Response to referees' comments – manuscript BG-2022-72 Forest-atmosphere exchange of reactive nitrogen in a low polluted area – Part II: Modeling annual budgets

We thank the anonymous referees for their comments to the manuscript. We recognized that the structure of the manuscript needs improvements. The discussion on fluxes is rather quantitative, and descriptions of the results are too detailed making a comprehensive understanding challenging. Based on the reviewers' suggestions, we will shorten the manuscript and improve the discussion.

Comments of Referee 1 range from R1.1 to R1.21, Comments of Referee 2 range from R2.1 to R2.44. Line numbers in the answers, where new information will be added to the manuscript, refer to the original submitted version. Text marked in red will be deleted, text marked in blue will be implemented in the manuscript.

Response to Referee 1

General Comment: This study provides dry deposition estimates of total reactive nitrogen at a mixed forest site using four different measurement and modeling methods. Annual and seasonal concentrations, dry deposition velocities and fluxes were discussed and compared between different methods. The study provides a useful dataset and some useful findings in terms of modeling uncertainties using different approaches. The presentation quality needs improvement as detailed below.

Response to R1.1: We thank the Reviewer for his/her comments to the submitted manuscript. As written above, we agree that the structure of the manuscript needs to be improved as outlined below.

Specific comments

Comment R1.2: The second paragraph of the Abstract needs to better summarize major findings.
Response to R1.2: We agree. Please note the comments R1.3 to R1.6.

Comment R1.3: Lines 20-21: this sentence is not clear, hard to tell which flux number is from which method. You mentioned DEPAC-1D method in this sentence, and the next sentence provides a flux number again using this method.
Response to R1.3: We agree that the sentences are difficult to understand. In a first approach, the mean diurnal variation (MDV) was applied to short-term gaps and DEPAC-1D to remaining, long-term gaps. In a second approach, only DEPAC-1D was used for gap-filling. We will replace using DEPAC-1D only, and the Mean-Diurnal-Variation method in combination with DEPAC-1D as gap-filling approaches, respectively by depending on the gap-filling approach.

Comment R1.4: Lines 24-27: while v_d parameterization certainly needs some improvements, partitioning among the different N_r species in the total N_r budget might be the dominant factor for the temperature-dependent v_d in this case. This issue can be discussed more in section 3.2, but this statement may not be needed in the Abstract because such an statement provides little useful information.
Response to R1.4: The difference in the parametrization of v_d during that time was found to be a key issue for the overestimation in ΣN_r fluxes of DEPAC-1D. Therefore, we think this statement should be in the abstract. However, the subclause leading to the conclusion that the parametrizations may need revision is not needed here and will be deleted.

Comment R1.5: Lines 31: It is better to first present the dominant N species in the measured concentrations (and Nr flux if available) before discussing modeling results.
Response to R1.5: The contribution of the measured N_r species to ΣN_r is discussed in the previous study (Wintjen et al., 2022) and written in its abstract. Therefore, we think a repetition is not needed here. In line 31ff, we want to highlight the reason for the overestimation of LOTOS-EUROS compared to TRANC fluxes.

Comment R1.6: Lines 36-37: this single sentence should not be a separate paragraph; it can be presented in the first paragraph of the Abstract or at the end of the Introduction.
Response to R1.6: We decided to delete this sentence.

Comment R1.7: Section 2.1 and line 131: I assume most of the materials in this section were already reported in your previous study. If this is the case, this section can be simplified since the manuscript is very long. The same recommendation applies to other sections where applicable.

Response to R1.7: We agree that this section provides information, which was already given in Wintjen et al. (2022). Thus, we decided to rewrite the entire Sec. 2.1 and entitle as it "Dataset description". In this section, we will provide a table giving an overview about each method with input and output parameters. Information on sample resolution and data coverage will be added. We will replace Sec. 2.1 in the originally submitted version with the text given in this response. Information on the gap-filling of TRANC and DEPAC-1D (lines 217 to 235) will be moved to Sec. 2.1.

Table 1 Overview of all methods used for estimating ΣN_r dry deposition.

Method	**Primary input variables and temporal resolution**	**Primary output variables and temporal resolution**
TRANC	Wind components (u,v,w), sonic temperature (T_s), and ΣN_r concentration at 10 Hz resolution	Fluxes of ΣN_r flux at half-hourly resolution
DEPAC-1D	Measurements of micrometeorological variables at half-hourly resolution	Fluxes of NH_3, NO_2, NO, HNO_3, and particulate NH_4^+ and NO_3^- at half-hourly resolution
	NH_3, NO, NO_2 at half-hourly resolution	
	SO_2, HNO_3, NH_3, and particulate NO_3^- and NH_4^+ at monthly resolution	
TRANC (MDV+DEPAC-1D)		A combination of MDV and DEPAC-1D for adding missing fluxes
TRANC (DEPAC-1D)		Only DEPAC-1D is used for gap-filling
LOTOS-EUROS	Micrometeorological data from ECMWF weather forecasts and modeled concentrations of SO_2, NH_3, NO_2, NO, HNO_3, and particulate NH_4^+ and NO_3^- at hourly resolution	Fluxes of NH_3, NO_2, NO, HNO_3, and particulate NH_4^+ and NO_3^- at hourly resolution
Canopy budget technique	Samples from throughfall measurements collected in weekly intervals	Dissolved inorganic nitrogen fluxes consisting of NO_3^- and NH_4^+ ions on monthly/annual basis

“For the comparison to modeled ΣN_r deposition fluxes, TRANC eddy-covariance flux measurements described in Wintjen et al. (2022) in detail were used. These flux measurements were available at half-hourly resolution and had a data coverage of 41.0 % considering the entire campaign period. Data gaps were related to violations of the EC theory and performance issues of the instruments.

For the application of DEPAC-1D, time series of micrometeorological parameters (i.e. temperature, atmospheric pressure, relative humidity, global radiation, Obukhov length, friction velocity) and air pollutant concentrations (NO, NO_2, HNO_3, NH_3, NO_3^-, NH_4^+, and sulphur dioxide (SO_2)) are required for flux calculations. NH_3, NO_2, and NO concentrations obtained from Quantum cascade laser and chemiluminescence measurements as well as micrometeorological parameters were available at half-hourly resolution, whereas the remaining N_r species and NH_3 were obtained from DELTA (DEnuder for Long-Term Atmospheric sampling e.g., Sutton et al., 2001; Tang et al., 2009) and passive sampler measurements of the IVL type (Ferm, 1991) for NH_3 only on monthly basis. Gaps in DEPAC-1D were mostly related to power outages causing gaps in micrometeorological data and issues in the measurements of N_r compounds. Respective half-hourly values in the flux time series of each gas (approx. 3.4% for NH_3, HNO_3, NH_4^+, and NO_3^- and 9.3% for NO and NO_2) were filled with results from LOTOS-EUROS. A detailed description to the site and the instrumentation is given in Wintjen et al. (2022). For LOTOS-EUROS flux modeling, modeled input variables were used to predict deposition fluxes for NO, NO_2, HNO_3, NH_3, NO_3^-, and NH_4^+. LOTOS-EUROS fluxes were resampled to half-hourly timestamps and missing fluxes were linearly interpolated. Throughfall measurements were regularly taken in weekly intervals and used for determination of total inorganic nitrogen on annual basis. An overview of all methods is given in Table 1.

To compare dry deposition estimates from modeling to TRANC measurements, we filled gaps in the TRANC flux data given with results from DEPAC-1D and henceforth, called this dataset TRANC(DEPAC-1D). In a second approach, we applied the mean-diurnal-variation (MDV) method to short-term gaps analogous to Wintjen et al. (2022) and replaced remaining gaps with results from DEPAC-1D. This approach was called TRANC(MDV+DEPAC-1D) Both approaches, DEPAC-1D alone and the combination of DEPAC-1D and MDV, were able to fill all gaps in flux time series. Uncertainties of the gap-filled fluxes determined by MDV were calculated as the standard error of the mean. Cumulative uncertainties of TRANC fluxes solely based on the uncertainty of the gap-filling and were calculated according to Eq. (3) of Wintjen et al. (2022). The error calculation scheme proposed by Brümmer et al. (2022) (Eq. (1)) was applied to fluxes filled with DEPAC-1D. Flux uncertainty of those half-hours was given as

$$F_{\text{unc,DEPAC}-1\text{D}} = \frac{\tilde{X}}{F_{\text{DEPAC}-1\text{D}}}\ ;\ \text{with}\ \tilde{X} = \frac{F_{\text{unc,meas}}}{F_{\text{meas}}} \quad \textbf{(1)}$$

where $\tilde{X}$ represents the median of the ratio of the uncertainty of the measured fluxes ($F_{unc,meas}$) to their corresponding flux values (F_{meas}). The uncertainty of the measured fluxes was given by Finkelstein and Sims (2001). Systematic uncertainties were not accounted in the error calculation. A discussion on systematic uncertainties is given in Wintjen et al. (2022).”

Comment R1.8: Line 136: explicitly show which species (only NH3?) uses the bi-directional approach.
Response to R1.8: In DEPAC, a compensation is only implemented for NH_3. This sentence will be added to line 138.

Comment R1.9: Line 138: “as” should be “and”.
Response to R1.9: Agreed.

Comment R1.10: Line 139: this part is not clear.
Response to R.1.10: We will add brackets to *z-d*.

Comment R1.11: Line 169: why choose 2.5 m knowing that the measurement is at 30 m? In most regional scale CTMs, dry deposition is typically calculated at the mid-layer height of the first model layer (typically at 10-40 m) while some model may also use 10 m.
Response to R1.11: The land-use specific and total dry deposition were calculated by LOTOS-EUROS on hourly basis for NH_3, NO, NO_2, HNO_3, NO_3^-, and NH_4^+. In the model, the dry deposition velocity and flux are calculated for the mid-layer height of the first model layer, which has a depth of ca. 20m. By assuming a constant flux and using the stability parameters, the concentrations can be diagnosed for the canopy top and the typical observation height (2.5 m above roughness length (z_0)) in air quality networks. We will add this information to line 169.

Comment R1.12: Line 170: most CTMs would have many more NO_y species than listed here (PAN, MPAN, etc.). Are these species not available in this model?
Response to R1.12: Line 170 will be rephrased as follows: On hourly basis, the land-use specific and total dry deposition were calculated in LOTOS-EUROS by applying DEPAC for NH_3, NO, NO_2, HNO_3, dry deposition of particles (NO_3^-, and NH_4^+) was calculated according to Zhang et al. (2001) (see Manders-Groot et al. (2016, Sec. 5.2).

PAN is of course in the model but is not considered in DEPAC. Hence, it is removed after it decomposes. Note that trial calculations including PAN, NO_3 radical, and N_2O_5 dry deposition showed that these contribute less than 1% to the total flux.

Comment R1.12: Line 182, Are the resistance formulas in DEPAC-1D (Section 2.2.3) the same as those in DEPAC (Section 2.2.1)?
Response to R1.12: Yes, except for R_a and R_b. They are not provided by DEPAC and determined externally.

Comment R1.13: Lines 171 and 187: This is a size-resolved v_d model. What size distributions were used for particle nitrate and ammonium when calculating their v_d?
Response to R1.13: For the fine fraction of NO_3^- and NH_4^+, a mass median diameter of 0.7 µm was used. For the coarse fraction of NO_3^-, 8 µm was taken.

Comment R1.14: Simplify Section 2.2 if possible since the manuscript is very long and the description here is a bit too long.
Response to R1.14: Please note the response to R1.7. Lines 217 to 235 will be shortened and moved to Sec 2.1.

Comment R1.15: Section 2.3: Is direct DON wet deposition considered in this budget? You may also want to briefly mention the important contribution of the dry deposition of organic N in the total Nr dry deposition budget somewhere in the manuscript.
Response to R1.15: DON is accounted in wet deposition measurements (l243f). The CBT approach focuses on inorganic N species. The „contribution of the dry deposition of organic N in the total Nr dry deposition“ is not directly measured by CBT. Dry deposition of ΔDON could had been happened, but it may be also caused by microbial conversion of inorganic N_r or by leaching from textures. We are not able to draw a firm conclusion about the contribution organic N to dry deposition of ΣN_r. Please note the responses to comments R2.17 to R2.19.

Comment R1.16: Section 3.1: With too many details (such as seasonal high and low values), but lacking of a summary of the big picture, makes the section difficult to read. I assume you can take the measured concentrations as the benchmark, and then evaluate model performance. Then you should first present the annual (and seasonal where needed) average concentrations for each N species from the measurements (this way the dominant species and their relative contributions to the total Nr can be easily observed), then mention the model-measurement differences.

Response to R1.16: We agree that the current structure of Sec. 3.1 makes the reading difficult since the level of detail in the results section is rather high and thus it is challenging to take conclusions from Sec. 3.1. We appreciate your suggestions to the structure of Sec. 3.1 and will change the way of presenting facts according to your suggestions.

Comment R1.17: Section 3.1: Figure 1: You may just show statistics in the main body of the paper and move the time series of the data to SI to avoid a too-crowed picture. The same comment applies to other similar figures.

Response to R1.17: In order to solve this issue, we will change the y-axis limits of corresponding figures like Fig. 1 and Fig. 3 making the differences between the medians more visible.

Comment R1.18: Section 3.2-3.4: v_d is compared in two sections while Nr fluxes are compared in three sections. Why not use one section focusing on v_d comparisons and another section focusing on Nr flux comparison? v_d comparison can be very brief (knowing that v_d modeling has larger uncertainties from literature, e.g., Flechard et al, 2001 cited in this study).
Response to R1.18: We thought a separation by model is appropriate for a manuscript highlighting performance of flux modeling. However, we agree with the Referee to change the structure of the sections making a section for comparisons on v_d and fluxes. We will further shorten the description of the figures, in particular Figures S4 and S5 (Lines 304 to 328 and 368 to 405), thereby focusing on the main messages of these figures.

Comment R1.18: My comments above on section 3.1 also apply to other sections below. A general impression I have is that: there are too many details in the results and discussion, but a good summary of the major findings is lacking.
Response to R1.18: As written in the responses above, changes to the results section will me made including a reduction in details and a presenting of the important messages. Please also note the responses to R2.22 and R2.28.

Comment R1.19: Sections 4.2 and 4.3: Discussions on methodology uncertainties are important in any studies, but detailed discussions on modeling uncertainties (and without firm conclusions) like the ones presented here seem to be out of the scope of the present study.
Comment R1.19: In Sections 4.2 and 4.3, we elaborated the current uncertainties in flux modeling of DEPAC-1D and LOTOS-EUROS which probably caused the discrepancies to TRANC flux measurements. We think that it necessary to highlight these uncertainties making readers, developers, and potential users aware of them. However, we agree that the discussion, in particular Sec. 4.2, should be more focused on the uncertainties of DEPAC-1D than on the overall modeling performance (see R2.35).

Comment R1.20: Lines 819-824: No need to repeat what you have done in the conclusion section.
Response to R1.20: We will delete these lines and additionally rephrase parts of the conclusion, which are likely a repetition of the results.

Comment R1.21: In summary, the methods used in this study are valid, data analysis results are scientifically sound, but presentation quality should be improved for smooth reading.
Response to R1.21: We will consider your comments in the revision of the manuscript, which will lead to a better readability of the manuscript.

Response to Referee 2

General comments: This study deals with the modeling of reactive nitrogen (N_r) deposition fluxes on a German forest, and with the calculation of annual budgets of N_r, based on a previous paper (https://doi.org/10.5194/bg-19-389-2022), where measurements of Nr concentrations are published. Different modeling approaches are used, from 1D (with DEPAC-1D) to 2D (with LOTOS- EUROS) and with a Canopy Budget Technique. Differences and similarities of results between the different approaches are highlighted. The study is useful and interesting, but is presented with too many details and lacks synthesis. The reader gets lost when all results are described. The comparison with literature results is not always useful: if a comparison is made with other results, this should help the authors to explain their own results. In this paper, the comparison with results often remains at a stage of quantification without giving any keys on how to go further to explain the processes leading to differences between studies. In other words, the discussion lacks depth.

The question of soil resistance is not addressed, especially for NH_3 deposition modeling, and could help to adjust NH_3 deposition flux overestimation. Point of attention: I have the impression that the authors would like to include results from the previous paper (Wintjen et al., 2022, Part I on measurements) and add modeling results in this paper. Be careful of correctly synthesize the right information needed for this study. I did not understand how the authors selected the figures to be put in supplementary material of in the main manuscript. The figures in supp mat are often widely described (this suggests that they are important for the study), the authors should revise either the description or the location of the figures.

Generally speaking, the manuscript needs to be synthesized, shortened, and above all deeper discussed with highlighted scientific questions. The abstract and the conclusion should be further adapted. Therefore, the manuscript needs major revisions.

Response to R2.1: We thank the Referee for his/her comments to our work. As outlined in the responses to Referee 1, we agree that the structure of the results needs to be optimized and the description of the results is too detailed, in particular Sec. 3.2 and 3.3, which also includes the description of supplementary figures. We further agree a comparison to literature is not useful in its current state. We will consider your suggestion to the discussion section. The conclusion section will be adapted since parts sound like a summary. We agree that the discussion on soil resistance should be extended. Please note the response to the corresponding comment R2.10. Comment R2.10 as well as the other specific comments to the manuscript are addressed below.

Specific comments

Abstract

Comment R2.2: L10: “total atmospheric deposition”: precise if you talk about wet and dry or only dry but for all N reactive species.

Response to R2.2: We will add (wet+dry) after total.

Comment R2.3: L36: this statement is not at the right place and should be placed at the beginning of the abstract.
Response to R2.3: As written to R1.6, the sentence will be deleted.

Introduction

Comment R2.4: L92 "Using the so- called canopy....": if you give information on the CB technique, you should also explain the inferential method using measured concentrations of gases and particles and modeled deposition velocities in comparison to throughfall measurements.
Response to R2.4: The inferential method is introduced in lines 75-79.

Comment R2.5: L95: redundant with sentence above.
Response to R2.5: We don't know why this introductory sentence should be redundant.

Comment R2.6: L100: rewrite the sentence by replacing the numbers (1), (2) and (3) by first..., then we..., and we finally...at the beginning of each statement. Add a fourth statement on the uncertainties assessment.
Response to R2.6: We will place the numbers in front of each statement and add a statement about uncertainties.

Material and methods

Comment R2.7: L110 Beudert and Breit, 2010: this reference is not recent, Anthropogenic activities and influence may have changed. Please update or confirm that the situation is still as described in 2010.
Response to R2.7: As shown in Wintjen et al. (2022), average concentrations reported by Beudert and Breit (2010) were still similar to concentrations measured during the 2.5 years campaign. Please note that this sentence will be deleted.

Comment R2.8: L115: "…are responsible for the contribution to these networks": what do you mean?
Response to R2.8: This part of the sentence is not corrected and should be "… are responsible for carrying out the ICP IM protocol at the site." Please note that this sentence will be deleted.

Comment R2.9: L122: specify the sampling resolution for all variables.
Response to R2.9: We will add the sampling resolution. Please note the response to R1.7.

Comment R2.10: L155: The soil resistance may have an important influence on NH_3 bidirectional fluxes. Is the parameterization used adapted to the soils considered in this study? Are the values realistic? Did you realize sensitivity tests?
Response to R2.10: We used the standard soil parametrization implemented in DEPAC, which is described in Van Zanten et al. (2010). In DEPAC, soil resistance is set to a constant value depending on if the soil is frozen, dry, or wet. In addition, the in-canopy resistance (as part of the effective soil resistance) is dependent on land-use, and even infinity for some classes (grass and "other"), thus effectively shutting off exchange with the soil. We did not conduct any measurements of soil conductance at the site. Thus, we cannot evaluate the representativeness of the current soil parametrization. Since a soil compensation point is not implemented in DEPAC yet, sensitivity tests are probably less useful without having any information about the site soil characteristics.

Evaluating the soil resistance parametrization within this study, is probably beyond the scope of this manuscript. Still, we noted that the discussion on soil resistance should be extended, which will be done in the revised version.

Comment R2.11: L168: is there a reference for the national emission inventory?

Comment to R2.11: We will add the following reference to line 168. For Germany, the gridded emissions were obtained from the GrETa system (*GRETA – Gridding Emission* Tool for ArcGIS v1.1; Schneider et al., 2016).

Comment R2.12: Paragraph 2.2.3: a table summarizing what model provides what output, and with what inputs are calculated deposition fluxes, would be useful
Response to R2.12: Please note the response to R1.7.

Comment R2.13: L191: "concentration measurements on monthly and half hourly…": for which species? Based on available measurements?
Response to R2.13: HNO_3, SO_2, NH_3, and particulate NO_3^- and NH_4^+ were available at monthly resolution, NO, NO_2, and NH_3 at half-hourly resolution. All concentration based on available measurements. This information is given in the following lines and will be provided in Sec. 2.1 (see R1.7).

Comment R2.14: L197: what do you mean by instationarities? This explanation on gap filling is not clear. Please try to be more concise.
Response to R2.14: We will replace instationarities by large variabilities.

Comment R2.15: L221: avoid the use of etc…, not precise enough.
Response to R2.15: We will delete etc. and at other occurrences in the manuscript.

Comment R2.16: I do not understand how this 2.2.3 paragraph is organized. It should be about site base modeling with DEPAC-1D, and this paragraph from L215 to L230 deals with LOTOS-EUROS model and TRANC measurements. A bit of storage is needed.
Response to R2.16: We agree. The last paragraph explain how flux estimates of the different methods are compared with each other and how gaps in DEPAC-1D and TRANC measurements are filled. Please note that lines 217 to 230 will be integrated into Sec. 2.1.

Comment R2.17: L239 to 246: in this paragraph, you should specify where you talk about dry, wet deposition and what you mean when you talk about total deposition: total atmospheric (wet+dry) or total species?
Response to R2.17: We agree and will adapt lines 239 to 244 as follows:

The canopy budget technique (CBT) is the most common method for estimating total atmospheric deposition of dissolved inorganic nitrogen (DIN_t) based on wet inorganic nitrogen fluxes in open site precipitation (bulk deposition) and throughfall (NO_3^-, NH_4^+-ions) only (see Staelens et al., 2008, Table 1). DIN_t was estimated on yearly basis after the CBT approach of Draaijers and Erisman (1995) and de Vries et al. (2003) whose results differed only marginally and were therefore averaged. The biological conversion of deposited inorganic nitrogen into dissolved organic nitrogen (DON) in the phyllosphere (bacteria, yeasts, and fungi) or the dry deposition of atmospheric DON onto the canopy or the exudation of DON from plant tissues is not addressed in CBT. Here, the difference of DON fluxes between throughfall and bulk deposition (ΔDON) was used as an estimate of N_r dry deposition not accounted for by CBT.

Comment R2.18: L243: what do you include in "deposite inorganic nitrogen into dissolved organic nitrogen?
Response to R2.18: soluble amino compounds

Comment R2.19: L246: Does total nitrogen deposition Nr include organic and inorganic species?
Response to 2.19: Yes.

Results

Comment R.2.20: Add "particulate" in front of NO_3^- and NH_4^+ if this is about dry deposition to avoid confusion with ions in DIN.
Response to R2.20: We will add a "p" in front of NO_3^- and NH_4^+.

Comment R.2.21: L295: "However…" combine this sentence with the one above.
Response to R.2.21: Second sentence will be replaced by "… were different to results from measurement, which stated NO_x as predominant compound in the ΣN_r concentrations (Wintjen et al., 2022).

Comment R.2.22: L305 to 330: too much description. Go directly to what is important and speaks above all of what poses a question.
Response to R2.22: As written before, we will shorten the description of the figures and will provide the essential messages of the figures a few lines. We agree that a detailed description of the individual fluxes and deposition velocities is not needed here if not essential for further interpretation of the results. The sentences given below will be major lines for the description of Figures S4 and S5.

NH_3 deposition velocities of LOTOS-EUROS and DEPAC-1D exhibited similar values in winter, but disagreements were found in summer and autumn. In summer, DEPAC-1D determined systematically larger median deposition velocities, whereas LOTOS-EUROS predicted a large variability in NH_3 deposition velocities during autumn, which was not supported by DEPAC-1D. NO_2 deposition velocities agreed well in their temporal pattern and the median deposition velocities. Still, variability in DPEAC-1D deposition velocities was slightly higher apart from the winter months. In both model applications, NO deposition velocities were practically zero. For particulate NO_3^- and NH_4^+, deposition velocities DEPAC-1D and LOTOS-EUROS agreed well with median deposition velocities close to zero during summer, but large disagreement were found during winter. The same phenomenon was found for HNO_3.

For the fluxes, similar observations can be made. Differences to the observations made for v_d are related to differences in the concentration input. Since the description of Figures S4 and S5 will be shortened substantially, there is no need to place the figures in the manuscript

Comment R2.23: Your first paragraph in the results section deals with concentrations. Rather than separating the analyses model by model, why don't you write another paragraph on deposition velocities, and another paragraph on deposition fluxes? The comparison between each approach would be easier.
Response to R2.23: Please not the response to R1.18. In the revised version, Sec. 3.2 will be related to deposition velocities and Sec. 3.3 to flux measurements.

Comment R.2.24: L354: give the principle of the scheme in one sentence.
Response to R2.24: We will replace the sentence In addition, we separated dry and wet leaf surfaces following the calculation scheme by Wintjen et al. (2022) by the following sentences:

Leaf surface wetness was measured at the site with sensors attached to a spruce and a beech tree. In order to classify the sensor as dry or wet, the half-hourly leaf wetness value was compared to a threshold value based on the calculation scheme given by Wintjen et al. (2022).

Comment R.2.25: L372: if the authors want to talk about errors in the stability parameterization, they should be more precise, because it is not possible to understand which process is involved, and why it influences the result. If it is crucial, it should be corrected for this study and not for after the study.
Response to R.2.25: We agree that the explanation is currently imprecise. As shown in Figure S4, large deposition velocities for particulate NO_3^-, NH_4^+, and HNO_3 were observed in the LOTOS-EUROS

simulations during winter. Deposition of these compounds is mostly driven by the aerodynamic resistance and quasi-laminar resistance, R_a and R_b. Surface roughness is probably lower compared to the summer month due to snow-covered surfaces and smaller leaf area indexes. This affects the friction velocity (u_*) calculation leading to potentially higher values, which lowers these resistances resulting in large v_d values. The Monin-Obukhov-length (L) determines the integrated stability functions and depends on wind speed close to the surface, cloud cover, and solar zenith angle (Manders-Groot et al., 2016). Snow cover is not considered in the parametrization of L yet. Including L in the parametrization change the albedo of the surface and thus the prevailing stratification of the boundary layer probably leading to more occurrences of stable stratification. An implementation of snow cover in the parametrization of L may reduce the deviations of simulated stability and friction velocity to measured counterparts. We will rephrase line 370ff according to this response.

Comment R.2.26: Generally speaking, this paragraph page 12 is too long. The reader gets lost. Please synthesize, and find a way to classify.
Response to R2.26: Please note the response to R2.22.

Comment R.2.27: L495: An example of sentence not useful which could be removed to clarify the important text: "Until June 2018, measured deposition was higher than the half of the previous years". Same comment for the sentence after this one.
Response to R.2.27: The sentences Until June 2018, measured deposition was higher than the half of the previous years. DEPAC-1D deposition was nearly identical for 2016 and 2017, but lower than measured deposition until June 2018 will be deleted.

Comment R.2.28: Line 500 to 510: please synthesize and shorten this paragraph
Response to R2.28: We will delete lines 501 to 505.

Comment R.2.29: The comparison with literature data should be based on a discussion on processes. If the comparison is only quantitative, it is not a discussion. The first paper by Wintjen et al. (2022) has shown a predominant role of NO_x, contrary to model results. This particular result could be a major axis of the discussion, by analyzing which processes and which parameterizations are responsible for such a result. The comparison with literature should give ideas for the interpretation of your data and not only give possible range of values.

Once again, axis of discussion should be highlighted as scientific questions, and illustrated by results, rather than willing to illustrate all concentrations, than all fluxes, because we get lost with too many quantitative details.

Response to R2.29: We agree that the current discussion, in particular Sec. 4.1 and partly Sec. 4.2, is quite quantitative and the discussion on possible mechanisms causing differences between models and measurements is missing or can be extended in the case of critical loads for example. Quantitative comparison to literature will be removed.

In the revised version, the differences in the contribution of N_r species to ΣN_r will be one aspect of Sec. 4.1. Second will be the difference in fluxes between LOTOS-EUROS, DEPAC-1D, and TRANC with focusing on differences in the ΣN_r exchange. The results from individual compounds will be used to interpret these differences. Responsible processes in modeling these compounds will be discussed. The discussion on critical loads will be extended (see R2.34).

Comment R.2.30: L552: How can you prove the application of fertilizers? What do you exactly mean by "these times"? Any indication about the practices? It is interesting to discuss this question about fertilizer application, but this discussion should not be flooded among the other statements.
Response to R2.30: We agree that the phrase "these times" is not precise. Here, we refer to spring and autumn. Generally, the sentence may indicate that fertilization techniques and type of fertilizer

were known, which is not the case. Since the site was several kilometers away from possible nitrogen emission sources, the sentence should more general. Instead of in particular…, we will write like emission from the application of fertilizer or animal householding.
As written in Wintjen et al. (2022), sparse livestock farming is done in the surroundings of the tower. Disagreements in NH_3 concentration lead to the conclusion that modeled emissions of NH_3 from agriculture are overestimated substantially for this grid cell.

Comment R2.31: L566: this is theory, not discussion.

Comment R2.32: L580 to 600: same remark as above: the comparison with literature remains too quantitative and this is not a discussion.

Response to R2.31 and 2.32: These lines will be rephrased and used for the interpretation of the results if possible.

Comment R.2.33: L611: are the conditions in Trebs et al. (2005) comparable to your study to allow you give this interpretation?
Response to R2.33: The sentence will be deleted since we know that NH_3 concentrations are overestimation in the LOTOS-EUROS-simulations. The chosen reference is not suitable for this sentence, since conditions compared to Trebs et al. (2005) were different. Since both, DELTA and LOTOS-EUROS, found an excess of NH_4^+ over NO_3^-, particulate NH_4^+ was probably most responsible for deposition of ΣN_r.

Comment R2.34: L616 to 622: this sounds like a partial conclusion.
Response to R2.34: We agree and will move them to the conclusion.

Comment R2.35: L651 to 661: this paragraph on critical load is very small despite being an interesting point of discussion. Should be expanded, while previous paragraphs should be reduced.
Response to 2.35: We will delete the sentence Thus, the investigated forest ecosystem is in a potentially endangered state. and add the following information after line 656:

Physical examinations of tree species showed that the critical load concept, which showed that the forest stand is still below critical limits, is valuable tool to evaluate the functionality of an ecosystem. Long-term observations of effects in different environmental media indicating nitrogen loads, however, revealed normal ecosystem functioning: healthy tree stands with medium nitrogen nutrition and balanced ratios of nitrogen to other nutrients in tree foliage and normal growth of (Beudert and Breit, 2014) and intact nitrogen retention and storage results in very low nitrate concentrations in soil water, aquifers and streams (Jung et al., 2021). Moreover, green algae coatings on spruce needles indicating higher NH_y dry deposition (Grandin, 2011) could not be found even at exposed high elevation sites.

Comment R2.36: L664: the paragraph is entitled uncertainties in DEPAC 1D, but deals with all model results. Change the title and adapt the content to your own results. Another solution could be to include uncertainty considerations in each question, influence of meteorological parameters, influence of soil resistance parameterizations on NH3 deposition velocity, etc…(these are examples, not mandatory to follow).
Response to R2.36: We thank the Referee for his/her suggestions to this section. We will change the title and discuss the potential uncertainty questions regarding their impact on the fluxes.

Comment R2.37: L687: “no distinct…”: what do you mean?
Response to R2.37: We will replace the sentence starting with no distinct with …since no well-defined time lag of NH_3 was found in the EC flux analysis.

Comment R2.38: L699: you mention hydrochloric acid. Do you have any idea of the concentrations in your study? Is it relevant to mention it?
Response to R2.38: From DELTA measurements, average hydrochloric acid (HCl) concentration was ca. 0.1 µg m^{-3}. Probably, is not relevant for the discussion of HNO_3 emission.

Comment R2.39: L701 to 707: this is not a discussion topic.
Response to R2.39: We agree and will remove these lines.

Comment R2.40: L751: 761: OK for the discussion on the combination of long-term measurements and intensive campaigns but why is it mentioned in the paragraph about uncertainties in DEPAC-1D?
Response to 2.40: The combination of long-term measurements and high-resolution measurements are useful to reduce uncertainties in inferential model applications. Since this partly a conclusion of this study, the lines will be moved to the conclusion section.

Comment R2.41: L771: considerations about NH_3 emission inventory should be merged with discussion on NH_3 above. What is the range of these emissions?
Response to R2.41: We agree that the information about the NH3 emission can be moved to line 532. If the information on the range of these emission is needed after merging, we will provide a range of possible values.

Comment R2.42: L815: "overestimation was only partly related to other issues like grid cell size", but you have written above that the grid cell size was not a problem. Please rephrase.
Response to R2.42: Agreed. We will delete and overestimation was only partly related to other issues, for example, the grid cell size of 7x7 km^2

Summary and conclusion

Comment R2.43: In my opinion, should be conclusion only. Summary is abstract.
Response to R2.43: We agree and will rephrase the conclusion section.

Comment R2.44: L837: DEPAC-1D and DEPAC-1D only: is there a mistake?
Response to R2.44: No. For both gap-filling approaches DEPAC-1D was used, but in one approach DEPAC-1D was applied to gaps, which could not be filled with MDV.

Comment R2.45: L845: erroneous parameterizations? If this is true then the modeling results should not be published and the parameterizations should be corrected before. Once again, the discussion should focus on some important processes and how they are represented in the model, which could explain discrepancies with observations.
Response to R2.45: We apologize for this inappropriate phrase. We will replace erroneous by uncertainties in the.

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