

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 R2.41, and the comment from R2.42 to 2.75 refer to technical corrections/suggestions. Comments of Reviewer 3 range from R3.1 to R3.5.

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 will split the preprint into two parts. We appreciate your suggestions to the discussion of the measurements. We include a comparison of the TRANC measurements to results from DELTA samplers and investigate the effect of micrometeorology on the deposition velocity. As outlined in the author comment, discussions on the modeling results will be shifted to the modeling manuscript, and questions related to the nitrogen modeling will not be answered in this response. 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 reword 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 is the flow rate after the critical orifice. The mass flow rate before the critical orifice is the same as after the critical orifice. Since mass flow is equal to both sides of the critical orifice, a difference in flow velocity is 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 NH₃. The exposition duration was approximately one month. We specify the type of passive samplers in the revised manuscript.

Comment R1.8 *Line 207: What was the typical magnitude of this correction to the total Nr flux?*

Response to R1.8 The correction contributes approximately 132 g N ha⁻¹ to two years of TRANC flux measurements if the Mean-Diurnal-Variation (MDV) approach is used as gap-filling approach. The half-hourly magnitude is -0.22 ng N m⁻² s⁻¹ on average. We add the information given in this response to the description of the water vapor correction.

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 a reduced pump performance leading to an increase in sample cell pressure. If pressure in the sampling cell is outside the range given by the manufacturer, low pressure conditions needed for the detection of photons emitted by excited NO₂ molecules may not hold. Pump efficiency was controlled at least monthly, and the pump was replaced if necessary. Issues in maintaining a stable temperature regime within the air-conditioned box, and the change in the O₂ supply from gas tanks to ambient, dried box air could affect the measurements. Also, sample cell temperature was checked at least monthly. Gas tanks of O₂ were replaced regularly. 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 concentration were carefully checked for irregularities like spikes or drop-outs by visual screening.

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.

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.

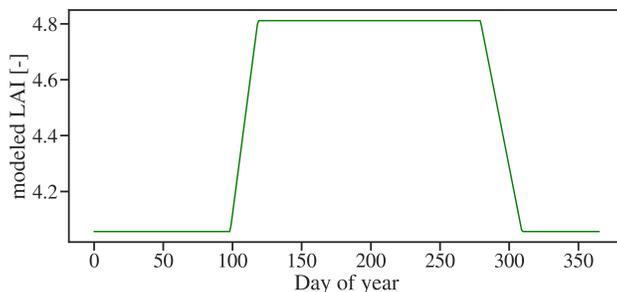


Figure R1: Modeled LAI following van Zanten et al. (2010) for measured fractions of coniferous forest (81.1%) and deciduous forest (18.9%) within the flux foot print for a year.

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 $NO_x > 20ppb$ 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 is comparatively low. NO_x peaks above 20 ppb 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 prepare a supplemental file.

Comment R1.16 *Line 303: How does the sum of the concentrations measured by the DELTA compare to the TRANC N_r measurement?*

Response to R1.16 According to the reviewers suggestion, we add a stacked bar graph (Fig. R9) showing monthly concentrations of the DELTA measurements compared to the TRANC ΣN_r concentrations. Latter are averaged to the exposition period of the DELTA samplers. The comparison of the absolute values revealed significant underestimations of TRANC ΣN_r from March to mid of May 2018 and from July to mid of August 2017. Differences to DELTA+ NO_x were about $0.6 \mu g N m^{-3}$ in summer 2017 and $1.2 \mu g N m^{-3}$ in spring 2018. We found that the zero-air calibration value of the TRANC-CLD system was incorrect for the mentioned time periods 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 and evaluations shown in the response are made with the bias-corrected data. Figures of the manuscript will be updated accordingly. On average, the TRANC values are slightly higher by $0.3 \mu\text{g N m}^{-3}$ than DELTA+NO_x. Please see R2.3 for further details.

Comment R1.17 *Line 317: What fraction of the non gap-filled half-hourly fluxes exceeded the flux detection limit?*

Response to R1.17 52% of the non gap-filled fluxes were higher than the flux detection limit. It shows that nitrogen dry deposition was close to detection limit of the used measuring device. It confirms our findings that nitrogen exchange happens 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.

Comment R1.18 *Line 350: Should “based on” be “are based on”?*

Response to R1.18 Yes, you are right.

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 is 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 highlight the effect of the measurement heights.

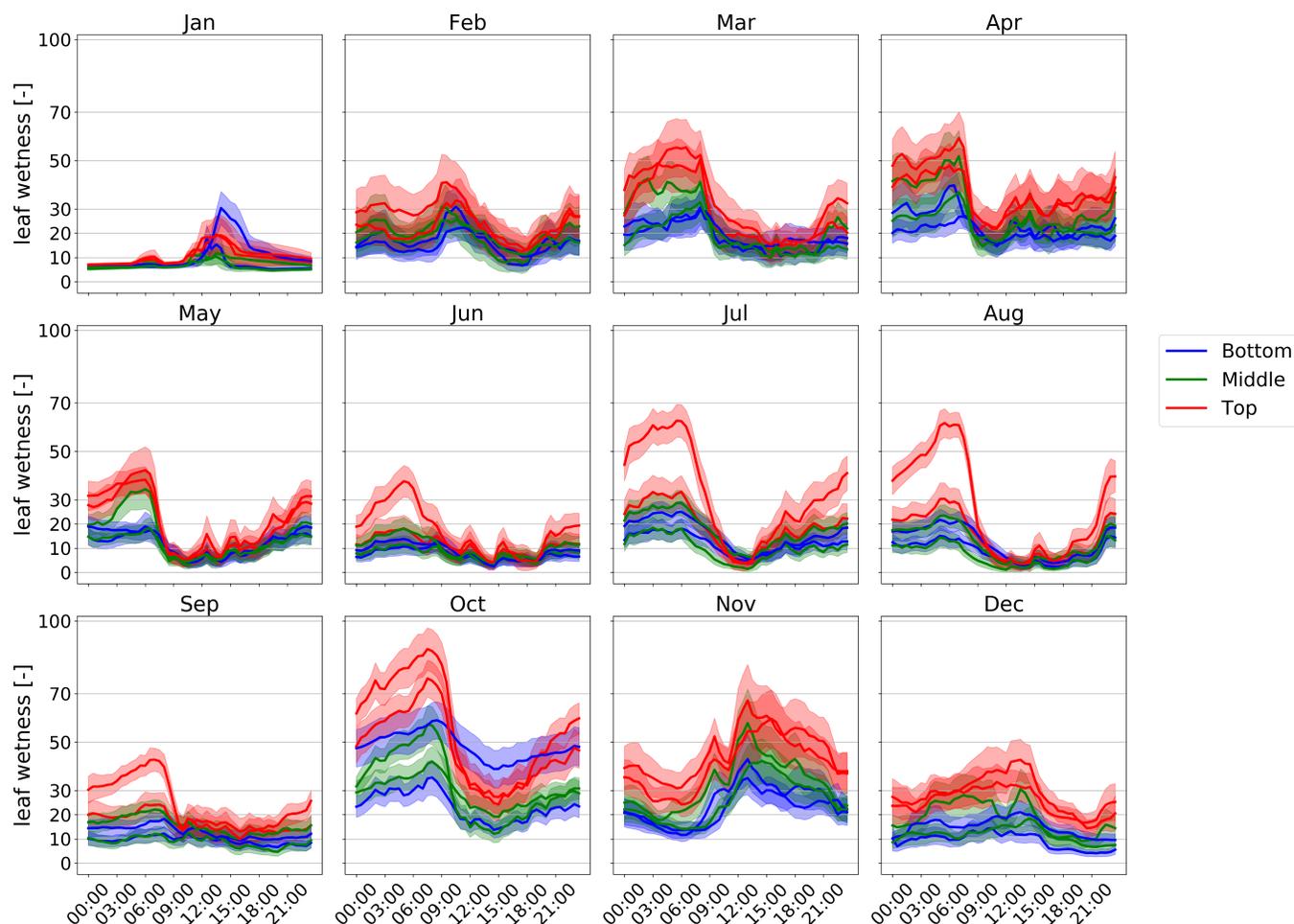


Figure R2: Daily cycles of the leaf wetness for 2017. Colors indicate installation heights of the sensors (red=bottom, green=middle, blue=top). Shaded areas represent the standard error of the mean.

On monthly basis, the diurnal patterns of the sensors were almost the same for a season. During autumn and winter, no difference between the sensors heights was found. From April, the start of the growing season, to September highest values were measured during dawn and lowest values during the day. Sensors from the mid and the top exhibited slightly higher values than the sensors from the lowest position, but the former showed a sharper decrease after sunrise. During daylight, only slight differences in measurement height are visible. Considering the standard error, the differences in measurement heights diminish, especially between the lowest and middle sensor. Also, sensors from the mid and the top are 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 lowers its uncertainty. We will correct the statement in the revised manuscript. Figure R2 will be available 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 NH_4NO_3 during this period? Given the lower V_d of parti-

cles 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. We agree that the contribution of particles to ΣN_r must be higher or equal to compounds which are most responsible for driving N deposition during this period. Since we discussed the observed deposition event with regard to the results of LOTOS-EUROS, we will move the interpretation to the modeling manuscript.

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 is 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 is 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 NH_4^+ 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 Reviewers 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 N_r species concentrations (Delta compounds, NO_x , QCL NH_3 , passive NH_3) 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 N_r 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 is misleading and additional context is required (see also R2.4). We will modify the interpretation accordingly.

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 will change it to "Concentration values of NH_3 and NO_x ..."

Comment R1.33 *Line 472: "...confirm the seasonal pattern of N_r ". Do you mean that those studies show patterns consistent with the current study?*

Response to R1.33 The interpretation somewhat is misleading. The seasonal flux pattern of different N_r compounds reported by the mentioned studies are comparable to the monthly pattern observed for ΣN_r from our site. They exhibit a similar shape and the same order of magnitude. In the present case, the word "confirm" is too strong. We rephrase the statement in the revised manuscript accordingly.

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, NO_x 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 will modify the sentence accordingly.

Comment R1.35 *Line 477: This sentence should include references for the "few studies focusing on N_r ".*

Response to R1.35 Agreed. Namely Ammann et al. (2012), Brümmer et al. (2013), and Zöll et al. (2019) measured ΣN_r fluxes with the eddy-covariance method.

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” refers to Taylor and Jones (1990). However, by reflecting the suggestions of the Reviewers 1 and 2 to the paragraph, we doubt that NH₃ is most responsible for the observed emission fluxes in December 2017. Since NH₃ is water soluble given by the molecular structure a diffusion through a partially wet snow layer seems improbable. 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 , the latter as cumulative curves, 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.

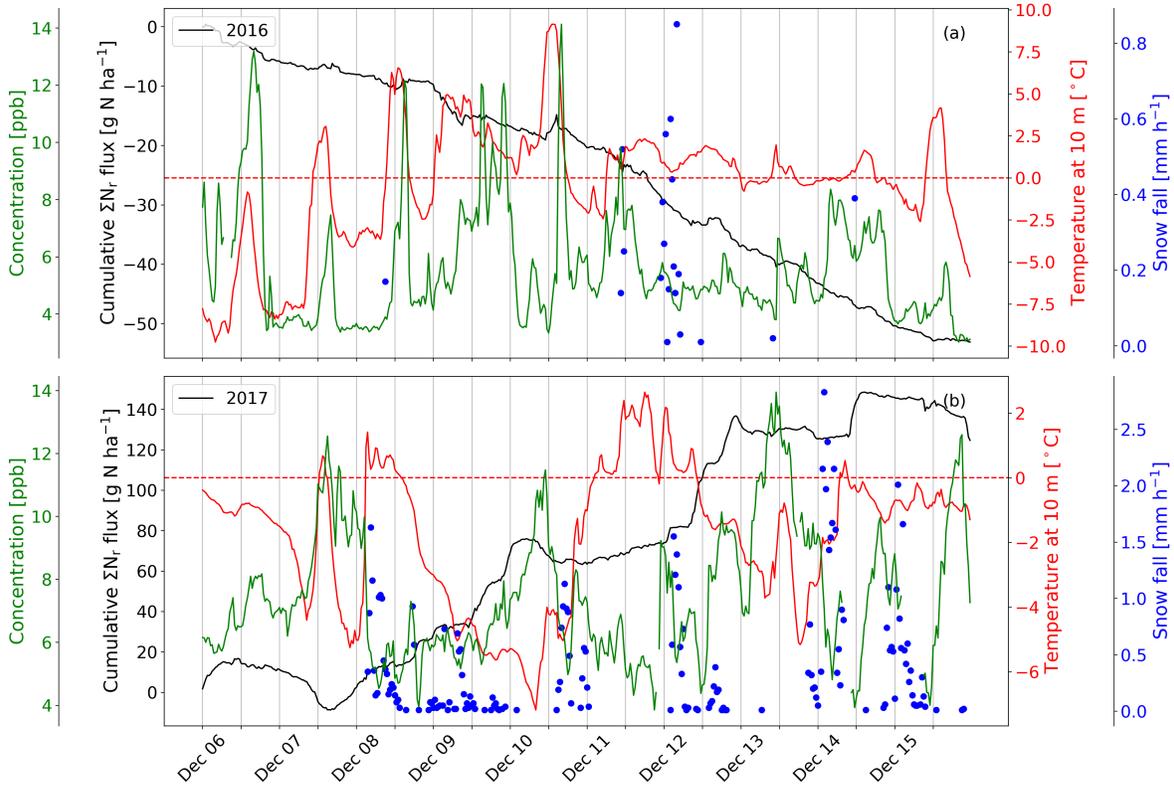


Figure R3: Air temperature at 10 m height above ground (red), snow fall (blue), ΣN_r (green) concentrations, and fluxes as cumulative sums (black) from 6 December to 15 December. In (a), the measurements are displayed for the period in December 2016, in (b) for December 2017. Gaps are filled with the MDV approach with fluxes being in a range of ± 3 days.

In 2017, we observed substantial snow fall and a slower varying temperature compared to 2016 leading to significant snow depth compared to 2016. On the 1st of December, 1 cm and 20 cm snow depth were measured in the catchment 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 $0^{\circ}C$ in December 2016. In 2017, temperatures were mostly below $0^{\circ}C$ and only for one day above $0^{\circ}C$, 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 happen 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 for the snow insulation case. 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 inhibits molecular diffusion and the release of N_r species. Thus, emission of nitrogen from the soil or the decomposition of leaves was probably reduced

compared to 2017. A deeper snow layer reduced the impact of soil frost penetration, promoted micro bacterial activities, 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 been 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 $< 3^\circ C$). They found that NO fluxes were positively correlated to air and soil temperature. Only minor response were found to freeze-thaw events. Their influence on the cold season NO fluxes was neglectable. 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 happen in the topsoil and lower 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 (Medinets et al., 2016, and references therein). A 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 are expected. We will include Figure R3 in the revised version and improve the lines substantially.

Comment R1.38 *Line 508: Change "proposed" to "reported".*

Response to R1.38 Done.

Comment R1.39 *Line 511: Change "by DELTA" to "by the DELTA".*

Response to R1.39 Done.

Comment R1.40 *Line 528: Change "high" to "large".*

Response to R1.40 Done.

Comment R1.41 *Line 529: Change "at less" to "at a less".*

Response to R1.41 Done.

Comment R1.42 *Line 530: Change "It shows" to "These studies indicate"*

Response to R1.42 Done.

Comment R1.43 *Line 539: Please consider splitting up this long sentence for clarity.*

Response to R1.43 Agreed.

Comment R1.44 *Lines 544 – 547: The last two sentences of this paragraph seem more appropriate for the conclusions section.*

Response to R1.44 We agree and will add them to the conclusion section.

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 N_r fluxes above forests.

Response to R1.45 We appreciate the Reviewers' suggestions to section 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 and canopy resistance R_c of ΣN_r will be added. The discussion related to the effect of precipitation on the ΣN_r will be reworked.

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 will change, we will also consider to adjust the header. A proper header can be "Influence of micrometeorology and nitrogen concentration on deposition and emission".

Comment R1.47 *Line 549: What is the proposed mechanistic relationship between N_r flux and global radiation? What about the diurnal pattern of turbulent mixing and its role in air-surface exchange?*

Response to R1.47 As shown by Zöll et al. (2019), ΣN_r and CO_2 fluxes exhibit a similar daily cycle and show a strong dependence on global radiation 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 compound like NO_2 (Thonen et al., 1996) or NH_3 (Wyers and Erisman, 1998; Hansen et al., 2015) 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) has to control 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 global radiation and concentration, respectively, affect the uptake of ΣN_r . Further shown by Zöll et al. (2019) other parameters like friction velocity (u_*) were not identified as important drivers for ΣN_r . Photochemistry appears to be more important than turbulent mixing. Radiation changes the composition of ΣN_r due to the formation of ozone (O_3). In addition, global radiation has an influence on u_* as seen by their similar shapes in daily cycle. 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. 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 has to be noted that the findings of Zöll et al. (2019) were conducted for ΣN_r at a natural, unmanaged site where micrometeorological parameters are 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 large-scale effects as written by Zöll et al. (2019). We will add the information given in this answer to the revised section.

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 will add plots similar to Fig. 2 and 3 but for the ΣN_r concentration to the supplement. Figure R4 shows a scatter plot of the measured fluxes for different u_* classes and global radiation (R_g) higher than 50 W m^{-2} .

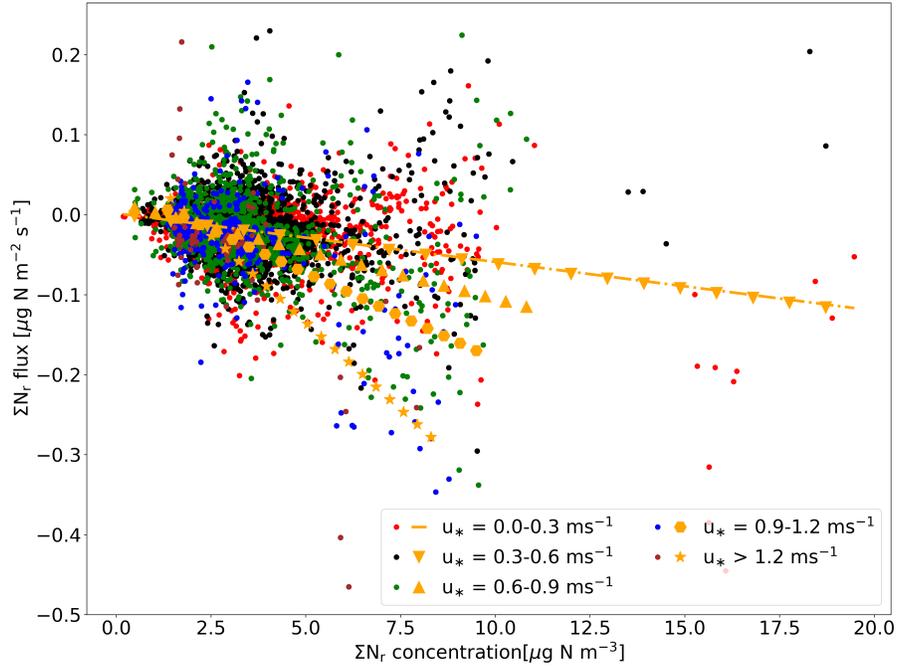


Figure R4: Dependency of measured concentrations on corresponding ΣN_r fluxes shown as scatter plot during daylight ($R_g > 50 \text{ W m}^{-2}$). Colors indicate different u_* classes. Linear regressions between concentrations and fluxes are made for each u_* class indicated by different symbols.

We find a decreasing slope with increasing u_* . The slope corresponds to the deposition velocity (v_d). The results of the linear regression, v_d and squared correlations (R^2), are listed in Table R1.

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

Table R1: Results of the linear regressions from Fig. R4 for the selected u_* ranges.

For u_* values lower than 0.9 m s^{-1} , v_d is almost similar. For u_* values close to 1 m s^{-1} or even higher, deviations in v_d can be found. However, a significant R^2 is only given for u_* higher than 1.2 m s^{-1} . Since the aerodynamic resistance (R_a) (Garland, 1977) and quasi-laminar resistance (R_b) (Jensen and Hummelshøj, 1995, 1997), after stability corrections of Paulson (1970); Webb (1970), decrease with increasing u_* , v_d also increases. Thus, an influence of atmospheric turbulence on the deposition of ΣN_r appears to be relevant for the highest u_* values. Figure R5 shows the daily cycle of concentration, R_g , u_* , air temperature (T_{air}), and v_d for the period from May to September.

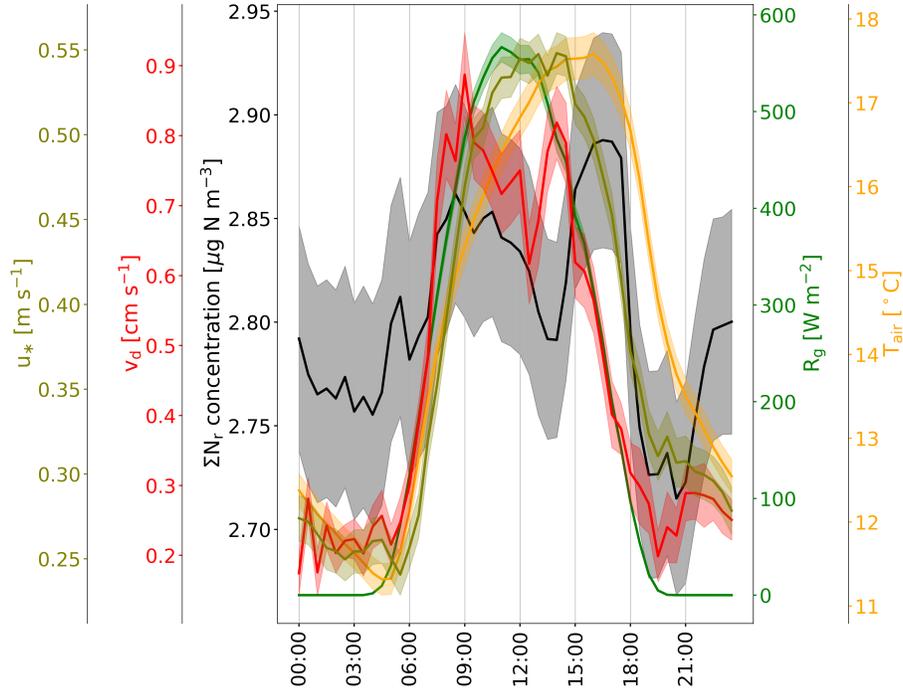


Figure R5: Daily cycle of ΣN_r (black) concentration, R_g (green), u_* (olive), air temperature T_{air} (orange), and v_d (red) for the period from May to September. Shaded areas represent the standard error of the mean.

We observe that R_g , u_* , and T_{air} exhibit a similar shape with highest values during the day and lowest values during the night. u_* and T_{air} reach their maximum two to three hours later. A similar observation is made for the concentration whereas v_d shows largest values around noon and lowest values during the night. Figure R6 is made for the same variables but for December, January, and February.

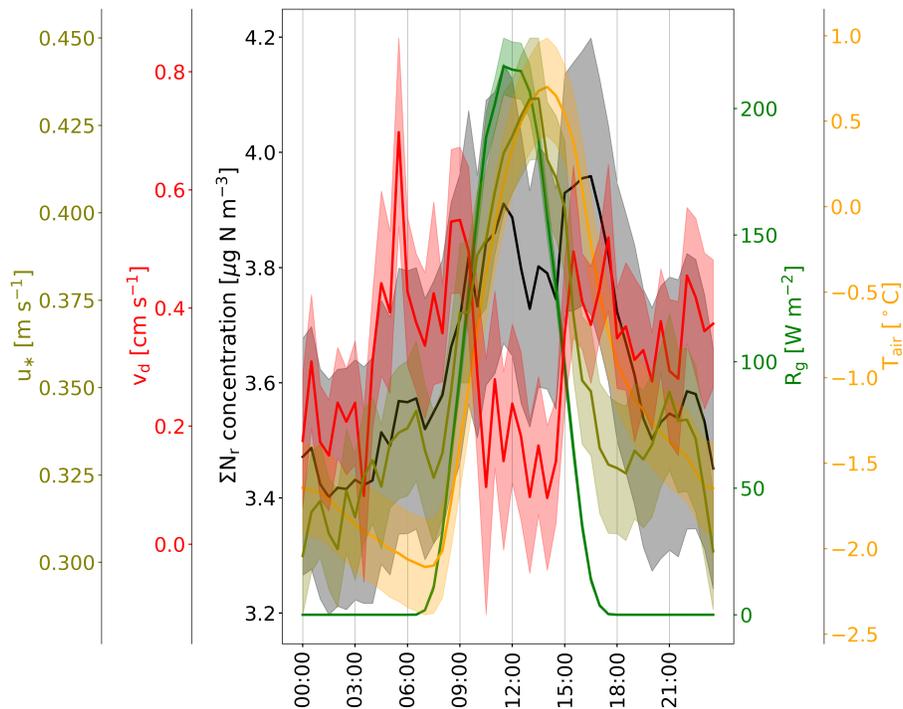


Figure R6: Daily cycle of ΣN_r (black) concentration, R_g (green), u_* (olive), air temperature T_{air} (orange), and v_d (red) for the period from May to September. Shaded areas represent the standard error of the mean.

Compared to the summer period, similar observations can be made for R_g , u_* , T_{air} , and concentration. Obviously, significant differences in the amplitude are visible for global radiation and temperature. However, v_d is almost equal and even lower during the day, which results in lower deposition of ΣN_r during winter. Thus, enhanced deposition during summer is related to global radiation, which lead to an increase in temperature and plant activity under sufficient temperatures, and to the stomatal compensation point, which also affects v_d . Since slight deposition of ΣN_r observed in periods with less global radiation, deposition likely happens through the cuticular or soil pathway.

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. We will replace it by “influence”.

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. 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 (VPD) is considered as secondary driver. Additionally, we found that v_d is reduced for high ambient humidity and wet leaf surfaces. Since we measured NH_3 indirectly by ΣN_r and above an ecosystem characterized by lower humidity than a peatland, global radiation

favoring the exchange through the stomatal pathway appears to be more important for ΣN_r at the measurement site. We will correct the sentence with the information given here.

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 We found significant differences in global radiation, in particular for spring deposition. In March 2018, R_g was about 50 to 100 Wm^{-2} lower than in April 2018 and about 30 to 50 Wm^{-2} lower than a year before during daytime. DELTA measurements displayed in Fig. R9 showed that NH_3 measured in March 2017 is about 0.5 $\mu\text{g N m}^{-3}$ higher than in March 2018. In April 2017, NH_3 was about 1.0 $\mu\text{g N m}^{-3}$ lower compared to April 2018. Due to lower global radiation and temperature, fertilization season was probably shifted to mid/end of April 2018 whereas the fertilization season started one month earlier in 2017. The different contributions in NH_3 and conditions in radiation affect v_d and the stomatal resistance. Thus, v_d was slightly lower than zero in spring 2017 whereas v_d was close to zero and even positive in spring 2018.

After the bias correction, concentration differences between the summer periods of 2016 and 2017 are still existent but differences in the ΣN_r median concentration are lower than 1 ppb. No remarkable differences in micrometeorology were found between summer 2016 and 2017. Again, Fig. R9 reveals that the contribution of components to ΣN_r differs between the investigated time periods. From July to September 2017, the mean NH_3 concentration is about 0.3 $\mu\text{g N m}^{-3}$ lower than a year before. HNO_3 , NH_4^+ , and NO_3^- are remarkably low in July 2017 compared to July 2016. We conclude that the deviations in the median deposition are not related to differences in the ΣN_r concentration. The differences in the composition of ΣN_r affect v_d more and therewith the measured fluxes. The discussion of the deposition periods will be improved accordingly.

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 the fluxes and v_d were found. Thus, conditions for deposition through stomatal and cuticular pathway are comparable. We implement 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 will be removed in the revised version.

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. The sentence will be changed.

Comment R1.55 *Line 572: What is the proposed mechanism by which dry conditions enhance N_r 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 R_c . It shows if the association between the 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 driver analysis since ΣN_r compounds like NH_3 , HNO_3 and NH_4^+ are affected by rain. Thus, the entire paragraph (lines 571 - 598) will be rewritten. Updated versions of Fig. 4 for v_d and R_c are shown in R2.2. Also, further details on aerodynamic and surface resistance are given in R2.2.

In regard to the question, under dry conditions higher temperatures and lower relative humidity caused by radiation lead to higher stomatal deposition during daylight. A clear diurnal pattern is observed for v_d with high values for v_d around noon and low, non-zero values during the night. The stomatal uptake is only present for the period from May to September. During the rest of the year, no diurnal pattern is found under dry conditions since stomata are closed, or requirements for stomatal deposition are not fulfilled (stomatal compensation point). Since we still observe a low, non-zero v_d during seasons with lower radiation, cuticular and soil deposition and turbulent processes are 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 is greatly reduced or even inhibited due to reduced radiation or leaf area surfaces. Precipitation also reduces incident radiation and reduces stomatal uptake. In those periods, v_d is slightly positive with periods of negative values and almost constant during the day. No difference is found for the temperature and relative humidity threshold. Since stomata are most likely closed but deposition is still observed, cuticular processes, an exchange with soil, or turbulent processes appear to be the possibilities for the ΣN_r exchange. Generally, cuticular deposition is 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 may also be lower under wet conditions because the requirements for cuticular deposition are not met (cuticular compensation point) since water soluble compounds N_r can be washed out below the measurement height. 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. To conclude, stomatal deposition seems to be more important than other in-canopy uptake processes for the ecosystem in close proximity to the measurement site during times of high incident radiation.

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. The sentence will be modified.

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 N_r 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). We will improve the sentence accordingly.

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 is less useful. As written before, we did a reanalysis of Fig. 4 by separating fluxes, v_a , and R_c in dry and wet classes. With the improved versions of Fig. 4, Figure F1 adds no additional information and will not be shown in the revised version. The quality of fluxes measured during rain is almost similar to flux measurements with no measured precipitation. For example, 15% of the “wet“ fluxes are classified as flag two following the Mauder and Foken flagging system (Mauder and Foken, 2006). Also, 15% of the “dry“ fluxes are 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 will be 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 is assumption needs further clarification. Wyers and Erisman (1998) measured highest NH₃ deposition if the canopy has a high water storage level (CWS) (> 2 mm). The deposition efficiency was reduced if CWS was higher than 0.25 mm but lower than 2 mm. By comparing different measurement years, they found differences in the deposition efficiency even if the canopy is saturated with water. They attributed the effect to the solubility of NH₃ in the water film. If canopy gets drier, evaporation of water occurs and the concentration of NH₃ increases in the water film. The cuticular resistance increases and deposition of NH₃ is reduced. Even emission of NH₃ was observed Wyers and Erisman (1998), especially during the day. During the day the canopy was dry, and NH₃ exchange was bidirectional. They showed that

stomatal resistance was higher than canopy resistance. The authors identified cuticular deposition as more important for NH_3 than stomatal deposition. As written in the manuscript, Wyers and Erisman (1998) measured an average NH_3 concentration of $5.2 \mu\text{g m}^{-3}$. We measured $0.65 \mu\text{g m}^{-3}$ on average and found that the contribution of NH_3 to ΣN_r is comparatively low at the measurement site. Probably, cuticular deposition of water soluble N_r species is reduced at our measurement site. Fig. R7 shows that v_d during rain is significantly lower than during dry conditions and exhibits no diurnal pattern. If we further exclude wet leaf surfaces, the difference between the diurnal patterns separated by RH and T diminishes.

Thus, the statement has to be modified as follows: 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. Wyers and Erisman (1998) 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, the measurement site is several kilometers away from potential (anthropogenic) emission sources. Concentrations of NO_3^- , NH_4^+ , sulphur dioxide (SO_2), and NO_x are comparatively low at the site, in particular during summer. Thus, stomatal deposition appears to be more important for ΣN_r under dry conditions. 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 total ammonium (tot-NH_4^+) and total nitrate (tot-NO_3^-) during dry conditions. 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 $0.45 \mu\text{g m}^{-3}$ 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 $0.78 \mu\text{g m}^{-3}$ for NH_3 , HNO_3 , NH_4^+ , and NO_3^- , respectively. Measured tot-NO_3^- and tot-NH_4^+ of (Wolff et al., 2010) exhibit 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 resistances 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.

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 will be 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 N_r species like HNO_3 is relatively straightforward compared to NH_3 ” 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 NO₃⁻ and NH₄⁻ 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. 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 concen-

trations and a figure similar to Fig. R13 with NH_3 from LOTOS-EUROS.

Comment R1.76 Line 722: *“The difference to LOTOS-EUROS NH_3 concentrations was highest during periods with significant amount of NH_3 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 are located. Thus, a reduction of the size may reduce the modeled concentrations of grid cell, in which the measurement site is 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 NH_4^+ and NO_3^- concentrations during this period compared to other months? These data should be presented.*

Response to R1.78 Unfortunately, we had no DELTA results for that period.

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 of CBT approach 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 will be reduced. We will state 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 (N_r) to a forest site, interpretation of the results in the light of measurements of N_r 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 N_r dry deposition is rare and such a long dataset of N_r 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 N_r flux measurements were accompanied by NH_3 flux measurements (by QCL), which I hoped would have been used to elucidate the non- NH_3 component of the N_r flux. However, I was let down in various aspects: the NH_3 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 NH_3 exchange and its impact on the total N_r flux and with NH_3 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 N_r flux with one instrument. The disadvantage is that it does not shed light on the behaviour of the individual N_r 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 N_r 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 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 the 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 is too long and can be shortened. We plan to shift the discussion of the individual ΣN_r compounds to the modeling manuscript. As stated in the author comment, the analysis will be done on v_d and R_c . Finally, the total nitrogen budget will be discussed. Therefore, we will include measurements of wet deposition taken close to the tower by bulk and wet-only samplers and show the effect of micrometeorological filters on the MDV approach used for the calculation of the nitrogen dry deposition budget.

In the modeling study, individual flux components of DEPAC-1D will be compared to values reported in literature. As done for the TRANC ΣN_r v_d , 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 addressed all mentioned points related to the flux measurements and will implement your suggestions in the revised manuscript. Since we make 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 mentioned before, a calculation of the NH₃ fluxes did not work out with the eddy-covariance method. No consistent NH₃ 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 can be excluded. We suppose that the variability in the measured NH₃ concentrations was not detectable by the instrument. Significant short-term variability in the ΣN_r raw concentrations were not found in the NH₃ signal even in spring or summer. Thus, no robust time lag estimation had been conducted between the vertical wind component of the sonic anemometer and the NH₃ concentration.

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 N_r , the deposition of N_r 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 V_d 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 V_d . For example, Fig. 4 would be more meaningful if presented for V_d . In fact, an analysis in terms of R_c would be even more meaningful as it would normalise for the effect of turbulence on R_a and R_b both of which contribute to V_d . Because particles are not really subject to a boundary-layer resistance in the way it is applied to gases, R_c is not really meaningful. However, the analysis could be done in terms of $V_{ds} = V_d(z_0)$, i.e. after normalising at least for R_a .*

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 will benefit from an analysis of v_d in order to separate the effects of v_d and concentration for ΣN_r . Figure R7 is done in accordance to Fig. 4 in the manuscript but for v_d . We further determined R_a and R_b after Dyer and Hicks (1970); Paulson (1970) and Garland (1977), respectively. 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 HNO₃, NH₃, NO and NO₂ multiplied with their individual molecular diffusivities adapted from Massman (1998) and Durham and Stockburger (1986). Therefore, R_c should be seen as an effective resistance.

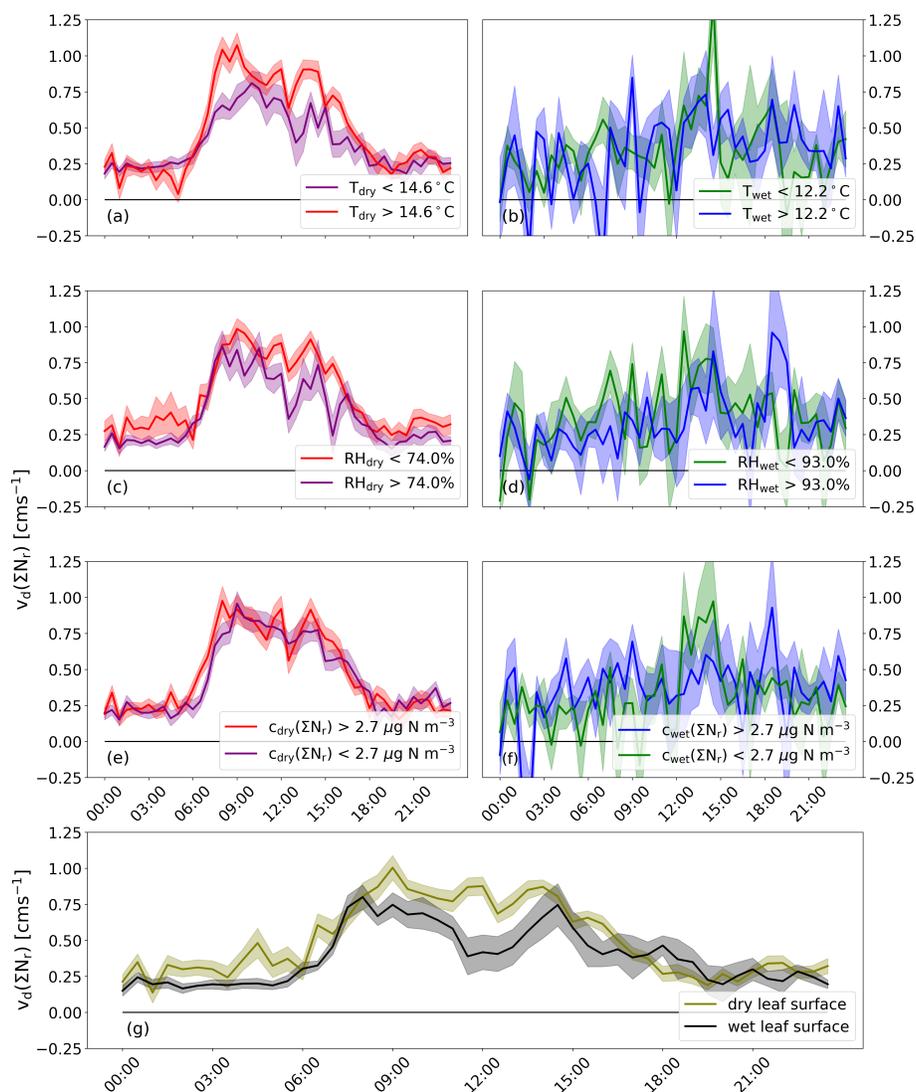


Figure R7: Mean daily cycle from May to September of v_d for low and high temperature, humidity, and concentration separated by dry and wet conditions. Median values of temperature, humidity, and concentration, which are derived for the same time period, are used as threshold values for separating v_d . For separating dry and wet leaf surfaces, the scheme proposed in Sec. 2.2 (and further explained in R2.21) is applied. Further details are given in R2.21 and R1.19. The shaded areas represent the standard error of the mean.

In general, v_d is lower than values proposed for NH_3 and HNO_3 by Schrader and Brümmer (2014) and closer to values of NO_2 implicating a lower contribution of NH_3 than NO_2 to the measured flux. During dry conditions, a clear diurnal pattern can be observed in v_d with highest values around noon and lowest values in the night. Differences are still visible in micrometeorological parameters, but the difference for different concentration regimes diminishes compared to the ΣN_r fluxes. In case of precipitation, v_d is significantly reduced during daytime and almost constant for the entire day. No difference can be found for low and high temperature, humidity, and concentration regimes during precipitation. During other seasons of the year, no diurnal pattern is observed during dry conditions. Figure R8 is in accordance to Fig. R7 but for R_c .

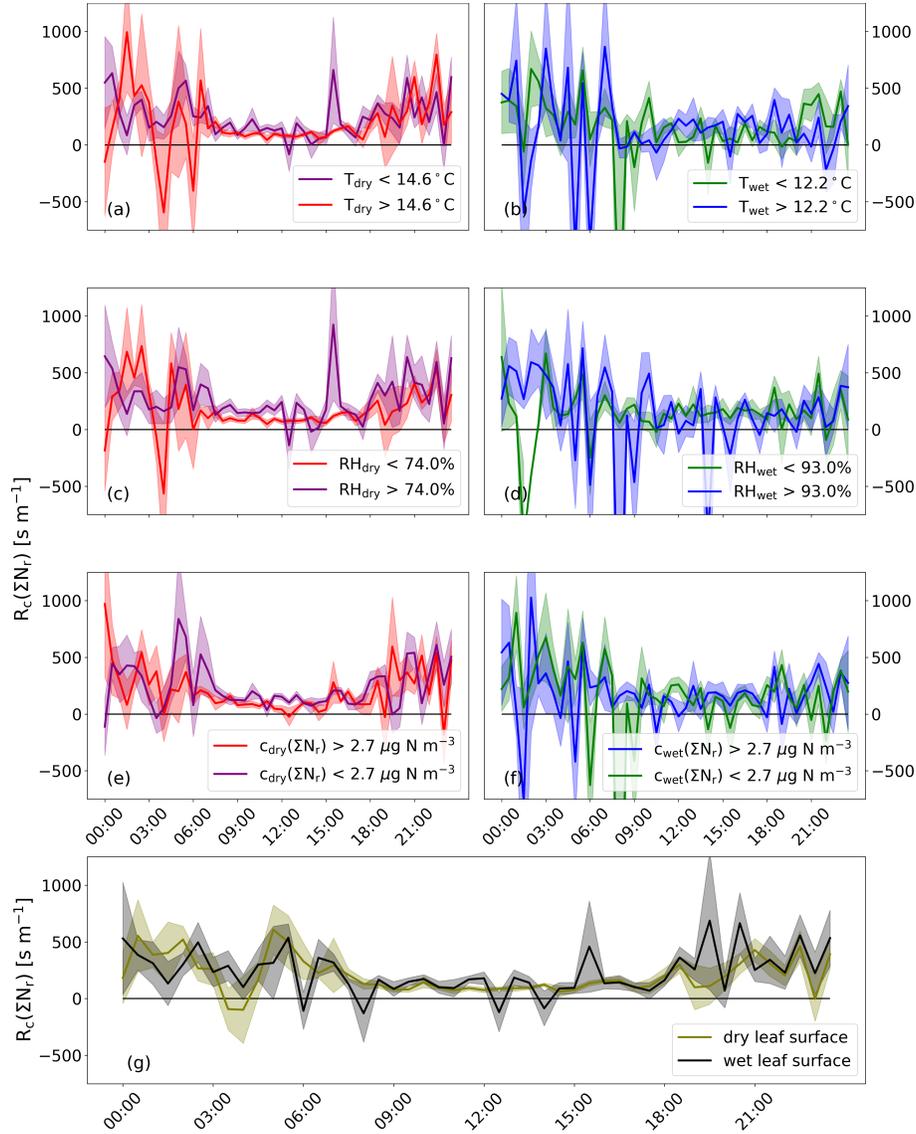


Figure R8: Mean daily cycle from May to September of R_c for low and high temperature, humidity, and concentration separated by dry and wet conditions. Median values of temperature, humidity, and concentration, which are derived for the same time period, are used as threshold values for separating r_c . For separating dry and wet leaf surfaces, the scheme proposed in Sec. 2.2 (and further explained in R2.21) is applied. Further details are given in R2.21 and R1.19. The shaded area represents the standard error of the mean.

R_c exhibits lowest values during the day and highest values at night. During nighttime, the variability in R_c is enhanced whereas R_c is almost stable during daylight, which is most likely related to radiation. Only slight differences between the applied threshold are found. R_c slightly lower for drier conditions and higher concentrations only for short periods during daylight, for example around noon. Higher concentrations lead to lower R_c , which is probably related to the (stomatal) compensation point resulting in higher v_d . Since the impact of concentration on R_c is comparatively low, it is superimposed by slight differences induced by R_a and R_b . Thus, concentration has almost no effect on v_d . Since we had measured the ΣN_r exchange in a low nitrogen environment, nitrogen concentration in the plants was probably at low level, and thus the influence of the stomatal compensation point on the uptake of N_r species may be reduced. Zöll et al. (2019) calculated a

light response curve of ΣN_r for the same site. The increase in deposition gets lower for R_g between 300 and 500 $W m^{-2}$ and reaches a reversal point around 600 $W m^{-2}$. We find slight differences in R_c for the concentration threshold around noon, at times with highest radiation. It shows that a stomatal compensation point exists but its influence is limited by the low, ambient concentration.

A similar analysis was made for R_a and R_b . During daylight, values of R_a and R_b are close to zero showing that v_d is mostly driven by the pattern of R_c . During the night or at lower radiation, R_a and R_b are comparable in magnitude to R_c . In autumn and winter, R_c shows negative values and no diurnal pattern suggesting that uptake through the stomatal pathway is reduced, and even emission of ΣN_r occurs from canopy or soil. During these times, uptake of ΣN_r can happen through the cuticular pathway or by the soil. Deposition also appears to be driven by turbulence during these times.

The observed differences in v_d for relative humidity and temperature are related to R_a and R_b . Lower resistances were found for drier conditions (lower air humidity and higher temperature). In case of RH, also R_c exhibits slightly lower values for less humid air. Temperature has nearly no effect on R_c . As written in R1.55, the differences for v_d to micrometeorological parameters diminishes if wet leaf surfaces are excluded. The same is valid for the resistances. Wet leaf surfaces reduce the uptake of ΣN_r at the measurement site. We showed that the contribution and concentrations of N_r species, which can deposit on wet leaf surfaces, is 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. These issues may reduce the cuticular contribution to exchange processes with the canopy. Presumably, cuticular deposition is probably not as important as stomatal deposition during periods of high incident radiation, in particular from May to September. 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. Additionally, the investigation of 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 N_r (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 V_d 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 N_r 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 N_r components very useful. This would convey how the total and their contribution to total N_r changed seasonally. Also, an assessment of how well the sum of the individual N compounds compares with the total N_r 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. Concen-

trations 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 will not be shown since we found no significant deviations of the minimum and maximum TRANC ΣN_r cases to average after the bias correction of the concentrations.

The comparison of the absolute values revealed slight overestimations by DELTA+NO_x from August 2016 to October 2016 and from January to March 2017. On average, the underestimation by DELTA+NO_x is approximately $0.3 \mu\text{g N m}^{-3}$ with an standard deviation of $0.7 \mu\text{g N m}^{-3}$. The median value is about $0.35 \mu\text{g N m}^{-3}$. Fig. R9 shows monthly stacked DELTA concentrations compared to the TRANC measurements.

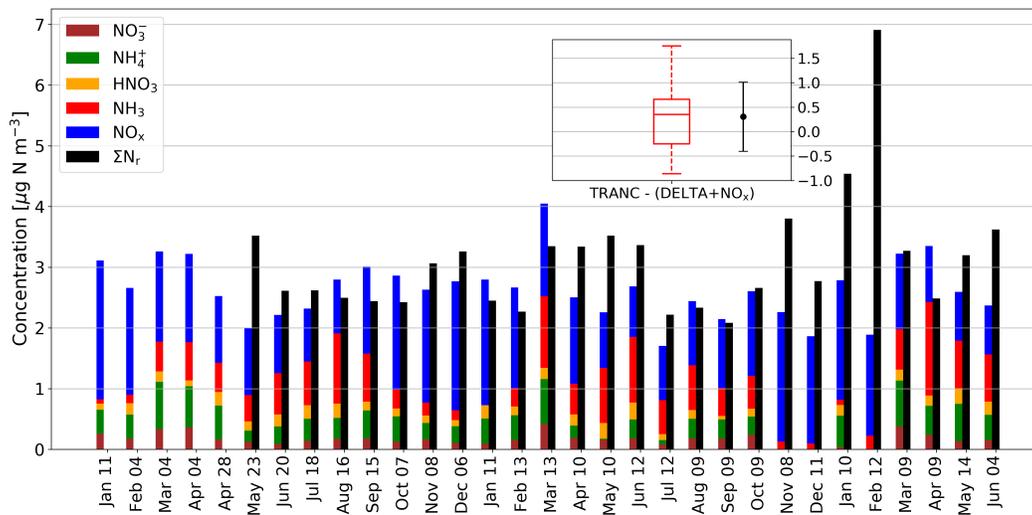


Figure R9: Monthly stacked concentration of TRANC, DELTA, and NO_x in $\mu\text{g N m}^{-3}$ for the entire measurement campaign. Colors indicate different N_r compounds. Missing NH₃ measurements from the DELTA measurements caused by a low pump flow were filled with passive sampler values from 30 m. 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.

Differences in total sums are probably related to short-term events, which could not be sufficiently captured by the DELTA system, and by a degrading of the coated denuder surfaces due to temperature and moisture. HNO₃, NH₄⁺, and NO₃⁻ concentrations are nearly equal through the entire measurement campaign. Seasonal differences exist mainly for NH₃ and NO_x. We measured average concentrations of 0.56 , 0.17 , 0.40 , 0.19 , and $1.40 \mu\text{g N m}^{-3}$ for NH₃, HNO₃, NH₄⁺, NO₃⁻, and NO_x for the entire campaign, respectively. On average, the contribution of NH₃, HNO₃, NH₄⁺, and NO₃⁻ to ΣN_r is less than 50% for the entire measurement campaign as visualized by Fig. R10. It shows that NO_x contributes most to the concentrations of ΣN_r and has a high potential to influence the exchange pattern of ΣN_r . However, seasonal differences are visible.

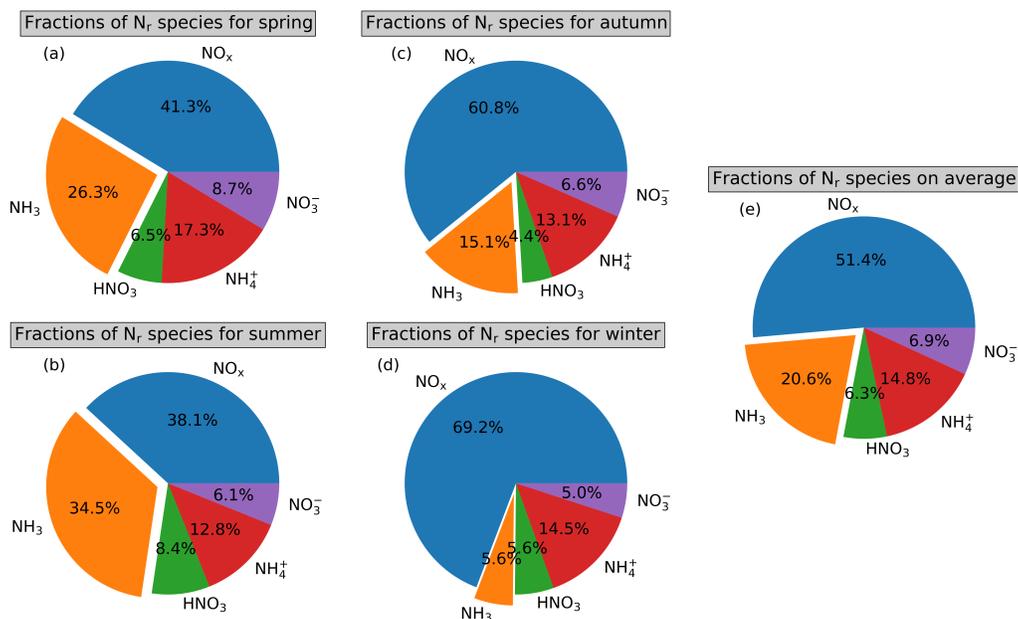


Figure R10: Pie charts showing the contribution of NO_x , NH_3 , NO_3^- , NH_4^+ , and HNO_3 to ΣN_r based on DELTA samplers and NO_x measurements for different seasons of the year. NO_x measurements are averaged to exposition periods of the DELTA samplers. (a) to (d) refer to spring, summer, autumn,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

In general, NO_x shows the highest contribution to ΣN_r and follows the expected seasonal changes with highest values during winter and lowest values in summer. NH_3 shows the expected seasonality with concentrations lowest in winter and higher values in spring and summer. The contribution of HNO_3 is almost stable. A slight increase in the contribution is found for summer. As reported by Tang et al. (2020), HNO_3 should be taken with care since HONO is included in HNO_3 signal. NO_3^- and NH_4^+ exhibit slightly higher values for spring. The changes in the composition of ΣN_r are also affecting v_d . The slight increase in HNO_3 and decrease of NH_4^+ can be related to the evaporation of NH_4NO_3 (Wyers and Duyzer, 1997; Van Oss et al., 1998).

Only slight seasonal changes in the overall ΣN_r concentration are observed. We measured 3.3, 2.6, 2.5, and $3 \mu g N m^{-3}$ for spring, summer, autumn, and winter, respectively. Consequently, it is not only the change in the overall ΣN_r concentration that influences v_d .

The comparison of the total N concentrations shows that the TRANC can sufficiently measure ΣN_r concentration. Obviously, not all components of ΣN_r are included in this comparison, for example higher oxidized components like N_2O_5 . NO_x had been measured at 51 m. Seok et al. (2013) found comparatively higher NO_x concentrations from 30 to 50 m. 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 .

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 V_d . The pollution climate the ecosystem is in is equally important and does not necessarily correlate with the ecosystem type (think of an urban woodland or a heavily grazed pasture in otherwise pristine environment). The comparison needs to be reworded. Generalisation that N_r 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 will rewrite the corresponding comparisons.

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 will reword the corresponding text and remove 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^+ are 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 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.” is certainly misleading and will be deleted. Wet deposition samplers were in close proximity to the flux tower. Thus, wash out processes also affect 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 TRANC measurements, the flux uncertainty of the gap-filled fluxes is calculated as the standard error of mean. The random uncertainty determined following (Finkelstein and Sims, 2001) will also be included in the discussion. Total from random error estimates will be calculated as square root of the sum of the squared random uncertainties according to Pastorello et al. (2020). The uncertainty discussion of DEPAC-1D 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 will improve the lines 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 will reword 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 add 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. It shows that global radiation contains most of the 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 the correlation of the input parameter with the flux. In general, correlations could also be influenced by other parameters, which have not or could not be 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 will implement the explanations given in this response in the revised version.

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 will preferentially remove smaller fluxes, which occur at night-time. 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 a gap-filling approach. The threshold was set to 0.1 ms^{-1} , and the window for filling each gap was set to ± 5 days. Uncertainties of the gap-filled fluxes are estimated by the standard error of the mean. The total uncertainties are calculated as the sum of the standard errors.

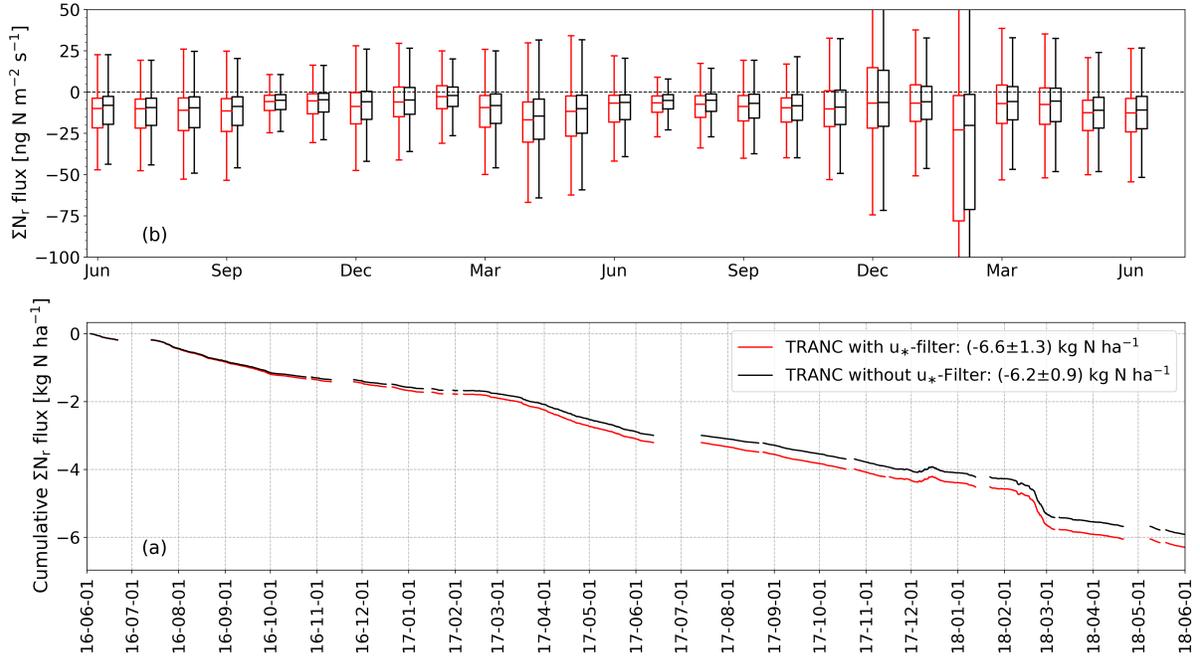


Figure R11: Panel (a) shows the non-gap filled ΣN_r fluxes depicted as box plots with (red) and without (black) u_* filter in $\text{ng N m}^{-2} \text{ s}^{-1}$ (box frame = 25% to 75% interquartile ranges (IQR), bold line = median, whisker = $1.5 \cdot \text{IQR}$). The threshold for u_* is set to 0.1 ms^{-1} . In panel (b), the cumulative dry deposition of ΣN_r is plotted for both cases in kg N ha^{-1} . Time frame is from June 2016 to June 2018.

The difference in dry deposition is approximately 400 g N ha^{-1} after 2 years and 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 are almost equal to or larger than the median depositions without u_* -filter. Apparently, we measured large and small fluxes below 0.1 ms^{-1} as seen by Fig. R4. The latter reveals that large fluxes are found for low and high u_* values. Thus, the applied u_* -threshold removes not only small fluxes resulting in a non-consistent bias between the median depositions. We further investigated the impact of temperature, humidity, and precipitation on the usage of the MDV approach compared to the MDV approach without restrictions 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 $\pm 3^\circ\text{C}$, humidity by $\pm 5\%$, or precipitation was recorded. Remaining, long-term gaps were filled by the ΣN_r monthly mean diurnal cycles estimated from the non gap-filled fluxes. Daily cycles were calculated for low and high humidity and temperature regimes separated by their monthly median. The calculations were made with and without the application of an u_* -filter. Figure R12 shows the annual dry deposition of the measurement years from the beginning of June to end of May.

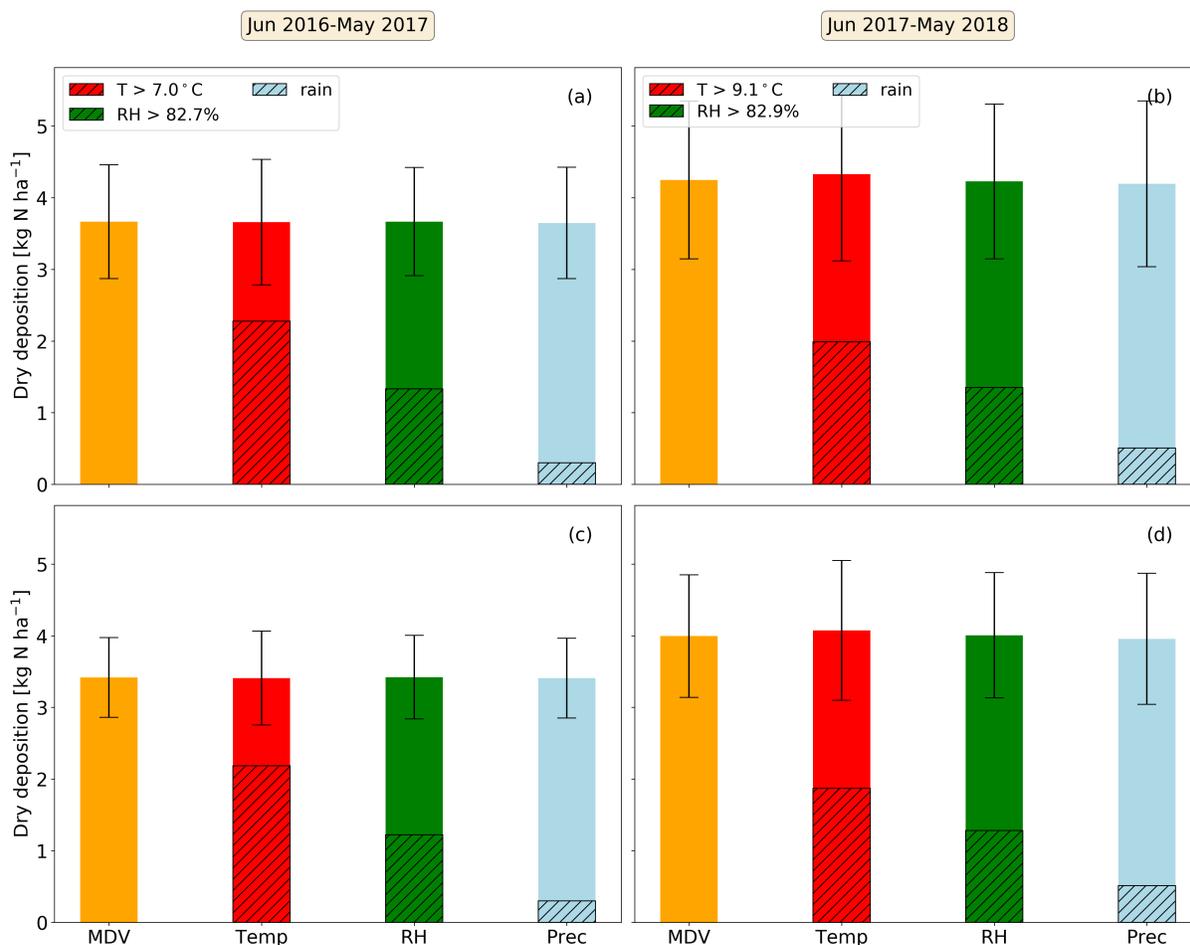


Figure R12: Annual ΣN_r dry deposition for different micrometeorological conditions compared to the original MDV approach depicted as bar graph from June to May in kg N ha^{-1} . Short-term gaps were filled with the MDV approach, long-term gaps with the averaged monthly diurnal cycles. The latter were calculated from the non gap-filled fluxes. (a) and (b) were made for fluxes with u_* -filter, (c) and (d) without u_* criteria. 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 can be found between the micrometeorological and the original MDV approach for both measurement years. Dry deposition estimated during rain is less than 500 g N ha^{-1} due to missing water soluble compounds in ΣN_r and probably lower radiation leading to closed stomata. Warm, drier conditions exhibit a higher contribution to the annual dry deposition highlighting the importance of the stomatal deposition. Although we found differences in the shapes of the diurnal patterns, especially for precipitation, no clear impact on the annual budgets was found. The diurnal patterns differed significantly only from May to September. Otherwise, low, almost constant deposition fluxes were observed for the entire day. No difference was found for fluxes measured during dry or wet conditions during those times. As shown before, the difference in the application of an u_* -filter is existent and within the uncertainty range. Dry deposition was higher in 2017/2018, which is related to the large deposition fluxes observed in February 2018. In total, we estimated $(3.7 \pm 0.8) \text{ kg N ha}^{-1}$ and $(4.2 \pm 1.1) \text{ kg N ha}^{-1}$ with the original MDV approach and u_* -filter for 2016/2017 and 2017/2018, respectively. For the revised version, we will add a

discussion of the impact of flux filters on the MDV approach.

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 average 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. In the revised version, we will add it to the method section. We will implement a detailed description of the usage of monthly DELTA concentrations and related uncertainties in the modeling study.

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 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 V_d 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 R_c . 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 were right. The drawn conclusion regarding NH₃ being responsible for the observed emission is most likely incorrect. Based on your suggestions and Reviewer 1, we will improve 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 We agree that some phrases and 2.5-year dry deposition numbers can be removed. Due to the separation of the manuscript, the abstract length will be reduced.

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 (NaNO₃), ammonium nitrate (NH₄NO₃), and ammonium sulfate ((NH₄)₂SO₄) 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 0.3mm 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 NaNO₃, NH₄NO₃, and (NH₄)₂SO₄, respectively. A comparison with a twin differential mobility particle sizer (TDMPMS) (Birmili et al., 1999) showed similar conversion efficiencies for NaNO₃ and (NH₄)₂SO₄ but differences for NH₄NO₃ (Marx et al., 2012, Fig. 6). At higher temperatures (>20°C) and relative humidity (>50%), NH₄NO₃ 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 provide the details about the aerosol conversion efficiency in the revised manuscript.

Comment R2.18 *Line 33ff. I am not aware that deposition of Nr components threatens human health. They do so by acting as precursors to PM2.5 and O3.*

Response to R2.18 We will correct the corresponding line(s).

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 fulfilled for ΣN_r and its components. Their exchange pattern is 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 or H_2O . 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 Reviewers remarks to the introduction of CTMs. Since the manuscript will be separated, the introduction will also change. The paragraphs to CTMs and DEPAC will be shifted to the modeling part, and a short introduction to v_d and resistance analysis is planned for the measurement part.

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 NH_4NO_3 ? 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 delete “resulting in an oxidization of reduced Nr compounds” and generally improved the description.

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 Median values of leaf wetness were between 1.9 and 6.2 for all sensors indicating that 50% of the leaf wetness values were close to zero for the entire measurement campaign. For daylight ($R_g > 20 \text{ W m}^{-2}$), 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 position of the medians, we set the threshold value to 10 for all sensors. A threshold of 5 lead to similar results. The threshold should be seen as highest guess, at which leaf surfaces can be considered as “wet”. The condition “wet” can be induced by the accumulation of particles or water droplets. We will improve the description of the threshold value accordingly.

Response to R2.21 Line 166ff. Please state the temporal resolution of the DELTA measurements. Also, later the text refers to ammonia diffusion samplers and NO_x measurements, which do not appear to be mentioned in Section 2.2.

Response to R2.21 The temporal resolution of the DELTA and Ferm-type passive samplers is approximately one month. We will improve the description of the DELTA measurements. NO and NO₂ measurements are mentioned in Sec. 2.2. Here, NO_x was determined by adding NO to NO₂ concentrations.

Comment R2.22 *Line 199. Does the flux loss depend on the chemical composition of Nr?*

Response to R2.22 Wintjen et al. (2020) determined flux loss factors for two different ecosystem, which exhibit differences, for example, in the composition of ΣN_r . They assumed that the differences in flux losses also related to the chemical composition of ΣN_r .

Comment R2.23 *Line 207. Please state the relative magnitude of the water correction. What is its uncertainty?*

Response to R2.23 ΣN_r interference fluxes were between 0.3 and -3 ng N m⁻²s⁻¹. The uncertainty ranges between 0.0 and 0.5 ng N m⁻²s⁻¹. Considering two years of TRANC flux measurements with MDV as gap-filling approach, the correction contributes with 132 g ha⁻¹ to the estimated dry deposition of 6.6 kg ha⁻¹.

Comment R2.24 *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 R2.24 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 3·1.96 σ . 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 are left out for preparation of the revised manuscript.

Comment R2.25 *Line 264f. How were compensation points derived from long-term measurements of SO2 and NH3? This would seem problematic.*

Response to R2.25 We agree that additional details are needed to 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 R2.26 *Line 266. Why was the LAI modelled for a site-based application? Why was this not based on a measured value?*

Response to R2.26 The LAI was not measured at the site. Please also see comment R1.11.

Comment R2.27 *Line 390. How do the diurnal cycles compare between measurements and model results? Does this shed additional light on model deficiencies?*

Response to R2.27 We appreciate the Reviewers suggestions. A comparison of measured and modeled diurnal cycles will be made for the modeling study.

Comment R2.28 Line 434. *No, concentration is not proportional flux. The flux is proportional to the concentration. The concentration is the driver.*

Response to R2.28 Revised.

Comment R2.29 Line 468. *What do the concentration ranges refer to?*

Response to R2.29 On average, $1.0 \mu\text{g m}^{-3}$ NH_3 was measured from mid of 2003 to 2005, 4 to $9 \mu\text{g m}^{-3}$ for NO_2 and 0.5 to $2 \mu\text{g m}^{-3}$ for NO . Concentration ranges refer to 1992 until the end of 2008.

Comment R2.30 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 R2.30 We agree. The sentence will be modified accordingly.

Comment R2.31 Line 507. *The DELTA samplers does not measure NOx.*

Response to R2.31 Agreed. NO_x was measured by the NPBW.

Comment R2.32 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 R2.32 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^+ . The description of the coatings will be added to the method section.

Comment R2.33 Line 551. *Presumably in addition to total Nr concentration, its speciation also affects the net deposition rate and thus the flux.*

Response to R2.33 Probably, yes. Therefore we would like to add the following sentence: “The different ΣN_r deposition rates reported in the mentioned publications is may be also related to differences in the composition of ΣN_r .”

Comment R2.34 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 R2.34 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.

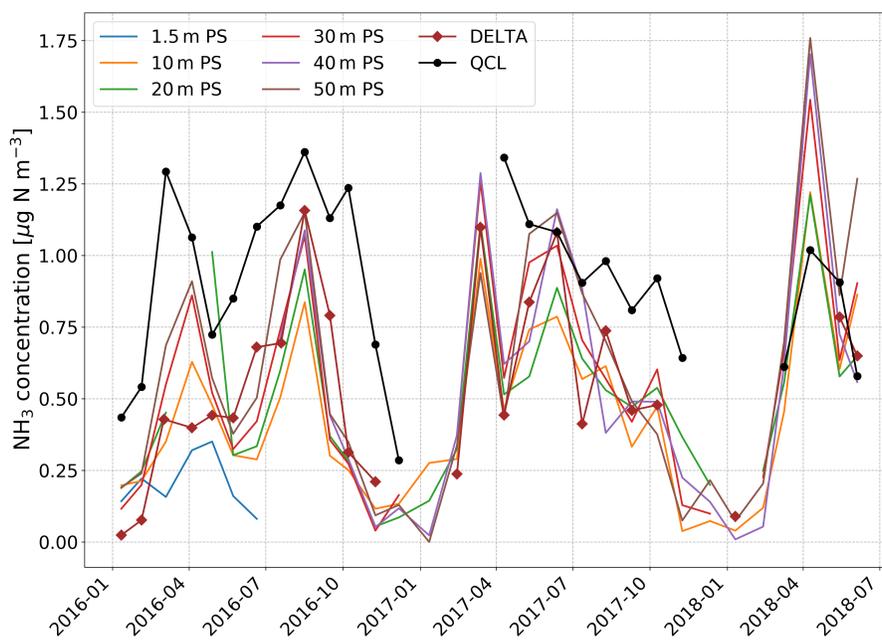


Figure R13: Concentrations of NH_3 measured by the DELTA and passive samplers, and the QCL in $\mu\text{g N m}^{-3}$. NH_3 of the QCL was averaged to the exposition period of the long-term samplers. Colors of the passive samplers indicate different measurement heights.

Overall, the agreement in the yearly pattern is good, but a bias between the QCL and the diffusion samplers is obvious. From passive sampler measurements, an increase in the NH_3 concentration with measurement height can be found. At 10 m (in the canopy), the lowest NH_3 concentrations were measured. No systematic difference was found between 20 m and 30 m due to uptake and turbulent mixing processes at the top of the canopy. At 50 m, NH_3 was slightly higher ($0.1 \mu\text{g N m}^{-3}$) than 30 m, which might be related to an inhibited air-mass exchange caused by atmospheric stratification. During wintertime, the difference in measurement heights diminishes. Slightly higher NH_3 concentration were observed at 10 m. 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 $\text{TRANC } \Sigma \text{N}_r$ will be made for the modeling part.

From R2.35 to R2.40, suggested modifications to the text and recommendations of the Reviewer are related to the modeling part and will be implemented in second manuscript.

Comment R2.35 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 R2.35 The deviation in u_* is not related to the topography. u_* is calculated with the wind speed given at the reference height. As written in the manuscript, the reference height of LOTOS-EUROS is 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_* can be expected.

Comment R2.36 *Line 754. “input NH₃ concentrations” Do you refer to emissions or long-range transport?*

Response to R2.36 For our measurement site, the elevated NH₃ concentrations are most likely caused by emissions from nearby agriculture.

Comment R2.37 *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 R2.37 Agreed.

Comment R2.38 *Line 793f. But you say the V_d 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 R2.38 In case of the modeled v_d , yes. v_d of ΣN_r is significantly lower than the modeled v_d of NH₃ and closer to v_d of NO₂. Figure R10 reveals that NO_x, in particular NO₂, was the dominant N_r species and not NH₃. Presumably, a measured v_d of NH₃ would have been lower than modeled values.

Comment R2.39 *Line 795f. The wash-out could have occurred upwind and not contributed to the local wet deposition.*

Response to R2.39 Agreed. The sentence will be removed as written in R2.5.

Comment R2.40 *Line 798f. The good agreement seems entirely fortuitous given aerosol was not included in DEPAC-1D ...*

Response to R2.40 We work on including of NH₄⁺ and NO₃⁻ in DEPAC-1D for the modeling study.

Comment R2.41 *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 R2.41 We appreciate the Reviewer's suggestion for rephrasing and will modify the sentence accordingly.

Technical corrections / suggestions:

Comment R2.42 *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 R2.42 We will change the beginning of the corresponding sentences.

Comment R2.43 *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 R2.43 We will go carefully through the text and add articles if necessary.

Response to R2.43 *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 R2.43 We will check corresponding lines and replace “after” by appropriate words.

Comment R2.44 *General: in many cases units are incorrectly combined. For example ms-1 should read m s-1 and μgm^{-3} should read $\mu\text{g m}^{-3}$.*

Response to R2.44 We will improve the notation of the units and separate them correctly.

Comment R2.45 *Line 7. I was surprised to see N_r concentration given in ppb rather than $\mu\text{g N m}^{-3}$, especially since N_r contains aerosol compounds for which the use of ppb is rather unusual.*

Response to R2.45 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 . For the comparison to the DELTA measurements or the calculation of resistances, we changed the unit to $\mu\text{g N m}^{-3}$.

Comment R2.46 *Line 62. Better “EC studies of ...”*

Response to R2.46 Revised.

Comment R2.47 *Line 69 refers to “that site”, but it is not clear which site is meant at this point.*

Response to R2.47 It will be replaced by “conducted with the same instrumentation at the measurement site”.

Comment R2.48 *Line 96. “validation with flux measurements” (or “against”).*

Response to R2.48 Revised.

Comment R2.49 *Line 116. “Measurements were carried out in”. Actually, the authors should consider the alternative “Measurements were made” here and elsewhere.*

Response to R2.49 Revised. We will consider a rephrasing of the corresponding lines.

Comment R2.50 *Line 117. Remove “and”.*

Response to R2.50 Revised.

Comment R2.51 *Line 130. Remove “which is remote from significant sources of emissions.” This is repeating what was said before.*

Response to R2.51 Agreed.

Comment R2.52 *Line 139. “which was housed in an”*

Response to R2.52 Changed.

Comment R2.53 *Line 142. “oxidation”*

Response to R2.53 Changed.

Comment R2.54 *Line 145. “during which remaining oxidised N_r species”*

Response to R2.54 Changed.

Comment R2.55 *Line 219. “was caused by”*

Response to R2.55 Changed.

Comment R2.56 *Line 249. “filling the gaps in the flux data.”*

Response to R2.56 Changed.

Comment R2.57 *Line 274. “weighted using the actual land-use fractions” ?*

Response to R2.57 Agreed.

Comment R2.58 *Line 275. “when considering only deciduous”*

Response to R2.58 Revised.

Comment R2.59 *Section 3.1. Much of the section here and elsewhere should be put into past tense.*

Response to R2.59 Agreed.

Comment R2.60 *Line 303 and elsewhere. Please add charges to NO₃⁻ and NH₄⁺ (NO₃ is a radical).*

Response to R2.60 Charges will be added.

Comment R2.61 *Line 305. Redundant “with”*

Response to R2.61 Removed.

Comment R2.62 *Line 308. “the relative contribution of NH₃ is significantly higher”*

Response to R2.62 Agreed.

Comment R2.63 *Line 310 and elsewhere. A colon is followed by lower case in English.*

Response to R2.63 Revised.

Comment R2.64 *Line 311 “done following the criteria mentioned”*

Response to R2.64 Revised.

Comment R2.65 *Line 380 & 447. Should be “consequently” instead of “consequentially”*

Response to R2.65 Changed.

Comment R2.66 *Line 384. Should the units here be “kg N ha⁻¹ a⁻¹”?*

Response to R2.66 Yes, a N is missing here.

Comment R2.67 *Line 391. “Clearly, ...”*

Response to R2.67 Changed.

Comment R2.68 *Figure 6. The colours between upper and lower CBT estimate seem to be reversed.*

Response to R2.68 We agree. Colors will be switched.

Comment R2.69 *Line 417 and also line 816. “the range of ...”*

Response to R2.69 Changed.

Comment R2.70 *Line 450. “LOTOS-EUROS states out NH₃ ...” – meaning unclear.*

Response to R2.70 Sentence will be modified as follows: “LOTOS-EUROS determines NH₃ as the main contributor to ΣN_r ”.

Comment R2.71 *Line 479. “Apart from management events, fluxes above the arable ...”*

Response to R2.71 Changed.

Comment R2.72 *Line 528. “Munger et al. (1995) also made NO_y flux measurements ...”*

Response to R2.72 Changed.

Comment R2.73 *Line 607. “sometimes lead to non-stationarities”*

Response to R2.73 Changed.

Comment R2.74 *Line 612 “under certain circumstances”*

Response to R2.74 Changed.

Comment R2.75 *Conclusions. Re-introduce all acronyms, including Nr.*

Response to R2.75 In the revised version, acronyms will be 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. Since an investigation of resistances and v_d will be implemented in the revised version, equations needed to follow the resistance analysis will be given. The same will be done for the modeling study.

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 NH₃ measurement techniques (see R2.34). 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 NH₄⁺ and NH₃ concentrations were obtained from DELTA measurements. During the warmer month, NH₃ concentrations were higher than concentrations measured for NH₄⁺. From November to February, the situation is vice-versa. However, due to the denuder's low time resolution, we had no possibility to derive an influence of NH₄⁺ and NH₃ on stomatal

processes, which happen on a shorter time-scale.

References

- Ammann, C., Wolff, V., Marx, O., Brümmer, C., and Neftel, A. (2012). Measuring the biosphere-atmosphere exchange of total reactive nitrogen by eddy covariance. *Biogeosciences*, 9(11):4247–4261.
- Birmili, W., Stratmann, F., and Wiedensohler, A. (1999). Design of a DMA-based size spectrometer for a large particle size range and stable operation. *Journal of Aerosol Science*, 30(4):549–553.
- Bokhorst, S., Metcalfe, D. B., and Wardle, D. A. (2013). Reduction in snow depth negatively affects decomposers but impact on decomposition rates is substrate dependent. *Soil Biology and Biochemistry*, 62:157–164.
- Brümmer, C., Marx, O., Kutsch, W., Ammann, C., Wolff, V., Flechard, C. R., and Freibauer, A. (2013). Fluxes of total reactive atmospheric nitrogen (ΣN_r) using eddy covariance above arable land. *Tellus B: Chemical and Physical Meteorology*, 65(1):19770.
- Burkhardt, J., Peters, K., and Crossley, A. (1995). The presence of structural surface waxes on coniferous needles affects the pattern of dry deposition of fine particles. *Journal of Experimental Botany*, 46(288):823–831.
- Dämmgen, U., Thöni, L., Lump, R., Gilke, K., Seitler, E., and Bullinger, M. (2010). *Feldexperiment zum Methodenvergleich von Ammoniak- und Ammonium-Konzentrationsmessungen in der Umgebungsluft, 2005 bis 2008 in Braunschweig*, volume 337 of *Landbauforschung : Sonderheft*. Johann Heinrich von Thünen-Institut, Braunschweig. Jahresberichts-kategorie: 10-M4;10-3.
- Durham, J. and Stockburger, L. (1986). Nitric acid-air diffusion coefficient: Experimental determination. *Atmospheric Environment (1967)*, 20(3):559–563.
- Dyer, A. J. and Hicks, B. B. (1970). Flux-gradient relationships in the constant flux layer. *Quarterly Journal of the Royal Meteorological Society*, 96(410):715–721.
- Farquhar, G. D., Firth, P. M., Wetselaar, R., and Weir, B. (1980). On the gaseous exchange of ammonia between leaves and the environment - determination of the ammonia compensation point. *Plant Physiology*, 66(4):710–714.
- Ferm, M. (1991). *A Sensitive Diffusional Sampler*. Report L91-172. Swedish Environmental Research Institute, Gothenburg.
- Finkelstein, P. L. and Sims, P. F. (2001). Sampling error in eddy correlation flux measurements. *Journal of Geophysical Research: Atmospheres*, 106(D4):3503–3509.
- Garland, J. A. (1977). The dry deposition of sulphur dioxide to land and water surfaces. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 354(1678):245–268.
- Hansen, K., Pryor, S. C., Boegh, E., Hornsby, K. E., Jensen, B., and Sorensen, L. L. (2015). Background concentrations and fluxes of atmospheric ammonia over a deciduous forest. *Agricultural and Forest Meteorology*, 214-215:380–392.

- Hansen, K., Sorensen, L. L., Hertel, O., Geels, C., Skjoth, C. A., Jensen, B., and Boegh, E. (2013). Ammonia emissions from deciduous forest after leaf fall. *Biogeosciences*, 10(7):4577–4589.
- Hurkuck, M., Brümmer, C., Mohr, K., Grünhage, L., Flessa, H., and Kutsch, W. L. (2014). Determination of atmospheric nitrogen deposition to a semi-natural peat bog site in an intensively managed agricultural landscape. *Atmospheric Environment*, 97:296–309.
- Jarvis, P. G. (1976). The interpretation of the variations in leaf water potential and stomatal conductance found in canopies in the field. *Philosophical Transactions of the Royal Society B: Biological Sciences*, 273(927):593–610.
- Jensen, N. and Hummelshøj, P. (1997). Derivation of canopy resistance for water vapor fluxes over a spruce forest, using a new technique for the viscous sublayer resistance (correction to vol. 73, p. 339, 1995). *Agricultural and Forest Meteorology*, 85(3-4):289.
- Jensen, N. O. and Hummelshøj, P. (1995). Derivation of canopy resistance for water-vapor fluxes over a spruce forest, using a new technique for the viscous sublayer resistance. *Agricultural and Forest Meteorology*, 73(3-4):339–352.
- Kreyling, J., Haei, M., and Laudon, H. (2013). Snow removal reduces annual cellulose decomposition in a riparian boreal forest. *Canadian Journal of Soil Science*, 93(4):427 – 433.
- Marx, O., Brümmer, C., Ammann, C., Wolff, V., and Freibauer, A. (2012). TRANC – a novel fast-response converter to measure total reactive atmospheric nitrogen. *Atmospheric Measurement Techniques*, 5(5):1045–1057.
- Massman, W. J. (1998). A review of the molecular diffusivities of H₂O, CO₂, CH₄, CO, O₃, SO₂, NH₃, N₂O, NO, and NO₂ in air, O₂ and N₂ near STP. *Atmospheric Environment*, 32(6):1111–1127.
- Mauder, M. and Foken, T. (2006). Impact of post-field data processing on eddy covariance flux estimates and energy balance closure. *Meteorologische Zeitschrift*, 15(6):597–609.
- Medinets, S., Gasche, R., Skiba, U., Schindlbacher, A., Kiese, R., and Butterbach-Bahl, K. (2016). Cold season soil NO fluxes from a temperate forest: drivers and contribution to annual budgets. *Environmental Research Letters*, 11(11):114012.
- Milford, C., Hargreaves, K. J., Sutton, M. A., Loubet, B., and Cellier, P. (2001). Fluxes of NH₃ and CO₂ over upland moorland in the vicinity of agricultural land. *Journal of Geophysical Research: Atmospheres*, 106(D20):24169–24181.
- Moffat, A. M., Beckstein, C., Churkina, G., Mund, M., and Heimann, M. (2010). Characterization of ecosystem responses to climatic controls using artificial neural networks. *Global Change Biology*, 16(10):2737–2749.
- Nemitz, E., Dorsey, J. R., Flynn, M. J., Gallagher, M. W., Hensen, A., Erisman, J. W., Owen, S. M., Dämmgen, U., and Sutton, M. A. (2009). Aerosol fluxes and particle growth above managed grassland. *Biogeosciences*, 6(8):1627–1645.
- Pastorello, G., Trotta, C., and Canfora, E. e. a. (2020). The FLUXNET2015 dataset and the ONEFlux processing pipeline for eddy covariance data. *Scientific Data*, 7(1):225.

- Paulson, C. A. (1970). The mathematical representation of wind speed and temperature profiles in the unstable atmospheric surface layer. *Journal of Applied Meteorology*, 9(6):857–861.
- Peake, E. and Legge, A. H. (1987). Evaluation of methods used to collect air quality data at remote and rural sites in Alberta, Canada. In *Proc. 1987 EPA/APCA Symposium on Measurements of Toxic and Related Air Pollutants*. APCA.
- Peake, M. (1985). A preliminary report on the design and testing of the KAPS (Kananaskis Atmospheric Pollutant Sampler) for the collection of acidic and basic gases and fine particles. Document 0012e/July 8/85. Typskript University Calgary.
- Saccone, P., Morin, S., Baptist, F., Bonneville, J.-M., Colace, M.-P., Domine, F., Faure, M., Geremia, R., Lochet, J., Poly, F., Lavorel, S., and Clément, J.-C. (2013). The effects of snowpack properties and plant strategies on litter decomposition during winter in subalpine meadows. *Plant and Soil*, 363(1):215–229.
- Schrader, F. and Brümmer, C. (2014). Land use specific ammonia deposition velocities: a review of recent studies (2004–2013). *Water, Air, and Soil Pollution*, 225(10):2114.
- Seok, B., Helmig, D., Ganzeveld, L., Williams, M. W., and Vogel, C. S. (2013). Dynamics of nitrogen oxides and ozone above and within a mixed hardwood forest in northern michigan. *Atmospheric Chemistry and Physics*, 13(15):7301–7320.
- Tang, Y. S., Flechard, C. R., Dämmgen, U., Vidic, S., Djuricic, V., Mitosinkova, M., Uggerud, H. T., Sanz, M. J., Simmons, I., Dragosits, U., Nemitz, E., Twigg, M., van Dijk, N., Fauvel, Y., Sanz-Sanchez, F., Ferm, M., Perrino, C., Catrambone, M., Leaver, D., Braban, C. F., Cape, J. N., Heal, M. R., and Sutton, M. A. (2020). Pan-european rural atmospheric monitoring network shows dominance of NH_3 gas and NH_4NO_3 aerosol in inorganic pollution load. *Atmospheric Chemistry and Physics Discussions*, 2020:1–61.
- Taylor, B. and Jones, H. (1990). Litter decomposition under snow cover in a balsam fir forest. *Canadian Journal of Botany*, 68:112–120.
- Thonen, B., Rennenberg, H., and Weber, P. (1996). Absorption of atmospheric NO_2 by spruce (*picea abies*) trees. *New Phytologist*, 134(2):257–266.
- Van Oss, R., Duyzer, J., and Wyers, P. (1998). The influence of gas-to-particle conversion on measurements of ammonia exchange over forest. *Atmospheric Environment*, 32(3):465 – 471.
- van Zanten, M. C., Sauter, F. J., Wichink Kruit, R. J., van Jaarsveld, J. A., and van Pul, W. A. J. (2010). Description of the DEPAC module; Dry deposition modeling with DEPAC_GCN2010. Technical report, RIVM, Bilthoven, NL.
- Webb, E. K. (1970). Profile relationships: The log-linear range, and extension to strong stability. *Quarterly Journal of the Royal Meteorological Society*, 96(407):67–90.
- Wexler, A. S. and Seinfeld, J. H. (1991). Second-generation inorganic aerosol model. *Atmospheric Environment. Part A. General Topics*, 25(12):2731–2748.
- Wichink Kruit, R. J., van Pul, W. A. J., Sauter, F. J., van den Broek, M., Nemitz, E., Sutton, M. A., Krol, M., and Holtslag, A. A. M. (2010). Modeling the surface–atmosphere exchange of ammonia. *Atmospheric Environment*, 44(7):945–957.

- Wintjen, P., Ammann, C., Schrader, F., and Brümmner, C. (2020). Correcting high-frequency losses of reactive nitrogen flux measurements. *Atmospheric Measurement Techniques*, 13(6):2923–2948.
- Wolff, V., Trebs, I., Foken, T., and Meixner, F. X. (2010). Exchange of reactive nitrogen compounds: concentrations and fluxes of total ammonium and total nitrate above a spruce canopy. *Biogeosciences*, 7(5):1729–1744.
- Wyers, G. and Duyzer, J. (1997). Micrometeorological measurement of the dry deposition flux of sulphate and nitrate aerosols to coniferous forest. *Atmospheric Environment*, 31(3):333 – 343.
- Wyers, G. P. and Erisman, J. W. (1998). Ammonia exchange over coniferous forest. *Atmospheric Environment*, 32(3):441–451.
- Zöll, U., Lucas-Moffat, A. M., Wintjen, P., Schrader, F., Beudert, B., and Brümmner, C. (2019). Is the biosphere-atmosphere exchange of total reactive nitrogen above forest driven by the same factors as carbon dioxide? an analysis using artificial neural networks. *Atmospheric Environment*, 206:108–118.