentions micrometeorology but much of the following discussion involves the relationship between flux and air concentrations. Consider rewording.*
Response to R1.46 As the content of section changed, we adjusted the header as follows "Influence of micrometeorology and nitrogen concentrations on deposition and emission".

Comment R1.47 *Line 549: What is the proposed mechanistic relationship between Nr flux and global radiation? What about the diurnal pattern of turbulent mixing and its role in air-surface exchange?*
Response to R1.47 The following lines were added to line 551 of the manuscript: "As shown by Zöll et al. (2019), ΣN_r and CO_2 fluxes exhibited a similar daily cycle and showed a strong dependence on R_g during summer. The latter controls the opening of the stomata (Jarvis, 1976), i.e. lowers the stomatal resistance. Thus, photosynthesis controlling the CO_2 exchange through stomatal pathway appears to be the mechanism for controlling the ΣN_r exchange as compounds like NO_2 (Thone et al., 1996) or NH_3 (Wyers and Erisman, 1998) are taken up by the stomatal pathway, too. However, ΣN_r compounds are not willingly absorbed by the plants as seen by the light response curves of Zöll et al. (2019, Fig. 5). The light response curve of ΣN_r has a reversal instead of a saturation point as observed for CO_2 (Zöll et al., 2019). Consequently, a second mechanism, the stomatal compensation point firstly proposed by Farquhar et al. (1980) likely controls the uptake of the ΣN_r compounds. Basically, if the stomatal concentration is lower than the ambient concentration, deposition is observed. Thus, both parameters, the stomatal resistance and the stomatal compensation point, which are regulated by R_g and concentration, respectively, affect the uptake of ΣN_r. As further shown by Zöll et al. (2019), other parameters like u_* were not identified as important drivers for ΣN_r. Photochemistry and stomatal control appear to be more important than turbulent mixing. Radiation changes the composition of ΣN_r due to the formation of O_3. In addition, R_g had an influence on u_* as seen by their similar shapes in daily cycle (Fig. R5 and R6). The low correlations of ΣN_r fluxes to concentration for most of the selected u_* ranges show that atmospheric turbulence had a generally low influence on nitrogen deposition at the measurement site. Thus, u_* adds almost no additional information to the ΣN_r exchange and was not identified as important controlling factor for the ΣN_r exchange from July to September by Zöll et al. (2019). Similar conclusions can be drawn for temperature and relative humidity. They are also affected by light/energy input into the ecosystem and follow a similar diurnal pattern. It shows that R_g contains most of the information for the explanation of the ΣN_r fluxes."

It has to be noted that the study was conducted for ΣN_r at the same natural, unmanaged site from July to September. Micrometeorological parameters were controlled by natural processes. The low response to micrometeorological parameters may also related to other processes influencing the composition of ΣN_r, to opposing effects on N_r species, or effects happened on a shorter time scale such as molecular interactions between the ΣN_r compounds. R_g was not identified as

primary controlling factor for NH_3 by Milford et al. (2001). Milford et al. (2001) measured NH_3 fluxes above moorland, which has a generally higher humidity level than our measurement site. They concluded that NH_3 exchange is mostly driven by canopy temperature, wetness, and ambient concentrations. Radiation was not identified as primary controlling factor by the authors. They found higher deposition of NH_3 through the cuticular than through the stomatal pathway. However, Zöll et al. (2019) found only minor improvements in their driver analysis if water vapor pressure deficit was considered as secondary driver. Additionally, we found that v_d was reduced for high ambient humidity and wet leaf surfaces. Since we measured NH_3 indirectly by the TRANC and above an ecosystem characterized by lower humidity than a peatland, R_g favoring the exchange through the stomatal pathway appears to be more important for ΣN_r at the measurement site.

Comment R1.48 *Line 551: The authors discuss the relationship between air concentration and flux in several places in Section 4.2. Can the authors be a bit more quantitative in this analysis? What is the relationship (scatterplot) between concentration and flux if, for example, the dataset is filtered to include only mid-day fluxes (i.e., periods of high global radiation and friction velocity)? Is a clear relationship observed? What are the observed diurnal patterns in concentration? Do these patterns confound the relationship with global radiation mentioned in line 549? The authors should consider adding figures similar to figures 2 and 3 but for TRANC Nr concentration in supplemental material.*

Response to R1.48 We thank the Reviewer for his/her suggestion. We added plots similar to Fig. 2 and 3 but for the ΣN_r concentration to the supplement. A scatterplot showing the dependency of ΣN_r concentration on corresponding fluxes was added to Sec. 3.2. The following text was placed after line 348: "For visualizing the effect of turbulence on the fluxes, Fig. R4 shows the dependency of the measured fluxes on their concentrations for different u_* classes and global radiation (R_g) higher than 50 W m^{-2}.

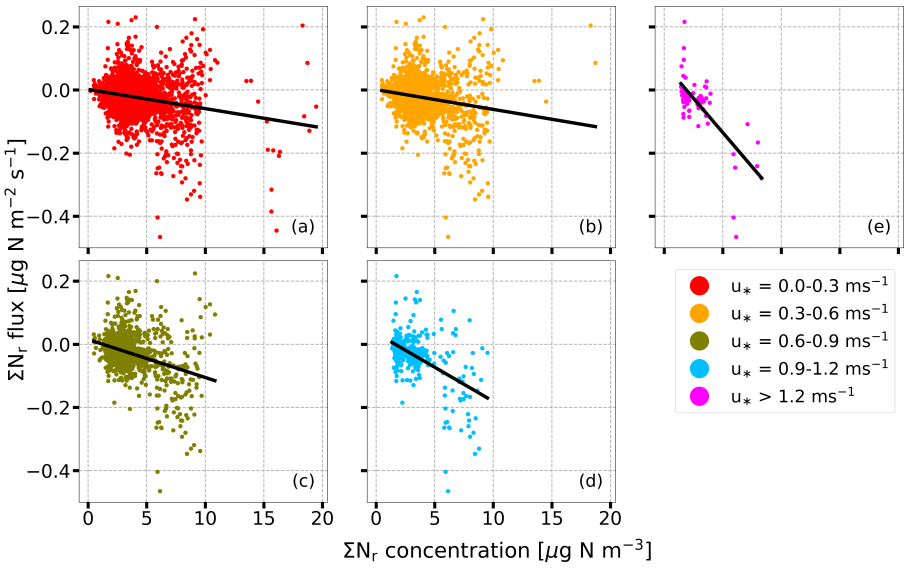

Figure R4: Dependency of measured concentrations on corresponding ΣN_r fluxes shown as scatter plots during daylight ($R_g > 50$ W m^{-2}). Colors indicate different u_* classes. Linear regressions between concentrations and fluxes are made for each u_* class indicated by black lines.

We found a decreasing slope with increasing u_*. The slope corresponds to v_d. Results of the linear regressions, v_d and squared correlations (R^2), are listed in Table R1. In addition, numbers

of half-hours used for the regressions are given.

Table R1: Results of linear regressions from Fig. R4 for selected u_* ranges. The slope of the linear function corresponds to v_d, R^2 is the squared correlation of concentrations and fluxes, and n is the number of half-hours used for the regression.

u_* range [m s^{-1}]	v_d [cm s^{-1}]	R^2 [-]	n [-]
0.0–0.3	0.61	0.07	9085
0.3–0.6	0.63	0.05	6124
0.6–0.9	1.20	0.14	2296
0.9–1.2	2.16	0.28	485
> 1.2	4.34	0.51	79

For u_* values lower than 0.6 m s^{-1}, v_d was almost invariant. For u_* values higher than 0.6 m s^{-1} or even higher, an increase in v_d was found. Since R_a (Garland, 1977) and R_b (Jensen and Hummelshøj, 1995, 1997) decrease with increasing u_*, v_d increases. The highest R^2 was determined for u_* higher than 1.2 m s^{-1}. For other u_* ranges, correlations were negligible. However, only 79 half-hourly concentrations and fluxes were available for u_* values higher than 1.2 m s^{-1}. Considering the number of half-hours, atmospheric turbulence had an influence on the deposition of $\Sigma\mathrm{N_r}$ but u_* could not be solely responsible for the observed exchange of $\Sigma\mathrm{N_r}$. ”

Figure R5 shows the daily cycle of concentration, $\mathrm{R_g}$, $\mathrm{u_*}$, air temperature ($\mathrm{T_{air}}$), and $\mathrm{v_d}$ for the period from May to September. Figure R6 is made for the same variables but for December, January, and February.

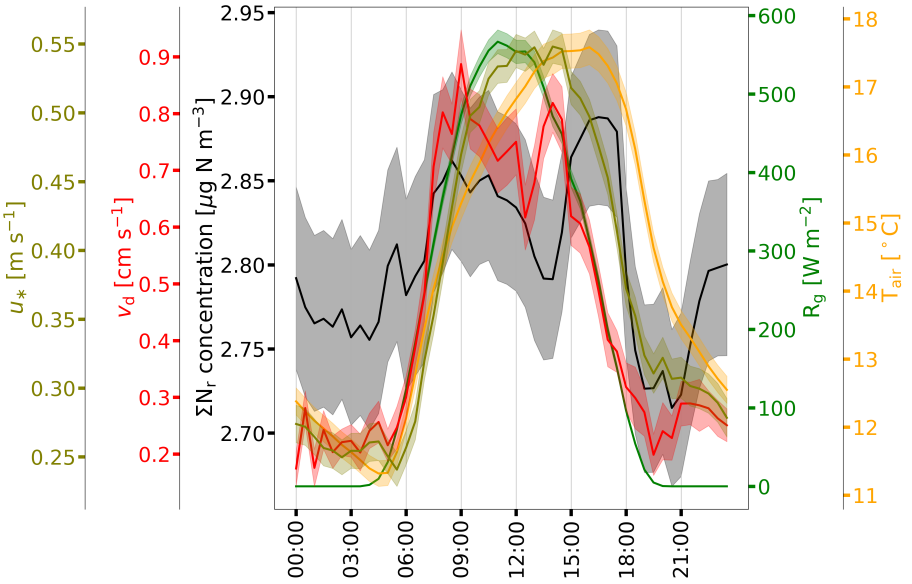

Figure R5: Daily cycle of $\Sigma\mathrm{N_r}$ (black) concentration, $\mathrm{R_g}$ (green), $\mathrm{u_*}$ (olive), air temperature $\mathrm{T_{air}}$ (orange), and $\mathrm{v_d}$ (red) for the period from May to September. Shaded areas represent the standard error of the mean.

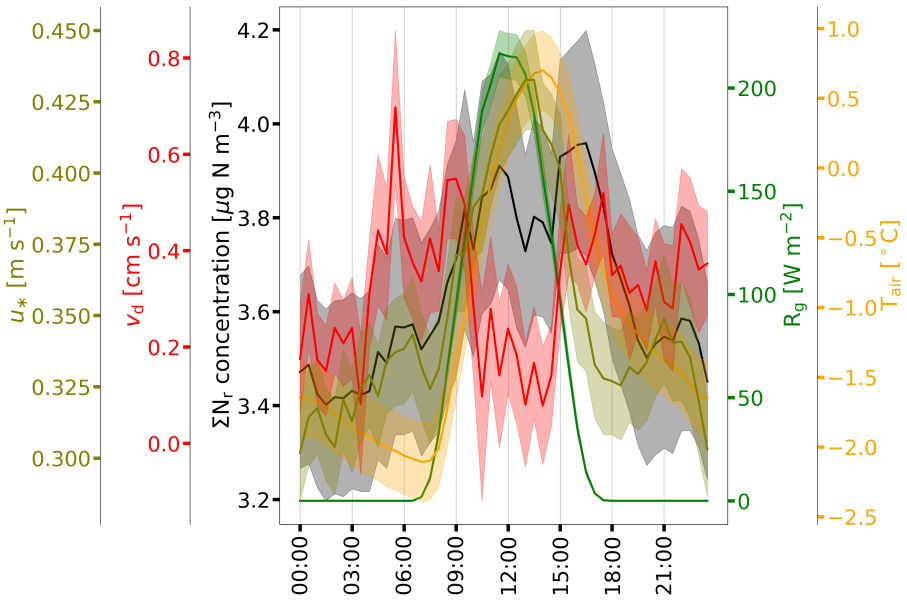

Figure R6: Daily cycle of ΣN_r (black) concentration, R_g (green), u_* (olive), air temperature T_{air} (orange), and v_d (red) for the period from May to September. Shaded areas represent the standard error of the mean.

From May to September, a clear diurnal pattern in v_d was observed with largest values around noon and lowest values during the night. During winter, v_d was almost equal and even lower during the day, which resulted in lower deposition of ΣN_r during winter. The different shapes of v_d were related to plant activity mainly controlled by R_g. Figures R5 and R6 were added to the supplement.

Comment R1.49 *Line 553: What do the authors mean by "favor" in this sentence?*
Response to R1.49 The word "favor" is confusing in this sentence. "influence" may be a more proper word. However, we decided to delete the sentence.

Comment R1.50 *Line 556: How is the last sentence in this paragraph justified by the preceding sentence? I must be missing something here.*
Response to R1.50 We agree that the last sentence is misleading. Please note the second paragraph of R1.47. We corrected the sentence with the information given there.

Comment R1.51 *Line 558: The authors compare March and April of 2017 and 2018 as an example of the potential role of photosynthesis in the interannual variability of fluxes. The explanation cites the role of temperature in stomatal function (and therefore the stomatal resistance) but what about the role of radiation? Are there differences in radiation between the two years that would also support this explanation?*
Response to R1.51 For March 2017 and 2018, we could not examine the reason for the slight difference median deposition. Micrometeorological conditions of both months were comparable. "The higher nitrogen deposition in April 2017 (Fig. 5) compared to April 2018 was mainly related to gaps in flux time series. In 2018, we had no flux measurements from mid of April to the beginning of May. During that time, foliage began in 2018 providing uptake of ΣN_r compounds. Increased plant activity was caused by continuously, high radiation values during daylight ($> 400\,W\ m^{-2}$) leading to higher temperatures in April 2018 ($\sim 11.0°C$) than in April 2017 ($\sim 6.0°C$). We further observed high NH_3 concentrations measured by passive samplers and the DELTA system for the

same time. Elevated NH_3 concentrations were likely caused by emissions from agricultural management in the surrounding region. In 2017, leaf emergence began in early May. Thus, measured N deposition would have been higher in April 2018 than a year before presumably related to a lower stomatal resistance in 2018. Almost equal patterns of v_d and $R_{c,eff}$ were determined for May 2017 and 2018. The conditions for uptake of ΣN_r by the canopy were comparable. Consequently, the different contributions in NH_3 and conditions in radiation and temperature strongly affected v_d and $R_{c,eff}$ and therewith the deposition of ΣN_r." The discussion of the April deposition was replaced by this response.

Comment R1.52 *Line 562: "...confirmed by the similar daily cycle for May 2017 and 2018." Similar daily cycle of what? Please specify.*
Response to R1.52 No deviations between the daily cycles of v_d and $R_{c,eff}$ were found. Thus, conditions for surface uptake were comparable. We implemented the explanation in the revised manuscript.

Comment R1.53 *Line 567: "Almost the same average...". This sentence is out of place relative to the rest of the paragraph. Please consider removing or consolidating with analysis of relative humidity and temperature in next paragraph.*
Response to R1.53 We agree that the sentence is out of place. The sentence was deleted.

Comment R1.54 *Line 571: The first sentence of this paragraph should either be removed or reworded. The use of "Therefore" implies a missing introductory sentence.*
Response to R1.54 We agree. Please note that the sentence was deleted.

Comment R1.55 *Line 572: What is the proposed mechanism by which dry conditions enhance Nr deposition? Are the authors proposing that the stomatal processes are a larger overall source of variability in the net canopy-scale flux than the cuticular processes? It is unclear from this paragraph, which seems to include multiple lines of analysis the connections or which are unclear as currently written. Please see my previous comment regarding the organization and clarity of section 4.2*
Response to R1.55 As suggested by Reviewer 2, the analysis on the parameters regulating the fluxes should be made for v_d and the $R_{c,eff}$. It shows if the association between fluxes and drivers is due to their effect on concentration or v_d. We further separated half-hours, which were influenced by precipitation, from the analysis since ΣN_r compounds like NH_3, HNO_3 and NH_4^+ are affected by rain. Thus, the entire paragraph (lines 571 - 598) was rewritten. Updated versions of Fig. 4 for v_d and $R_{c,eff}$ are shown in R2.2. Also, further details on R_a, R_b, and $R_{c,eff}$ are given in R2.2. The discussion regrading line 572 was modified as follows:

"Within the period of high incident radiation, in particular from May to September, a distinct diurnal pattern for v_d was observed, and no precipitation, high temperatures ($> 14.6°C$), low relative humidity ($< 74.0\%$), and dry leaf surfaces, were found to enhance the surface uptake, presumably through the stomatal pathway, of nitrogen during daylight. The observed differences in v_d for relative humidity and temperature were mostly related to R_a and R_b. $R_{c,eff}$ showed only a slight response to lower air humidity. Responses to the chosen temperature threshold and to dry leaf surfaces were not found.

During the rest of the year, no diurnal pattern was found under dry conditions (no precipitation) since stomata were likely closed, or requirements for stomatal deposition were not fulfilled (stomatal compensation point). Since we still observed a low, non-zero v_d but also short phases of ΣN_r

emission during seasons with lower radiation, cuticular, soil, and turbulent driven processes were likely to be responsible for the ΣN_r exchange. In periods of reduced plant activity, for instance in winter and autumn, the uptake through the stomatal pathway was greatly reduced or even inhibited due to reduced radiation or leaf area surfaces. Besides stomatal deposition, cuticular deposition is also an important pathway for ΣN_r compounds, which likely deposit on wet surfaces such as NH_3, HNO_3 or NH_4^+.

However, v_d was lower under wet conditions. Presumably, requirements for cuticular deposition were not fully met. Measurements of ΣN_r were conducted several kilometers away from nearby sources, and thus hydrophilic ΣN_r components could be washed out before air masses reached the site. We showed that the contribution and concentrations of N_r species, which can deposit on wet leaf surfaces, was comparatively low at the measurement site. Furthermore, those species were only indirectly measured, and wet leaf surfaces could be already saturated with water soluble N_r species leading to a high cuticular compensation point. These issues may reduce the cuticular contribution to exchange processes with the canopy. Presumably, cuticular deposition was probably not as important as stomatal deposition during periods of high incident radiation, in particular from May to September. Stomatal deposition seems to be more important than other in-canopy uptake processes for the ecosystem in close proximity to the measurement site for those months."

Comment R1.56 *Line 573: The sentence "Higher concentrations values lead to higher deposition values through the entire daily cycle." seems out of place. How does this statement relate to the preceding sentence?*
Response to R1.56 Higher concentrations of ΣN_r lead to a higher deposition as visualized by Panel (d) of Fig. 4. It is obvious that ΣN_r deposition scales approximately with its concentration since several components are included in the ΣN_r concentration signal. Thus, the statement was deleted in the revised version.

Comment R1.57 *Line 576: "Higher temperatures increase the opening size of the stomata leading to increased photosynthetic activity." What do the authors mean by "photosynthetic activity" in the context of the Nr fluxes?*
Response to R1.57 Higher temperatures lead to an increased plant activity and lower the stomatal resistance favoring ΣN_r deposition up to an optimum. As shown by (Wichink Kruit et al., 2010), stomatal conductance decreases with increasing temperature after reaching its maximum. Moreover, the maximal stomatal conductance depends on several parameters such as vegetation, RH, R_g, T, etc. (see Appendix E of Wichink Kruit et al., 2010). Please note that this sentence was deleted.

Comment R1.58 *"Thus we examined the influence of precipitation on fluxes." Would it not be more straightforward to compare fluxes during wet versus dry conditions as indicated by the leaf wetness sensors, perhaps binning by day versus night or air concentration to examine the relationship while controlling for other sources of variability? I'm not sure what precipitation rate in figure F1 is telling us about the relationship between flux and canopy wetness. Is the canopy any less wet (or leaf water layers thinner) after a prolonged 0.5 mm/h rainfall compared to short duration 5 mm/h rainfall? To clarify, are these flux measurements conducted during active precipitation? What is the quality of the EC fluxes during such periods? Please add another figure to F1 similar to plot b) but for the fluxes and include in discussion.*
Response to R1.58 We agree that a differentiation into precipitation classes was less useful. As written before, we did a reanalysis of Fig. 4 by separating fluxes, v_d, and $R_{c,eff}$ in dry and wet

classes. With the improved versions of Fig. 4, Fig. F1 added no additional information and was removed. The quality of fluxes measured during rain was almost similar to flux measurements with no measured precipitation. For example, 15% of the "wet" fluxes were classified as flag two following the Mauder and Foken flagging system (Mauder and Foken, 2006). Also, 15% of the "dry" fluxes were classified as flag two.

Comment R1.59 *Figure F1: Please begin the caption by describing plot a) rather than plot b).*
Response to R1.59 Please see the previous answer.

Comment R1.60 *Line 587: "It has to be considered that the catchment, in which the flux tower is located, has a size of approximately 0.69 km2 (Beudert and Breit, 2010) and is larger than the catchment of Wyers and Erisman (1998). Also, the surrounding forested area is much larger and the entire area is mountainous. The forest stand is relatively young since it is recovering from a bark beetle outbreak in the 1990s and 2000s (Beudert and Breit, 2014)." Please clarify how these statements are relevant to discussion of the relationship between surface wetness and flux.*
Response to R1.60 These statements were deleted since they add no relevant information to the discussion of surface wetness and flux.

Comment R1.61 *Line 592: "Presumably, if NH3 concentrations are low, Nr dry deposition seems to be favored by dry conditions." Please clarify how this conclusion follows from the analysis of the Wyers and Erisman (1998) and Woff et al (2010) studies. What would be the underlying leaf-level mechanism?*
Response to R1.61 We agree that this assumption needs further clarification. "Wyers and Erisman (1998) measured highest NH_3 deposition if the canopy has a high water storage level (CWS) ($> 2\,\mathrm{mm}$). The deposition efficiency was reduced if CWS was higher than $0.25\,\mathrm{mm}$ but lower than $2\,\mathrm{mm}$. By comparing different measurement years, they found differences in the deposition efficiency even if the canopy was saturated with water. They attributed the effect to the solubility of NH_3 in the water film. If canopy gets drier, evaporation of water occurs and the concentration of NH_3 increases in the water film. The cuticular resistance increases and deposition of NH_3 is reduced. Even emission of NH_3 was observed by Wyers and Erisman (1998), especially during the day when the canopy was dry, and NH_3 exchange was bidirectional. They showed that stomatal resistance was higher than canopy resistance. The authors identified cuticular deposition as more important for NH_3 as stomatal deposition. They measured an average NH_3 concentration of $5.2\,\mu\mathrm{g\ m^{-3}}$. We measured $0.65\,\mu\mathrm{g\ m^{-3}}$ on average and found that the contribution of NH_3 to ΣN_r was comparatively low at the measurement site. If contribution of NH_3 or other soluble N_r species to ΣN_r is comparatively low, cuticular deposition is most likely reduced under wet conditions. The authors proposed that even under low ambient humidity leaf surfaces can exhibit high humidity due to the accumulation of particles. In case of conifer needles, Burkhardt et al. (1995) showed that particles deposit close to their stomata. Most of them are hygroscopic. Therewith, cuticular deposition seems to be possible even under low ambient humidity. However, our measurement site was several kilometers away from potential (anthropogenic) emission sources. Concentrations of NO_3^-, NH_4^+, sulphur dioxide (SO_2), and NO_x were comparatively low at the site, in particular during summer. Thus, stomatal deposition appears to be more important for ΣN_r under high temperatures, low relative humidity, and no precipitation. This conclusion is valid for months with sufficient light/energy input leading to an increased plant activity, i.e. from May to September. Within the other seasons, aerosol concentrations originating from natural or

anthropogenic emission sources are probably higher resulting in a higher particle density on leaf surfaces promoting cuticular deposition.

Wolff et al. (2010) observed high deposition of tot-NH_4^+ and tot-NO_3^- during sunny days. During rain or fog, tot-NO_3^- exchange was almost neutral and emission was observed for tot-NH_4^+. They measured median concentration of 0.57, 0.12, 0.76, and 0.45 μg m^{-3} for NH_3, HNO_3, NH_4^+, and NO_3^-, respectively. For the September months, we measured average concentrations of 0.76, 0.46, 0.50, and 0.78 μg m^{-3} for NH_3, HNO_3, NH_4^+, and NO_3^-, respectively. Measured tot-NO_3^- and tot-NH_4^+ of Wolff et al. (2010) exhibited a higher particle than gaseous contribution. At our measurement site, the gaseous contribution was higher than the values reported by Wolff et al. (2010). Median deposition velocities of tot-NO_3^- and tot-NH_4^+ were higher than values measured for ΣN_r at our site, and they found that deposition was mainly driven by aerodynamic resistance rather than by surface resistance, in particular during periods of high radiation. It shows that changes in the contribution of N_r species to ΣN_r lead to different deposition pathways." The sentence was replaced by this response.

Comment R1.62 *Lines 595-598: It is unclear how the sentences on wet deposition relate to the rest of the paragraph. Please consider removing.*
Response to R1.62 We agree. The sentences were removed.

From R1.63 to R1.80, suggested modifications to the text and recommendations of the Reviewer are related to the modeling part and will be implemented in second manuscript.

Comment R1.63 *Line 609: "the implementation of Nr species like HNO3 is relatively straightforward compared to NH3" is out of place in this sentence. Consider removing.*
Response to R1.63 The sentence will be removed.

Comment R1.64 *Line 618: Change "uncertainties sources" to "sources of uncertainty".*
Response to R1.64 Agreed.

Comment R1.65 *Line 633: Change "much needed approach" to "much improved approach"*
Response to R1.65 Agreed.

Comment R1.66 *Line 663: "most of the studies.." Please indicate which studies the authors are referring to.*
Response to R1.66 Agreed. We consider to remove that sentence in the modeling study.

Comment R1.67 *Line 667: "and the inclusion of exchange mechanisms for NO3 and NH4 should be considered in-situ modeling approaches." Please clarify what is meant here.*
Response to R1.67 Currently, deposition of NO_3^- and NH_4^- is not included in DEPAC-1D. We will include particle deposition in DEPAC-1D for the modeling study. DELTA measurements will be used as input data.

Comment R1.68 *Line 671: As a general question, how well does the DEPAC total Nr flux reflect the relationships between measured TRANC Nr flux and radiation, temperature/RH/dryness described in section 4.2?*
Response to R1.68 We appreciate the Reviewer's comment. This will be part of the modeling

study and compared with results from the TRANC-CLD system. A similar analysis to Fig. R7 and R8 will be made for DEPAC-1D.

Comment R1.69 *Line 682: And at sites with sparse vegetation.*
Response to R1.69 Will be added to end of the sentence.

Comment R1.70 *Line 685: Change "almost similar" to "similar".*
Response to R1.70 Agreed.

Comment R1.71 *Line 688: Has VDI been explained/defined?*
Response to R1.71 "Verein deutscher Ingenieure" (Association of German Engineers) is missing.

Comment R1.72 *Line 689-690: The two sentences here related to NH3 should be move to the preceding paragraph.*
Response to R1.72 We will move the sentences to the preceding paragraph.

Comment R1.73 *Line 696: The use of "positive" to describe the deposition velocity is not necessary.*
Response to R1.73 Agreed.

Comment R1.74 *Line 712: Why is CBT mentioned here in the discussion of LOTOS-EUROS?*
Response to R1.74 The sentence seems out of place here and will be deleted.

Comment R1.75 *Line 720: As previously mentioned, a summary and comparison of the various measurement techniques would be helpful to this discussion. Could the authors add a table summarizing the statistics of QCL, DELTA, and passive measurements, along with the LOTOS-EUROS predictions, as supplemental material? How well did the measurement techniques agree?*
Response to R1.75 We will add a figure similar to Fig. R9 but for the LOTOS-EUROS concentrations and a figure similar to Fig. R13 with NH_3 from LOTOS-EUROS.

Comment R1.76 *Line 722: "The difference to LOTOS-EUROS NH3 concentrations was highest during periods with significant amount of NH3 in the atmosphere like in spring and autumn, which is caused by emissions from fertilizer leading to a high load of modeled concentrations." Please reword this sentence, avoiding the use of "like" and "load".*
Response to R1.76 Agreed.

Comment R1.77 *Line 726: I encourage the authors to revisit the point and usefulness of this paragraph. As written I can't see that it adds anything to the discussion.*
Response to R1.77 A reduction in grid cell size may lead to improvements in the localization of the emission sources. In close proximity to the flux tower, only a few emission sources were located. Thus, a reduction of the size may reduce the modeled concentrations of grid cell, in which the measurement site was located. We will modify the paragraph accordingly.

Comment R1.78 *Line 760: "The deposition event in February 2018 seen by the TRANC seems to be driven by particulate Nr." Do the DELTA measurements reflect higher NH4+ and NO3- concentrations during this period compared to other months? These data should be presented.*
Response to R1.78 Please see R1.20.

Comment R1.79 *Line 775: The details here (i.e., "were selected from a matured tree stand") highlight that more information is needed in the method section regarding CBT as it was specifically applied at this site.*
Response to R1.79 Further information about the tree stand will be added to the description of the CBT approach. The description will be shifted to the modeling manuscript.

Comment R1.80 *Line 783: And to CBT.*
Response to R1.80 Will be added.

Comment R1.81 *Line 779: Conclusions section. Much of the information contained in this section is a direct recap of the preceding results and discussions. The length of this section could be significantly reduced.*
Response to R1.81 Due to the separation into two studies, the length of the conclusion of the revised manuscript was reduced. We stated the conclusions more precisely.

Response to Reviewer 2

General Comments *The paper presents a 2.5-year long dataset of dry deposition of total reactive nitrogen (Nr) to a forest site, interpretation of the results in the light of measurements of Nr speciation, and a comparison of the results with alternative approaches: the prediction of a chemistry and transport model, a site-specific inferential model and a canopy budget technique. Direct measurements of Nr dry deposition is rare and such a long dataset of Nr dry deposition measurements to forest is unique and important, and thus generally publishable in Biogeosciences. I had high hopes for this paper, especially because the Nr flux measurements were accompanied by NH3 flux measurements (by QCL), which I hoped would have been used to elucidate the non-NH3 component of the Nr flux. However, I was let down in various aspects: the NH3 fluxes are not used in this paper (only concentrations). It is not stated whether they just did not work or whether they are left for another paper. However, this paper speculates a lot about the nature of the NH3 exchange and its impact on the total Nr flux and with NH3 flux data presumably available to explore this explicitly, this seems rather odd. In addition, the Discussion section is quite long and lacks structure and aim. The advantage of the TRANC is that it captures most of the Nr flux with one instrument. The disadvantage is that it does not shed light on the behaviour of the individual Nr components. Yet, much of the discussion is dedicated to relating the measured flux to the behaviour of individual compounds reported in the literature. I do not think this adds to the manuscript and should be shortened. Instead the paper should be more focussed on describing the flux in its totality. For example, the Nr dry deposition budget is not discussed in the context of the additional wet deposition which could be taken either from nearby measurements (if available) or the LOTOS-EUROS prediction. A number of serious concerns need to be addressed as raised below before the manuscript can be accepted for publication. This will require significant reworking and refocussing of the manuscript.*

We thank the Reviewer for his/her comments and criticism on this work. The determination of the NH_3 fluxes with the eddy-covariance method was not possible (see R2.1). If NH_3 fluxes by the QCL were available, an investigation of the non-NH_3 component would be included in the manuscript. Up to now, publications about flux measurements of ΣN_r are rare. Thus, we have not much comparison possibilities in case of ΣN_r. The discussion was extended to individual ΣN_r compounds in order to show that the flux magnitude of the individual compounds is in agreement with our measurements for similar ecosystems. However, we agree that the discussion on that topic was too long and can be shortened. We deleted lines 485-545 and shortened the discussion on individual ΣN_r compounds substantially. We plan to shift the discussion of the individual ΣN_r compounds to the modeling manuscript. As stated in the author comment, we made an analysis on v_d and $R_{c,eff}$ and determined the total nitrogen budget. We included measurements of wet deposition taken close to the tower by bulk and wet-only samplers and investigated the influence of micrometeorology on the nitrogen dry deposition sums using data-driven gap-filling methods.

In the modeling study, individual flux components of DEPAC-1D will be compared to values reported in literature. As done for the TRANC-CLD measurements, an analysis of the micrometeorological parameters will be made for DEPAC-1D. The discussion of the dry deposition budgets will be improved, and wet deposition estimates from LOTOS-EUROS will be included. We will discuss the ecological impact of nitrogen deposition on forest ecosystems. A comparison to annual N budgets reported for other forest ecosystems will be carried out. We addressed all mentioned points related to the flux measurements and implemented your suggestions in the revised manuscript. Since we made a separation of the modeling part, a detailed reply to the ΣN_r modeling results will

not be made yet.

Main scientific comments

Comment R2.1 *As mentioned above, if the NH3 fluxes could be worked into the manuscript this would strengthen the analysis a lot.*
Response to R2.1 As stated above, an evaluation of NH_3 fluxes with the eddy-covariance method was not possible. We added the following lines to the end of Sec. 2.3: "As outlined in Sec. 2.2, measurements of NH_3 were made with a QCL at high temporal resolution. In combination with the sonic anemometer, it gives the opportunity to determine NH_3 fluxes and to further investigate the non-NH_3 component of the ΣN_r flux. However, a calculation of the NH_3 fluxes with the EC method was not possible in this study. No consistent NH_3 time lag was found making flux evaluation impossible. Due to regular pump maintenance, cleaning of the inlet and absorption cell, issues related to the setup of the QCL were unlikely to be the cause. We suppose that the variability in the measured NH_3 concentrations was not sufficiently detectable by the instrument. Significant short-term variability in the ΣN_r raw concentrations were not found in the NH_3 signal even in spring or summer. Thus, no robust time lag estimation could be applied to the vertical wind component of the sonic anemometer and the NH_3 concentration. Recently, Ferrara et al. (2021) found large uncertainties for low NH_3 fluxes measured with the same QCL model. Cross-covariance functions had a low signal-to noise ratio indicating that most of the fluxes were close to the detection limit."

Comment R2.2 *The paper confuses the rate of deposition (deposition velocity) and the actual deposition. Ignoring the effects of compensation points on NH3 exchange and the contribution of soil NO emissions to the net flux of NO and NO2, and also changes in the relative contribution of different compounds to Nr, the deposition of Nr is expected to scale approximately with its concentration. This is trivial and fundamentally also the way the deposition is calculated in LOTOS-EUROS and DEPAC-1D. Changes in concentration therefore mask the mechanisms that regulate the deposition rate. Thus, the analysis would be much more meaningful if the authors examined the controls of the deposition velocity rather than of the flux. This is what is done in the literature for the different compounds and, currently, comparisons are not correct. For example, it is stated that NH3 fluxes are largest under wet conditions. In fact most studies report that Vd is larger for wet conditions, but at the same time the concentration may be reduced. For this reason statements like "dry conditions seem to favour nitrogen dry deposition (line 13, also line 793f)" are maybe not incorrect, but certainly misleading. Throughout the analysis it is rarely clear whether an association between the flux and drivers is due to their effect on concentration or Vd. For example, Fig. 4 would be more meaningful if presented for Vd. In fact, an analysis in terms of Rc would be even more meaningful as it would normalise for the effect of turbulence on Ra and Rb both of which contribute to Vd. Because particles are not really subject to a boundary-layer resistance in the way it is applied to gases, Rc is not really meaningful. However, the analysis could be done in terms of Vds = Vd(z0), i.e. after normalising at least for Ra.*
Response to R2.2 The ΣN_r compounds have different exchange pattern and differ in their interaction and reaction pathways. Thus, it is difficult to show one deposition velocity for ΣN_r. However, we agree that the manuscript benefits from an analysis of v_d in order to show if an influence of a driver on the flux is due to its effect on v_d or concentration. Figure R7 was done in accordance to Fig. 4 in the manuscript but for v_d.

We further determined the aerodynamic resistance (R_a) following Garland (1977) and the boundary-layer resistance (R_b) following Jensen and Hummelshøj (1995, 1997). R_b requires a

molecular diffusion coefficient of ΣN_r. We determined the molecular diffusion coefficient for ΣN_r as the weighted average of the campaign-wise averages of HNO_3, NH_3, NO, and NO_2 multiplied with their individual molecular diffusivities adapted from Massman (1998) and Durham and Stockburger (1986). The effective canopy resistance $R_{c,eff}$ was determined by subtracting the maximum deposition velocity allowed by turbulence from the measured deposition velocity. In Sec. 2.4 "Determining deposition velocity and canopy resistance of ΣN_r from measurements", equations needed for the calculation of v_d and resistances are given. Figure 4 and the corresponding description (lines 353-360) were deleted and replaced by the figures and text shown in this response.

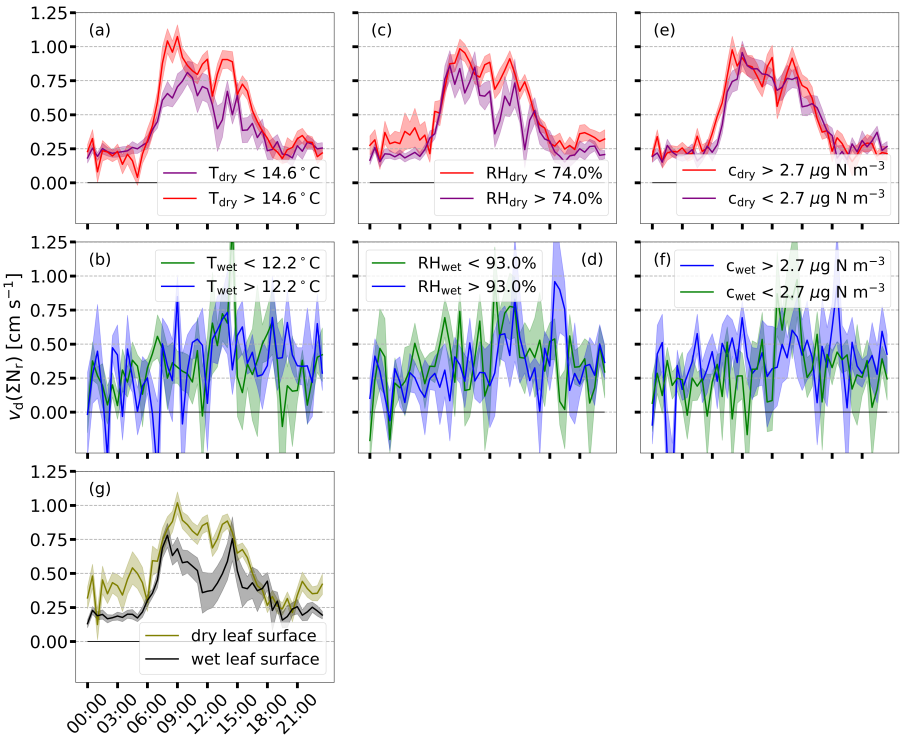

Figure R7: Mean daily cycle from May to September of v_d for low and high temperature, relative humidity, and concentration separated by precipitation in the conditions "dry" and "wet". Panel (a), (c), and (e) represent the case dry (no precipitation), (b), (d), and (f) the case wet. Median values of temperature, humidity, and concentration, which are derived for the same time period, are used as threshold values for separating v_d. In panel (g), the mean daily cycle of v_d for dry and wet leaf surfaces is shown. For classifying leaf surfaces as dry or wet, the scheme proposed in Sec. 2.2 is applied. The shaded areas represent the standard error of the mean.

"In general, higher temperatures, less humidity, dry leaf surfaces, and dry conditions (no precipitation) enhanced deposition of ΣN_r, and a clear diurnal pattern was observed for v_d with high values around noon and low, non-zero values in the night during dry conditions. During dawn/nighttime, deposition velocities exhibited no significant difference between the applied thresholds. Overall, no difference was found for low and high concentration regimes. In case of precipitation, v_d was reduced during daytime and exhibited a high variability for the entire day. No difference and distinct pattern could be found for low and high temperature, humidity, and concentration regimes during precipitation. During other times of the year, no diurnal pattern was observed during dry conditions. In those periods, v_d was almost constant and exhibited lower values during daylight compared to the May to September time frame. Occasionally, negative

deposition velocities referring to emission of ΣN_r were recorded during times of lower radiation. Figure R8 is in accordance to Fig. R7 but for $R_{c,eff}$.

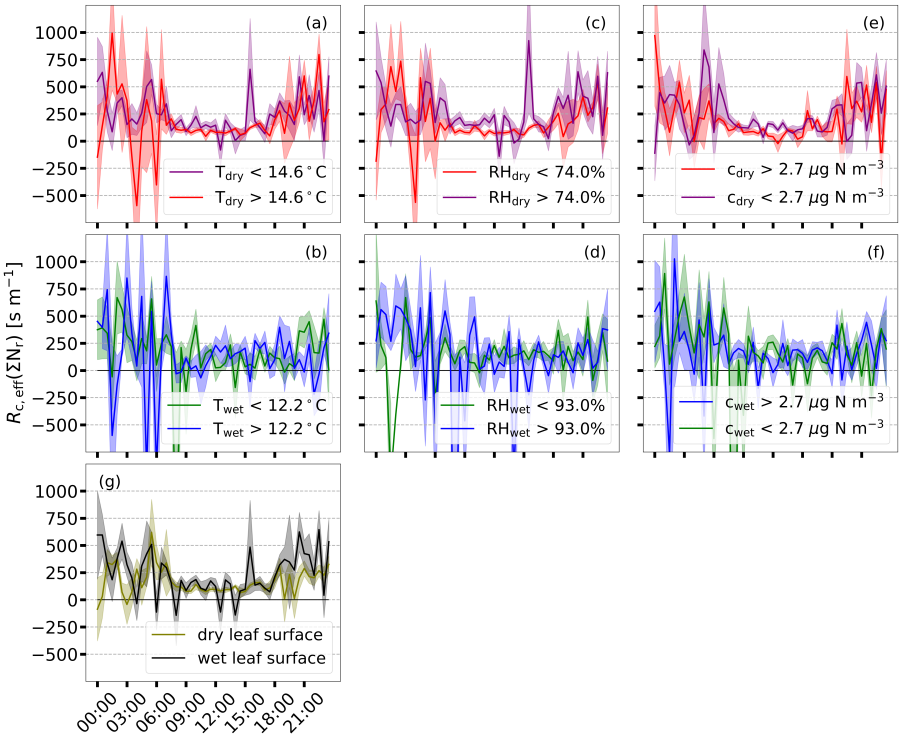

Figure R8: Mean daily cycle from May to September of $R_{c,eff}$ for low and high temperature, relative humidity, and concentration separated by precipitation in the conditions "dry" and "wet". Panel (a), (c), and (e) represent the case dry (no precipitation), (b), (d), and (f) the case wet. Median values of temperature, humidity, and concentration, which are derived for the same time period, are used as threshold values for separating $R_{c,eff}$. In panel (g), the mean daily cycle of $R_{c,eff}$ for dry and wet leaf surfaces is shown. For classifying leaf surfaces as dry or wet, the scheme proposed in Sec. 2.2 is applied. The shaded area represents the standard error of the mean.

$R_{c,eff}$ exhibited lowest values during the day and highest values at night. During nighttime, the variability in $R_{c,eff}$ was enhanced whereas $R_{c,eff}$ was almost stable during daylight. Only slight differences between the applied threshold were found. $R_{c,eff}$ was slightly lower at higher concentrations only for short periods during daylight, for example around noon. In case of relative humidity, $R_{c,eff}$ exhibited slightly lower values for less humid air. Temperature had nearly no effect on $R_{c,eff}$. During precipitation, no difference between the applied thresholds was found. Similar to v_d, $R_{c,eff}$ had a higher variability compared to dry conditions during the day resulting in higher uncertainties. Also phases with negative $R_{c,eff}$ values were observed during rain indicating emission of nitrogen from the canopy. A similar analysis was made for R_a and R_b. During daylight, values of R_a and R_b were close to zero showing that v_d was mostly driven by the pattern of $R_{c,eff}$. Lower values of R_a and R_b were found for lower air humidity and higher temperature. In case of wet leaf surfaces, R_a and R_b were higher in the morning and evening. If wet leaf surfaces were excluded from the analysis, the differences for v_d and resistances to micrometeorological parameters diminished. Wet leaf surfaces reduced the uptake of ΣN_r at the measurement site. During the night or at lower radiation, R_a and R_b were comparable in magnitude to $R_{c,eff}$. In autumn and winter, $R_{c,eff}$ showed partly negative values and no diurnal pattern. It should be noted that the shapes of the daily

cycles of each parameter shown in Fig. R7 and R8 are almost similar for the chosen threshold values and differ only in amplitude."

Due to the focus on v_d and resistances, the interpretation of the results had to be rewritten. Please see also R1.47, R1.48 R1.50, R1.51, R1.55, and R1.61. For the second part of this study, we plan to add a discussion of resistances and v_d calculated from the TRANC measurements compared to the results from LOTOS-EUROS and DEPAC-1D. Additionally, the investigation on micrometeorological controls will be applied to ΣN_r fluxes modeled by DEPAC-1D.

Comment R2.3 *The interpretation of the measurements is not helped by the lack of showing absolute concentrations. The relative composition of total Nr (Figs. B1 and E1) is useful, but also the absolute concentrations are needed to interpret the results. Again, because fluxes are discussed in terms of their magnitude and not their Vd the reader is left wondering whether whether it is really the change in relative composition that changes the flux or whether it is just the overall Nr concentration. By the way, it is unclear what time periods are shown by each pie chart and what frequency this maximum refers to (Caption and text Line 305ff). Presumably, these are monthly results given that the lowest data resolution (from the DELTA) is monthly? Indeed, I would find a figure showing monthly stacked bar graphs of the individual Nr components very useful. This would convey how the total and their contribution to total Nr changed seasonally. Also, an assessment of how well the sum of the individual N compounds compares with the total Nr concentration needs to be added as quality control.*

Response to R2.3 We agree that a comparison of the absolute concentration values is helpful for interpreting differences in the flux pattern. The pie chart (c) covers the entire measurement period of the DELTA system. (a) and (b) show a pie chart with the lowest and highest concentration of TRANC ΣN_r during the exposition periods of the DELTA samplers. Yes, the underlying time resolution is approximately monthly since the denuder were exchanged nearly every month. By the comparison of the absolute values, we found that the zero-air calibration value of the TRANC-CLD system was incorrect from July to September 2017 and from March to mid of May 2018 by approximately $0.9\,\mu$g N m^{-3} compared to the uncorrected TRANC-CLD concentrations. Concentrations and fluxes were recalculated with the bias correction. Figures shown in the response are made with the bias-corrected data. In the revised version, Figs. B1 and E1 were deleted since we found no significant deviations of the minimum and maximum TRANC ΣN_r cases to average after the bias correction. The following lines including the figures were added to Sec. 3.1 after line 302.

"The comparison of the TRANC with DELTA+NO_x revealed slight overestimations by the latter from August 2016 to October 2016 and from January to March 2017. On average, an underestimation by DELTA+NO_x of approximately $0.3\,\mu$g N m^{-3} with a standard deviation of $0.7\,\mu$g N m^{-3} was observed. The median value was about $0.35\,\mu$g N m^{-3}.

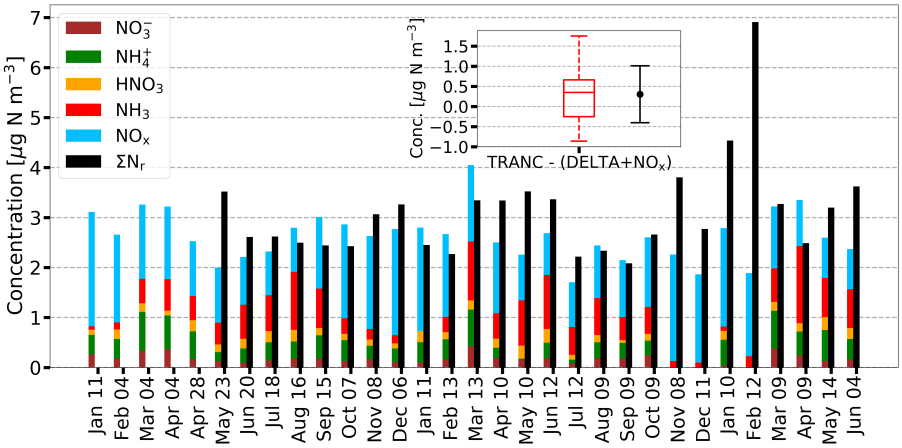

Figure R9: Monthly stacked concentration of TRANC, DELTA, and NO_x in μg N m^{-3} for the entire measurement campaign. Missing NH_3 measurements from the DELTA measurements caused by a low pump flow were filled with passive sampler values from 30 m. Replacing was done for December 2016 and 2017, January 2017, November 2017, and from February to April 2018. Gaps in the time series of the individual components were replaced by monthly averages estimated from other years if possible. NO_x and ΣN_r were averaged to the exposition periods of the DELTA samplers.

HNO_3, NH_4^+, and NO_3^- concentrations were nearly equal through the entire measurement campaign. Seasonal differences existed mainly for NH_3 and NO_x. We measured average concentrations of 0.56, 0.17, 0.40, 0.19, and 1.40 μg N m^{-3} for NH_3, HNO_3, NH_4^+, NO_3^-, and NO_x for the entire campaign, respectively. On average, the relative contribution of NH_3, HNO_3, NH_4^+, and NO_3^- to ΣN_r was less than 50% for the entire measurement campaign as visualized by Fig. R10. We further observed a low particle contribution to the ΣN_r concentrations ($\sim$ 22% on average) showing that the ΣN_r concentration pattern was mainly influenced by gaseous N_r compounds.

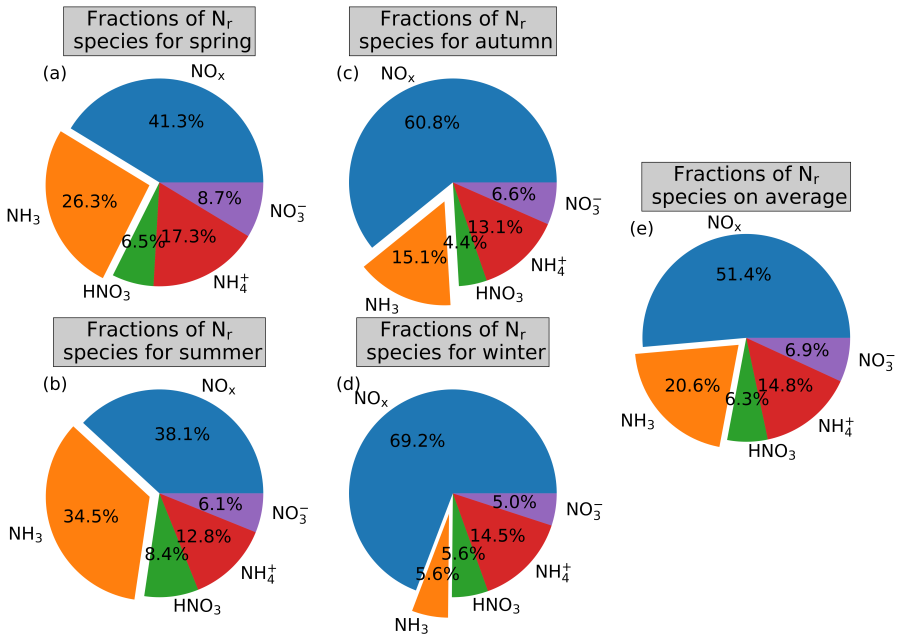

Figure R10: Pie charts showing the relative contribution of concentrations for NO_x, NH_3, NO_3^-, NH_4^+, and HNO_3 to ΣN_r based on DELTA samplers and NO_x measurements for different seasons of the year. NO_x measurements are averaged to exposition periods of the DELTA samplers. (a) to (d) refer to spring, summer, autumn, and winter, respectively. (e) shows the average relative contribution to ΣN_r for the entire measurement period.

In general, NO_x showed the highest contribution to ΣN_r and followed seasonal changes with highest values during winter and lowest values in summer. NH_3 showed also seasonal changes with concentrations lowest in winter and highest values in spring and summer. The contribution of HNO_3 was almost stable. A slight increase in the contribution was found for summer. As reported by Tang et al. (2020), HONO sticks to carbonate coated denuder surfaces, which are designed for collecting HNO_3. Thus, HNO_3 concentrations may be biased. NO_3^- and NH_4^+ exhibited slightly higher values for spring. Only small seasonal changes in the overall ΣN_r concentration were observed. As seen by Fig.R9, ΣN_r concentrations were mostly between 2 and 4 μg N m^{-3}. We measured 3.3, 2.6, 2.5, and 3 μg N m^{-3} with the TRANC system for spring, summer, autumn, and winter, respectively."

The comparison of the total N concentrations shows that the TRANC can adequately measure ΣN_r concentration. Obviously, not all components of ΣN_r were included in this comparison, for example, higher oxidized components like N_2O_5 could not be considered. As mentioned in Sec. 2.2, NO_2 had been measured at 50 m. However, Seok et al. (2013) found only slight differences in NO_2 concentrations above the canopy at a remote site. Thus, height differences in NO_2 are likely insignificant. Issues in the temperature stability or CO supply resulting in instabilities in the conversion efficiency of the TRANC, or a reduced sensitivity of the CLD could lead to differences to DELTA+NO_x. DELTA measurements report concentrations integrated over long time periods. 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. Please note the changes to the discussion of the DELTA results shown in Sec 4.1.

The changes in the composition of ΣN_r were also affecting v_d. Only slight seasonal changes in the overall ΣN_r concentration were observed. We measured 3.3, 2.6, 2.5, and 3 μg N m^{-3} for

spring, summer, autumn, and winter, respectively. Consequently, it was not only the change in the overall ΣN_r concentration that influenced v_d. Please note the new subsection 4.2.2 "Influence of N_r species on measured v_d."

Comment R2.4 *The measurements are compared to those made over other ecosystems and differences are explained by differences in ecosystems. Again, this is only part of the story, mainly the part that affects Vd. The pollution climate the ecosystem is in is equally important and does not necessarily correlate with the ecosystem type (think of an urbanwoodland or a heavily grazed pasture in otherwise pristine environment). The comparison needs to be reworded. Generalisation that Nr fluxes always behave above natural vegetation as they do at this particular site is not tenable (e.g. line 13 and throughout).*
Response to R2.4 We appreciate your comment and corrected the corresponding comparisons, e.g. line 458.

Comment R2.5 *The analysis of the effect of precipitation on the flux (Fig. F1a and associated text) is particularly problematic. During rain the eddy-covariance flux measurement of water soluble compounds (and many Nr compounds are) is highly uncertain because fluxes cannot be assumed to be constant with height due to the washout process. An increased Vd during rain may just reflect the presence of an additional sink (the washout process) below the measurement height. Rain episodes should potentially be filtered out, but certainly no process understanding should be derived from data taken during rain. How do the measurements demonstrate that wet deposition is important (Line 595)?*
Response to R2.5 Based on the suggestions of Reviewer 1 and your comment, we removed the corresponding text (lines 584-598) and Fig. F1. Yes, we agree that rainy episodes should be filtered out since water soluble N_r such as NH_3, HNO_3, and NH_4^+ were probably washed out from air masses before reaching the measurement height. As written before, we did a reanalysis of Fig. 4 by separating fluxes, v_d, and $R_{c,eff}$ in dry and wet classes. Please note the responses to comments 1.55 and 1.58 to 1.62. The sentence "It shows that wet deposition is important for the uptake of ΣN_r compounds at our measurement site." was certainly misleading and deleted. Wet deposition samplers were in close proximity to the flux tower. Thus, wash out processes also affected wet deposition measurements.

Comment R2.6 *The paper does not distinguish different types of error (e.g. lines 617f and 652f). The flux error according to Finkelstein and Sims describes a random error, whereas the uncertainty in the DEPAC-1D estimate is more likely to be systematic and thus provide a bias. The input parameters are considered the largest uncertainty in DEPAC-1D (lines 619f), but actually different inferential models give very different results which highlights their uncertainty (e.g. Flechard et al., 2011).*
Response to R2.6 The mixture of the different error types was not intended. In the revised version of the measurement part, the flux uncertainty of the gap-filled fluxes was calculated as the standard error of mean. The random uncertainty following Finkelstein and Sims (2001) was included in the discussion. Total uncertainty from random error estimates was calculated as square root of the sum of the squared random uncertainties according to Pastorello et al. (2020). Please note the substantial changes to Sec. 4.3, which was renamed to "Uncertainties in dry deposition estimates". The uncertainty discussion of DEPAC-1D and LOTOS-EUROS was deleted and will be moved to the modeling study and substantially improved.

Comment R2.7 *This then also relates to an apparent contradiction between the discussion of the importance of stomatal exchange (Line 575) which is temperature dependent but mainly regulated by PAR and the statement that the canopy resistance is mainly driven by water solubility (Line 702).*
Response to R2.7 We thank the reviewer for his/her hint to this contradiction. We improved the discussion on the uptake capacity of ΣN_r. Please note the new subsection 4.2.3 "Seasonal changes in ΣN_r uptake capacity". The information related to the modeling part will be improved accordingly.

Comment R2.8 *Still on the topic of drivers of the exchange, a similarity in the diurnal cycle between global radiation and flux is no proof of causality (line 549ff). A lot of parameters are driven by the radiation: turbulence, photochemistry etc.. Neural networks also do not derive causalities or 'drivers', only associations and determinants.*
Response to R2.8 We appreciate your comment and reworded the corresponding lines. Please note R1.47 and the description to Fig. R5 and R6. Zöll et al. (2019) showed that global radiation and concentration added independent information to the variability of the ΣN_r fluxes. Adding other parameters like temperature, u_*, or CO_2 as secondary driver resulted in lower values if global radiation was chosen as primary driver. Their investigation revealed that global radiation contained important information for the explanation of the ΣN_r fluxes. The word 'driver' is a paraphrase of the expression controlling input variable (Moffat et al., 2010). Drivers are identified by their correlation with the flux. In general, correlations could also be influenced by other parameters, which have not or could not considered by Zöll et al. (2019), for example chemical interactions of components contributing to ΣN_r. We agree that the word driver could be misinterpreted without proper explanation. We implemented the explanations given in this response to line 551.

Comment R2.9 *The filtering criteria will have removed preferentially the smaller fluxes during low turbulence conditions and the remaining dataset will therefore be biased. Whilst this is not an issue if a model is used for gap filling that accounts for changes in turbulence, it does impact the straight averages of the fluxes (Figure 2) the value of which then becomes questionable and also the MDV gap filling method. These issues and implications need to be discussed.*
Response to R2.9 Yes, the application of the filtering criteria like Mauder and Foken or a friction velocity threshold could preferentially remove smaller fluxes, which occurred at night-time. Therefore, we introduced a new section "Sensitivity of ΣN_r dry deposition sums to micrometeorological parameters". In this chapter beginning at line 362, we investigated possible dependencies of the ΣN_r dry deposition sum on micrometeorological parameters if data-driven gap-filling methods like the Mean-Diurnal-Variation (MDV) method were used. Text and figures of this response were added to this chapter. We further calculated total annual depositions by using wet depositions measurements from wet-only samplers. Details about the wet deposition measurements were added to line 174.

Figure R11 shows the non gap-filled ΣN_r fluxes depicted as box plots and their cumulative sums with and without a u_*-filter if MDV is used as gap-filling approach. The threshold was set to 0.1 m s^{-1}, and the window for filling each gap was set to $\pm$5 days. Uncertainties of the gap-filled fluxes were estimated by the standard error of the mean. The total uncertainties were calculated as the sum of the standard errors.

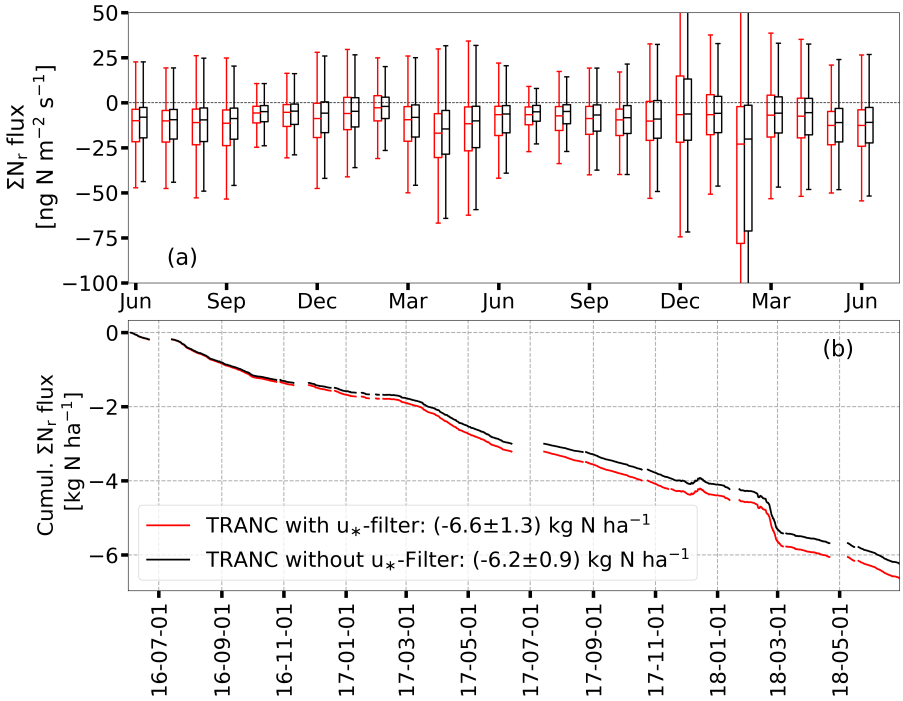

Figure R11: Panel (a) shows the non-gap filled ΣN_r fluxes depicted as box plots with (red) and without (black) u_*-filter in ng N m^{-2} s^{-1} (box frame = 25% to 75% interquartile ranges (IQR), bold line = median, whisker = 1.5· IQR). The threshold for u_* was set to 0.1 m s^{-1}. In panel (b), the cumulative dry deposition of ΣN_r is plotted for both cases in kg N ha^{-1}. For determining the cumulative curves, MDV was used as gap-filling method, and gaps were filled with fluxes being in a range of ±5 days. Remaining gaps were not filled.

The difference in dry deposition was approximately 400 g N ha^{-1} after 2 years and is within the uncertainty range of the estimated dry depositions. Panel (a) of Fig. R11 shows that median depositions of the ΣN_r fluxes with u_*-filter were almost equal to or larger than the median depositions without u_*-filter. Figure R4 indicates that we measured large and small fluxes below 0.1 m s^{-1}. Thus, the applied u_* threshold removed not only small fluxes resulting in a consistent bias between the median depositions. The contribution of the water vapor correction (Eq. (1)) to the estimated dry deposition was very low. ΣN_r interference fluxes were between -3 and -0.3 ng N $m^{-2}s^{-1}$. The uncertainty ranged between 0.0 and 0.5 ng N $m^{-2}s^{-1}$. Considering two years of TRANC flux measurements with MDV as gap-filling approach, the correction contributed with 132 g ha^{-1} to the estimated dry deposition of 6.6 kg ha^{-1}.

We further investigated the impact of temperature, humidity, and precipitation on the dry deposition sums of ΣN_r compared to the dry deposition without restrictions when using MDV as gap-filling approach since we found differences in the diurnal patterns of ΣN_r for micrometeorological parameters. Therefore, we considered only fluxes in the time frame of ±5 days, at which temperature varied by ±3°C, humidity by ±5%, or precipitation was recorded. Remaining, long-term gaps (see panel (b) of Fig. R11) were filled by a monthly average of the respective half-hourly value estimated from non-gap-filled fluxes (Fig. 6). Those averages were also calculated for low and high humidity and temperature regimes separated by their monthly median. The calculations were made with and without the application of a u_*-filter. Figure R12 shows the annual dry deposition of the measurement years from the beginning of June to end of May.

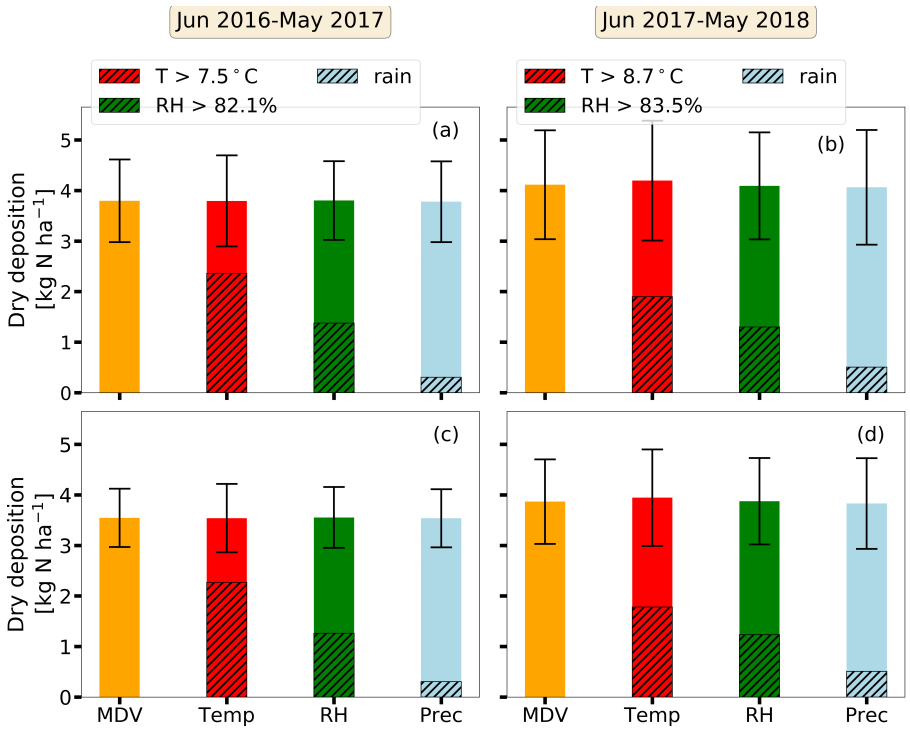

Figure R12: Annual ΣN_r dry deposition depicted as bar graphs from June to May in kg N ha^{-1}. For the orange bar, short-term gaps were filled with the MDV approach while using only fluxes in the time frame of ±5 days. In case of the red, green, and blue bar, fluxes used for gap-filling have to additionally fulfill criteria for temperature (±3°C), humidity (±5%), or precipitation (wet or dry). Remaining gaps were replaced by monthly averages estimated for each half-hour calculated from the non-gap-filled fluxes. For the meteorological cases, monthly medians were used to determine those averages for low and high humidity and temperature regimes. (a) and (b) were made for fluxes with u_*-filter, (c) and (d) without it. The hatched area of the bars represent the dry deposition for temperatures and relative humidity values higher than the annual median shown in the legend and for wet conditions.

No significant difference could be found between the dry depositions sums for both measurement years. Consequently, the applied selection criteria did not lead to biased sums compared to the dry deposition determined without restrictions for meteorological parameters. Warm, drier conditions exhibited a higher contribution to the annual dry deposition, in particular for the first measurement year. During rain, dry deposition was less than 500 g N ha^{-1} per 12-month period. As shown before, a difference in the application of a u_*-filter exists but is within the uncertainty range. Dry deposition was higher in 2017/2018, which was related to the large deposition fluxes observed in February 2018. In total, we estimated 3.8±0.8 kg N ha^{-1} and 4.1±1.1 kg N ha^{-1} with the MDV approach (orange bar) and u_*-filter for 2016/2017 and 2017/2018, respectively.

Comment R2.10 *The use of monthly mean concentrations for some of the compounds (DELTA measurements) adds significant uncertainty to DEPAC-1D model results. The first mention that the DELTA measurements are monthly seems to come in line 303 and the uncertainties are not mentioned until Line 622 (and there without references to, e.g., Schrader et al. 2018). The limitations of this approach should be more visible earlier on. Was the gap-filling of NH3 (Line 257) done in a mass-conserved way, i.e. was the available data removed from the long-term NH3 aver-*

age to work out what the average concentration during the gaps might have been? I suppose this would lower the uncertainty somewhat? Was a diurnal cycle superimposed on the long-temporal resolution measurements?

Response to R2.10 Yes, the usage of monthly mean values introduces a significant uncertainty to DEPAC-1D. We agree that the DELTA resolution has to be mentioned earlier. We added it to line 165 and will implement a detailed description of the usage of monthly DELTA concentrations and related uncertainties for the in-situ modeling in the second part.

Comment R2.11 *I do not follow the introduction of the DEPAC algorithm (Section 2.4.1). Erisman et al. (1994) does not describe a bidirectional resistance model (Line 224). Similarly, the references in lines 230-231 all describe deposition parameterisations, but most are almost certainly not the ones used in this version of DEPAC and contradict each other. The most correct description probably comes in Lines 243-247. Much of the description of the DEPAC-1D (Section 2.4.3), including the resistance parameterisations, probably also apply to the DEPAC version implemented in LOTOS-EUROS? It is all a little confusing. I did not realise until the Discussion section that DEPAC-1D does not treat the aerosol. This is a major and seemingly unnecessary shortcoming. My understanding was that DEPAC-1D is a stand-alone version of the deposition scheme implemented in LOTOS-EUROS and surely the latter treats the aerosol components. This seems hardly justifiable.*

Response to R2.11 We thank the Reviewer for his/her hints. We will improve the description of DEPAC and check the corresponding references within the preparation of the modeling study.

Comment R2.12 *I am confused throughout about the use of a compensation point for NH3 in the versions of LOTOS-EUROS and DEPAC-1D used. What is its magnitude for the forest types under consideration and where does it come from? Line 264 says that the DELTA concentrations were used for determining compensation points and additional deposition corrections? How was this done? Does this mean the models were not run with the standard scheme for these ecosystem types? Monthly concentrations do not lend themselves to deriving compensation points. Lines 671ff discuss uncertainties around cuticular compensation points. This would suggest that this was somehow adjusted based on the measurements?*

Response to R2.12 We appreciate the Reviewer's suggestions. Since the question relates to modeling part of the manuscript, this question will be answered in the modeling study.

Comment R2.13 *Given all this discussion about compensation points it is then highly surprising that Vd for HNO3 and NH3 are virtually identical (Line 374). How can this be? Apart from potential of evaporating NH4NO3 on leaf surfaces, HNO3 exchange is well understood and follows a near-zero Rc. NH3 does not.*

Response to R2.13 See above.

Comment R2.14 *I am similarly unclear about the discussion of the landcover (Lines 236-242). Given the resolution of LOTOS-EUROS of 7 x 7 km2 it is not surprising that the landcover of the grid cell containing the measurement site does not match that of the flux footprint which is much smaller. But I also do not see a big problem: is LOTOS-EUROS not based on a mosaic / tiling approach and predict fluxes to each landcover type separately? The associated description of the LAI values (Lines 273-279) is also unclear. Surely DEPAC-1D and LOTUS-EUROS simulate the deposition to all landuse types in a gridcell and from those a landcover-weighted average can then be calculated? In general, it should be made clearer what is identical and what is different*

between the LOTOS-EUROS and the DEPAC-1D simulation. What measurements were used for DEPAC-1D? Concentrations, meteorological parameters, canopy characteristics?
Response to R2.14 We thank the the Reviewer for his/her advice. We will implement your suggestions in the preparation of the modeling manuscript.

Comment R2.15 *The December emission fluxes are insufficiently explained. Were temperatures really sufficiently high to drive NH3 emissions from decomposition (Line 489)? Is there any evidence of freeze-thaw cycles affecting NH3 fluxes (Line 496)? Possibly, freeze-thaw cycle effects on soil NO are a more likely explanation? However, does the flux direction actually correlate with freeze-thaw events? Could it be caused by a problem with the measurement setup for a period of time given that December measurements differed between the two years?*
Response to R2.15 Yes, you are right that the emission fluxes were insufficiently explained. Previous conclusions regarding NH_3 being mainly responsible for the observed emission was most likely incorrect. Based on your suggestions and Reviewer 1, we improved the description. Please see R1.37. No issues with the instrument were found during the periods in December 2016 and 2017.

Minor scientific comments

Comment R2.16 *The abstract seems overly long and should be shorted. This can be done linguistically (e.g. remove phrases such as "We further showed that") and in terms of content. For example, it is sufficient to list the results in terms of annual deposition inputs and remove the numbers for the 2.5-year timeframe (line 19ff).*
Response to R2.16 Due to the separation of the manuscript, the abstract length was reduced. We removed redundant phrases and numbers for 2.5-year time frame.

Comment R2.17 *In Section 2.2 I am missing a fuller statement on the response of the TRANC to Nr compounds in the aerosol phase. What is the size-cut? What is the response to nitrate other than ammonium nitrate (e.g. sodium nitrate, calcium nitrate, ...)? Presumably they are not volatilised?*
Response to R2.17 Marx et al. (2012) conducted particle conversions test for sodium nitrate ($NaNO_3$), ammonium nitrate (NH_4NO_3), and ammonium sulfate ($(NH_4)_2SO_4$) since they are the most common nitrogen aerosol compounds (e.g., Wexler and Seinfeld, 1991; Nemitz et al., 2009). Aerosols were produced by a collision-type atomizer (TSI, St. Paul, USA) with a 0.3 mm nozzle from aqueous solutions of 0.5 g l^{-1}, 1 g l^{-1}, and 0.5 g l^{-1}, respectively (Marx et al., 2012). Conversion efficiencies were 78%, 142%, and 91% for $NaNO_3$, NH_4NO_3, and $(NH_4)_2SO_4$, respectively. A comparison with a twin differential mobility particle sizer (TDMPS) (Birmili et al., 1999) showed similar conversion efficiencies for $NaNO_3$ and $(NH_4)_2SO_4$ but differences for NH_4NO_3 (Marx et al., 2012, Fig. 6). At higher temperatures (>20°C) and relative humidity (>50%), NH_4NO_3 is semi-volatile resulting in higher fraction of NH_3 and HNO_3. Since TRANC-CLD detects the gaseous forms, a higher conversion efficiency than the one recorded by the particle detector can be expected. Overall, the results indicate that the TRANC is able to convert aerosols efficiently to NO. We added the determined conversion efficiencies for aerosols to the manuscript (line 149). For further details we refer to the publication of Marx et al. (2012)

Comment R2.18 *Line 33ff. I am not aware that deposition of Nr components threatens hu-*

man health. They do so by acting as precursors to PM2.5 and O3.
Response to R2.18 We added "by acting as precoursors for ozone (O_3) and PM2.5" to line 34.

Comment R2.19 *Line 80f. The critique of the MDS method is difficult to understand because it is not explained what it is. The introduction of CTM approaches is a little messy. Line 90 explains their workings by needing meteorological data and land-use information. Emissions and chemistry are only mentioned much further down.*
Response to R2.19 MDS utilizes the temporal correlation of micrometeorological parameters with fluxes to estimate gap-filled fluxes. In other words, "MDS requires a short-term stability of fluxes and micrometeorological parameters. This condition is not necessarily fulfilled for ΣN_r and its components. Their exchange patterns are characterized by a higher variability for different time scales leading to a lower autocorrelation and non-stationarities in flux time series compared to inert gases like CO_2." We replaced the lines related to MDS (lines 80-81) by the highlighted sentences given in this response. In addition, ΣN_r is a combination of several N_r species, which differ in physical and chemical properties and in their seasonal contribution. Thus, the application of data-driven gap-filling methods is suitable for gaps being a few days long. We appreciate the Reviewer's remarks to the introduction of CTMs. Since the manuscript was separated, the introduction changed. The paragraphs to CTMs and DEPAC were deleted, and a short paragraph to v_d and resistance analysis was added to the introduction (line 74).

Comment R2.20 *The introduction of the principle of operation of the TRANC is also not very logical. First reduced N is oxidised and then NH3 is formed from NH4NO3? Surely this happens before the oxidation (or in the same step).*
Response to R2.20 We agree that the description of the conversion steps is confusing. We deleted "resulting in an oxidization of reduced N_r compounds" (line 142) and generally improved the description (see R1.5 and R2.17).

Comment R2.21 *The description of turning the leaf wetness value into a boolean value needs to be improved (line 158ff). At present, a value of 10 in arbitrary units is meaningless.*
Response to R2.21 We agree that the explanation needs to be improved. We added the following sentences to line 158: "Due to a wetting of the sensor's surface, the electric conductivity of the material changes. This signal, the leaf wetness, was converted by the instrument to dimensionless counts. Based on the number and range of counts, different wetness states could be defined. Half-hourly leaf wetness values were in the range from 0 to 270. In this study, we defined the wetness states "dry" and "wet". The condition wet can be induced by the accumulation of hygroscopic particles extending the duration of the wetness state or water droplets. In order to classify a leaf as dry or wet, we determined a threshold value based on the medians of leaf wetness values." In order to clarify the determination of the threshold value used for classifying a leaf wetness sensor as wet or dry, we replaced the corresponding line 158 by the following sentences: "During daylight (global radiation $> 20\,W\ m^{-2}$), medians ranged from 1.1 to 2.0 and were between 4.1 and 9.4 during nighttime. During nighttime, medians are higher due to dew formation. According to the values determined during daylight, we set the threshold value to 1.5 for all sensors."

Comment R2.22 *Line 166ff. Please state the temporal resolution of the DELTA measurements. Also, later the text refers to ammonia diffusion samplers and NOx measurements, which do not appear to be mentioned in Section 2.2.*
Response to R2.22 Please see R1.7 and R1.10. NO and NO_2 measurements are mentioned in

Sec. 2.2. Here, NO_x was determined by adding NO to NO_2 concentrations.

Comment R2.23 *Line 199. Does the flux loss depend on the chemical composition of Nr?*
Response to R2.23 Wintjen et al. (2020) determined flux loss factors for two different ecosystems, which are different, for example, in the composition of ΣN_r. They assumed that the differences in flux losses are also related to the chemical composition of ΣN_r. We added the information to line 199.

Comment R2.24 *Line 207. Please state the relative magnitude of the water correction. What is its uncertainty?*
Response to R2.24 Please see R1.8.

Comment R2.25 *Line 211. Removal of fluxes outside a certain range appears to be arbitrary and subjective. Are these extreme fluxes not caught by the other tests, e.g. Foken's stationarity test or testing for stochastic significance via the random flux error? I presume the latter is what the "threshold of two times 1.96sigma" (Line 213) refers to? Currently, sigma is not defined and its calculation remains unexplained.*
Response to R2.25 We applied a limit filter for flux and concentration in order to filter out extreme outliers. Some of them were not identified by quality flags of Mauder and Foken (2006) or by the stochastic significance of the random flux error. σ represents the standard deviation of the variance. Fluxes were filtered out if variances of concentration, vertical wind, or temperature exceed the respective average plus $3 \cdot 1.96\sigma$. However, an investigation on the effectiveness of the filters revealed that quality flag criteria of Mauder and Foken (2006), a concentration limit filter, and a manual screening for periods of insufficient instrument performance, which resulted in irregularities in the raw signals (line 214-216), were sufficient to identify high-quality fluxes of ΣN_r. Please also note the answer to comment R1.9. Filters not needed were left out for preparation of the revised manuscript. We deleted the information to the variance filter (lines 212-213). Since other filters were chosen, the limits of flux filter and half-hourly fluxes also changed (lines 211 and 315).

Comment R2.26 *Line 264f. How were compensation points derived from long-term measurements of SO2 and NH3? This would seem problematic.*
Response to R2.26 We agree that additional details are needed to justify the determination of compensation points following Wichink Kruit et al. (2010). In the modeling study, the derivation of compensation points will be added to the description of DEPAC-1D.

Comment R2.27 *Line 266. Why was the LAI modelled for a site-based application? Why was this not based on a measured value?*
Response to R2.27 The LAI was not measured at the site. Please also see comment R1.11.

Comment R2.28 *Line 390. How do the diurnal cycles compare between measurements and model results? Does this shed add additional light on model deficiencies?*
Response to R2.28 We appreciate the Reviewers suggestions. A comparison of measured and modeled diurnal cycles will be made for the modeling study.

Comment R2.29 *Line 434. No, concentration is not proportional flux. The flux is proportional to the concentration. The concentration is the driver.*

Response to R2.29 Yes, you are right.

Comment R2.30 *Line 468. What do the concentration ranges refer to?*
Response to R2.30 Values for NO_2 and NO refer to 1992 until the end of 2008, NH_3 was measured from mid of 2003 to 2005. We added the information to line 468.

Comment R2.31 *Line 501. Both NO and NO2 contribute to Nr. So even if soil NO is converted to NO2 it will still contribute to the Nr flux except for the fraction that is removed by the canopy.*
Response to R2.31 We agree. The sentence was deleted, but we the implemented the Reviewer's suggestion in Sec 4.1.

Comment R2.32 *Line 507. The DELTA samplers does not measure NOx.*
Response to R2.32 Agreed. It should be DELTA+NO_x. Please note that the sentence was deleted.

Comment R2.33 *Line 514. There is a range of coatings available for the DELTA denuders. Clarify here and possibly also in the Methods section that carbonate coating was indeed used.*
Response to R2.33 We agree. For basic denuders, sodium carbonate and glycerol dissolved in water and methanol was used as coating for capturing HNO_3, SO_2, and NO_3^-, and citric acid and glycerol and also being dissolved in water and methanol as coating for acid denuders used for NH_3 and NH_4^+. Please note the changes to line 165 and comment R1.10.

Comment R2.34 *Line 551. Presumably in addition to total Nr concentration, its speciation also affects the net deposition rate and thus the flux.*
Response to R2.34 Probably, yes. Please note the revised discussion following line 551.

Comment R2.35 *Line 721. Is it worth adding DELTA, QCL and passive sampler data all to the graph to have an intercomparison between measurements? How do HNO3 compare between model and measurement? The modelled values of NH3 could also be too high because HNO3 in the model is too low (thus forming less NH4NO3).*
Response to R2.35 Figure R13 shows NH_3 concentrations of the DELTA system, passive samplers, and the QCL. NH_3 concentrations of the QCL were averaged to the exposition periods of the samplers. Figure R13 was added to supplement.

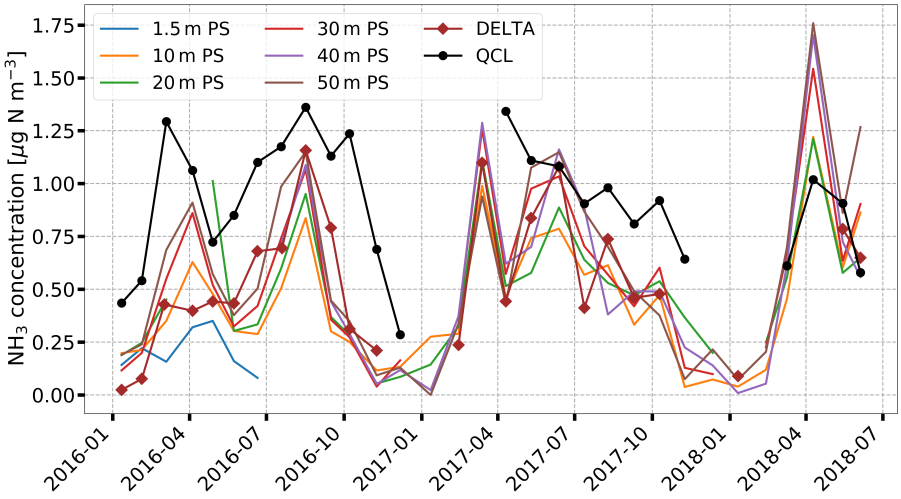

Figure R13: Concentrations of NH_3 measured by the DELTA and passive samplers, and the QCL in μg N m^{-3}. NH_3 of the QCL was averaged to the exposition period of the long-term samplers. Colors of the passive samplers indicate different measurement heights.

Averaged NH_3 concentrations of the QCL agreed well with NH_3 from passive samplers and DELTA measurements (Fig. R13). Overall, the agreement in the annual pattern was good, but a bias between the QCL and the diffusion samplers was found. From passive sampler measurements, an increase in the NH_3 concentration with measurement height could be observed. At 10 m (in the canopy), the lowest NH_3 concentrations were measured. No systematic difference was found between 20 m and 30 m. At 50 m, NH_3 was slightly higher (0.1 μg N m^{-3}) than 30 m. During winter, the difference in measurement heights diminished. Slightly higher NH_3 concentration were observed at 10 m in winter. A similar figure will be prepared for the modeling part including LOTOS-EUROS NH_3. As written in R1.29, a stacked bar graph to similar to Fig. R9 but with LOTOS-EUROS concentrations instead of TRANC ΣN_r will be made for the modeling part.

From R2.36 to R2.42, suggested modifications to the text and recommendations of the Reviewer are related to the modeling part and will be implemented in second manuscript.

Comment R2.36 *Line 739. The model presumably calculates u* from the ascribed canopy height and does not know about the complexity of the terrain. Are you saying that the measured u* is elevated because of topography? Would this not imply that the conditions for eddy-covariance are not met?*
Response to R2.36 The deviation in u_* was not related to the topography. u_* was calculated with the wind speed given at the reference height. As written in the manuscript, the reference height of LOTOS-EUROS was lower than the measurement height of the EC system. A single grid cell consists of various vegetation types, and all of them have different roughness lengths. We showed that the vegetation of the flux footprint differs significantly from the vegetation generated by the land-use classes for the grid cell. Thus, differences in u_* could be expected.

Comment R2.37 *Line 754. "input NH3 concentrations" Do you refer to emissions or long-range transport?*
Response to R2.37 For our measurement site, the elevated NH_3 concentrations were most likely

caused by emissions from nearby agriculture.

Comment R2.38 *Line 763. If the deposition event wasn't measured it maybe did not exist. I suggest to rephrase: "All models predicted at 2nd emission event which was not confirmed by the measurements."*
Response to R2.38 Agreed.

Comment R2.39 *Line 793f. But you say the Vd of NH3 is very high almost as high as HNO3. Thus, a large relative contribution of NH3 should give you large deposition fluxes.*
Response to R2.39 In case of the modeled v_d, yes. v_d of ΣN_r was significantly lower than the modeled v_d of NH_3 and closer to v_d of NO_2. Figure R10 reveals that NO_x, in particular NO_2, was the dominant N_r species and not NH_3. Presumably, a measured v_d of NH_3 would have been lower than modeled values.

Comment R2.40 *Line 795f. The wash-out could have occurred upwind and not contributed to the local wet deposition.*
Response to R2.40 Agreed. The sentence will be removed as written in R2.5.

Comment R2.41 *Line 798f. The good agreement seems entirely fortuitous given aerosol was not included in DEPAC-1D ...*
Response to R2.41 Currently, we are working on including of NH_4^+ and NO_3^- in DEPAC-1D for the modeling study.

Comment R2.42 *Line 803f. Maybe the gap filling methods are designed for compounds whose fluxes are actively regulated by production and consumption processes rather than the consequence of turbulence and concentrations such as deposition.*
Response to R2.42 We appreciate the Reviewer's suggestion for rephrasing and will modify the sentence accordingly.

Technical corrections / suggestions:

Comment R2.43 *General: avoid starting sentences with numbers. E.g. line 23 could better read "Deposition of 16.8 kg N ha-1 was calculated"*
Response to R2.43 We changed the beginning of the corresponding sentences.

Comment R2.44 *General: there are numerous places where an article is missing. E.g. line 86: "due to the low number", Line 146: "as a reducing agent", Line 179: "on an annual basis"*
Response to R2.44 We went carefully through the text and add articles if necessary.

Response to R2.44 General: there are several instances where the word "after" seems to be a mistranslation from German and needs to be replaced. Line 105: "were taken following the approaches of the International ...", Line 108: "nitrogen deposition using the canopy budget technique", Line 179: "bases following the CBT approach"

Response to R2.44 We checked corresponding lines and replace "after" by appropriate words.

Comment R2.45 *General: in many cases units are incorrectly combined. For example ms-1 should read m s-1 and µgm-3 should read µg m-3.*
Response to R2.45 We improved the notation of the units and separate them correctly.

Comment R2.46 *Line 7. I was surprised to see Nr concentration given in ppb rather than µg N m-3, especially since Nr contains aerosol compounds for which the use of ppb is rather unusual.*
Response to R2.46 Previous studies on measurements of ΣN_r by the TRANC also used ppb as unit for concentrations (e.g., Ammann et al., 2012; Brümmer et al., 2013; Zöll et al., 2019). In the TRANC, N_r species are converted to NO. The measured ΣN_r signal is basically NO, which is in a gaseous state under standard conditions. Therefore, the unit ppb seems to be appropriate for ΣN_r. In order to avoid switching between units, we changed the unit ppb to μg N m^{-3}. For comparing the measured concentrations to reported concentrations from other publications (lines 458-470), we changed the unit to ppb.

Comment R2.47 *Line 62. Better "EC studies of ..."*
Response to R2.47 Replaced by "Prior EC studies of..."

Comment R2.48 *Line 69 refers to "that site", but it is not clear which site is meant at this point.*
Response to R2.48 It will be replaced by "conducted with the same instrumentation at the measurement site".

Comment R2.49 *Line 96. "validation with flux measurements" (or "against").*
Response to R2.49 Revised.

Comment R2.50 *Line 116. "Measurements were carried out in". Actually, the authors should consider the alternative "Measurements were made" here and elsewhere.*
Response to R2.50 Revised. We made a rephrasing of the corresponding lines.

Comment R2.51 *Line 117. Remove "and".*
Response to R2.51 Revised.

Comment R2.52 *Line 130. Remove "which is remote from significant sources of emissions." This is repeating what was said before.*
Response to R2.52 Agreed.

Comment R2.53 *Line 139. "which was housed in an"*
Response to R2.53 Changed.

Comment R2.54 *Line 142. "oxidation"*
Response to R2.54 Word was deleted.

Comment R2.55 *Line 145. "during which remaining oxidised Nr species"*
Response to R2.55 Sentence was deleted.

Comment R2.56 *Line 219. "was caused by"*
Response to R2.56 Changed.

Comment R2.57 *Line 249. "filling the gaps in the flux data."*
Response to R2.57 Will be changed.

Comment R2.58 *Line 274. "weighted using the actual land-use fractions" ?*
Response to R2.58 Agreed.

Comment R2.59 *Line 275. "when considering only deciduous"*
Response to R2.59 Will be changed.

Comment R2.60 *Section 3.1. Much of the section here and elsewhere should be put into past tense.*
Response to R2.60 Agreed.

Comment R2.61 *Line 303 and elsewhere. Please add charges to NO3- and NH4+ (NO3 is a radical).*
Response to R2.61 Charges were added.

Comment R2.62 *Line 305. Redundant "with"*
Response to R2.62 Removed.

Comment R2.63 *Line 308. "the relative contribution of NH3 is significantly higher"*
Response to R2.63 Revised.

Comment R2.64 *Line 310 and elsewhere. A colon is followed by lower case in English.*
Response to R2.64 Revised.

Comment R2.65 *Line 311 "done following the criteria mentioned"*
Response to R2.65 Revised.

Comment R2.66 *Line 380 & 447. Should be "consequently" instead of "consequentially"*
Response to R2.66 Will be changed.

Comment R2.67 *Line 384. Should the units here be "kg N ha-1 a-1"?*
Response to R2.67 Yes, a N is missing here.

Comment R2.68 *Line 391. "Clearly, ..."*
Response to R2.68 Will be changed.

Comment R2.69 *Figure 6. The colours between upper and lower CBT estimate seem to be reversed.*
Response to R2.69 We agree. Colors will be switched.

Comment R2.70 *Line 417 and also line 816. "the range of ..."*
Response to R2.70 Will be changed.

Comment R2.71 *Line 450. "LOTOS-EUROS states out NH3 ..." – meaning unclear.*

Response to R2.71 Sentence will be modified as follows: "LOTOS-EUROS determines NH_3 as the main contributor to ΣN_r".

Comment R2.72 *Line 479. "Apart from management events, fluxes above the arable ..."*
Response to R2.72 Changed.

Comment R2.73 *Line 528. "Munger et al. (1995) also made NOy flux measurements ..."*
Response to R2.73 Sentence was deleted.

Comment R2.74 *Line 607. "sometimes lead to non-stationarities"*
Response to R2.74 Will be changed.

Comment R2.75 *Line 612 "under certain circumstances"*
Response to R2.75 Will be changed.

Comment R2.76 *Conclusions. Re-introduce all acronyms, including Nr.*
Response to R2.76 In the revised version, acronyms were mentioned in conclusions.

Response to Reviewer 3

General Comments *Wintjen et al. present an interesting and valuable data set on total nitrogen deposition to a forest spanning multiple years. The paper will be a worthy addition to N deposition literature, but would be improved by providing a few additional details and considering some additional analysis and interpretation.*

We thank the Reviewer for his/her comments and suggestions on this work. Since your comments and recommendations are discussed in the responses to Reviewer 1 and 2, we will add references to the given answers.

Comment R3.1 *Page: 8 line 252-254.It would be helpful to provide a little more detail on the calculation of resistances beyond just giving a reference. The actual equation itself would be ideal, but at least note what input variables are used in the parameterizations so that readers can know what the calculations are based on without having to consult multiple sources from the literature.*
Response to R3.1 We agree. We added a new chapter called "Determining deposition velocity and canopy resistance of ΣN_r from measurements" to the revised manuscript. In this chapter, equations needed for calculating the deposition velocity and canopy resistance of ΣN_r are given.

Comment R3.2 *line 257. Here it notes that alternate data sources are used for missing NH3 and HNO3. Is it stated anywhere how the data sources compare to one another when there are simultaneous measurements? Readers need this to assess whether there is any bias in the gap filling? Showing or mentioning a direct comparison would complement the plots showing cumulative deposition computed from different approaches. The direct comparison of simultaneous concentrations removes any confounding influence of other inputs to the calculated fluxes.*
Response to R3.2 Figure R13 shows a comparison of the NH_3 measurement techniques (see R2.35). In R2.3, a discussion is made on the agreement between the TRANC ΣN_r and ΣN_r derived from the DELTA samplers (see Fig. R9 and R10). We discuss the influence of micrometeorology on the MDV approach in R2.9.

Comment R3.3 *23 Line 449.Here it concludes that radiation is the primary driver affecting the diel cycle of N deposition. How have you discounted the role of wind speed/turbulence intensity, which will covary to radiation, as an alternative? If you account for the turbulence contribution to deposition velocity based on resistance model and thus compute an apparent canopy resistance from the residual is there still a dependence on radiation?*
Response to R3.3 Please see R1.47, R1.55, R2.2 and also the publication by Zöll et al. (2019). Turbulence was not identified as important driver for the ΣN_r flux by the authors. In R2.2, a discussion of the resistances and v_d is made (see Fig. R7 and R8).

Comment R3.4 *Page: 24 line 574 Do you consider the role of humidity and temperature on the partitioning between gaseous NH3 and NH4 aerosol? The patterns imposed by stomatal opening and NH3 partitioning might be difficult to distinguish. The observed pattern would be consistent with shifting the equilibrium toward gaseous NH3 during the warm and dry daytime conditions.*
Response to R3.4 NH_4^+ and NH_3 concentrations were obtained from DELTA measurements. During the warmer month, NH_3 concentrations were higher than concentrations measured for NH_4^+. From November to February, the situation was vice-versa. However, due to the denuder's low time resolution, we had no possibility to derive an influence of NH_4^+ and NH_3 on stomatal

processes, which happened on a shorter time-scale.

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