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 valuable comments to the manuscript. We agree that the structure of the manuscript needed improvements. The discussion on fluxes was rather quantitative, and descriptions of the results were too detailed making it difficult for the reader to understand the main findings. Based on the reviewers' suggestions, we reduced the level of detail and improved the readability of the discussion.

Comments of Referee 1 range from R1.1 to R1.22, comments of Referee 2 range from R2.1 to R2.45. Line numbers in the answers, where new information was added to the manuscript, refer to the revised version if not otherwise explicitly mentioned. Text marked in red was deleted, text marked in blue was 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 original submitted version needed improvements 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, longer gaps. In a second approach, only DEPAC-1D was used for gap-filling. We replaced 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 coclusion that the parametrizations may need revision is not needed here and was 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: We removed the sentence LOTOS-EUROS predicted an averaged ΣN_r concentration of 5.0±3.3 µg N m^{-3}. and rephrased the corresponding lines as follows:

LOTOS-EUROS showed substantial discrepancies to measured ΣN_r deposition during spring and autumn, which was related to an overestimation of ammonia (NH_3) concentrations by a factor of two to three compared to measured values as a consequence of a mismatch between gridded input NH_3

emissions and the site's actual, rather low, pollution climate. According to LOTOS-EUROS predictions, ammonia contributed most to modeled input ΣN_r concentrations, whereas measurements showed NO_x as the prevailing compound in ΣN_r concentrations.

Please note lines 28-31.

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 streamline the entire Sect. 2.1 and entitled it "Dataset description". In this section, we provided a table giving an overview about each method with input and output parameters. Information on sample resolution and data coverage were added. We replaced Sect. 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 of the original submitted version) was moved to lines 133-146.

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

Method	**Primary input/observation 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	ΣN_r fluxes at half-hourly resolution, no gap-filling applied
DEPAC-1D	Measurements of micrometeorological variables at half-hourly resolution	Fluxes of NH_3, NO_2, NO, HNO_3, pNH_4^+, and pNO_3^- at continuous half-hourly resolution
	Measured NH_3, NO, NO_2 concentrations at half-hourly resolution	
	Measured SO_2, HNO_3, NH_3, pNO_3^-, and pNH_4^+ concentrations at monthly resolution	
TRANC (DEPAC-1D)	See above	Continuous ΣN_r fluxes at half-hourly resolution, only DEPAC-1D is used for gap-filling
TRANC (MDV+DEPAC-1D)	See above	Continuous ΣN_r fluxes at half-hourly resolution, gap-filled with a combination of MDV (window size of ±5 days) and DEPAC-1D for adding further missing fluxes
LOTOS-EUROS	Meteorological data from ECMWF weather forecasts and modeled concentrations of SO_2,	Continuous fluxes of NH_3, NO_2, NO, HNO_3, pNH_4^+, and pNO_3^- at hourly resolution; fluxes were

	NH_3, NO_2, NO, HNO_3, pNH_4^+, and pNO_3^- at hourly resolution for 7x7 km^2 grid cell; concentrations were linearly resampled to half-hourly resolution	linearly resampled to half-hourly resolution
Canopy budget technique	Throughfall measurements from nearby spruce and beech trees and bulk deposition measurements at an open-site in weekly intervals	Dissolved inorganic nitrogen deposition (DIN) based on the exchange of NO_3^- and NH_4^+ ions on monthly basis following the approaches of Draaijers and Erisman (1995) and de Vries et al. (2003), dissolved organic nitrogen (DON) corresponds to difference of DON fluxes between throughfall and bulk deposition

For the comparison to modeled ΣN_r deposition fluxes, TRANC EC flux measurements described in detail in Wintjen et al. (2022) were used. These flux measurements were available at half-hourly resolution, carried out 30 m above the forest floor, 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 (L), friction velocity (u_*)) and air pollutant concentrations (NO, NO_2, HNO_3, NH_3, pNO_3^-, pNH_4^+, and sulphur dioxide (SO_2)) are required for flux calculations. NH_3 concentrations obtained from Quantum cascade laser measurements taken at 30 m above ground, NO_2 and NO obtained from chemiluminescence measurements taken at 50 m above ground as well as micrometeorological parameters were aggregated at half-hourly resolution, whereas the remaining N_r species and an additional NH_3 determination were obtained from DELTA (DEnuder for Long-Term Atmospheric sampling, e.g., Sutton et al., 2001; Tang et al., 2009) and passive sampler (NH_3 only) measurements of the IVL type (Ferm, 1991) on monthly basis. DELTA measurements were made at 30 m and passive sampler measurements at 10, 20, 30, 40, and 50 m above ground. Temperature and relative humidity were collected in a profile at 10, 20, 40, and 50 m above ground. Pressure and global radiation measurements were taken at 50 m. Indicators of stability and turbulence such as L and u_* were obtained from momentum flux measurements of the sonic anemometer.

Gaps in DEPAC-1D were mostly related to gaps in micrometeorological input 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, pNH_4^+, and pNO_3^- and 9.3% for NO and NO_2) were filled with results from LOTOS-EUROS. A detailed description of the site and the instrumentation is given in Wintjen et al. (2022). For LOTOS-EUROS flux modeling, modeled input data of the European Centre for Medium range Weather Forecast (ECMWF) and the national emission inventory of Germany (Schneider et al., 2016) were used to predict deposition fluxes for NO, NO_2, HNO_3, NH_3, pNO_3^-, and pNH_4^+. LOTOS-EUROS fluxes were resampled to half-hourly timestamps from the original hourly resolution and missing fluxes were linearly interpolated. For the canopy budget technique, throughfall measurements under spruce and beech trees close to the station (Beudert et al., 2014) and bulk deposition measurements at an open site (Wintjen et al., 2022) were taken in weekly intervals and used for determination of total nitrogen dry deposition on annual basis (Sect. 2.3). 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 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 TRANC 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-hourly values was given as

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

where $\tilde{X}$ represents the median of the ratio of the uncertainty of the measured fluxes ($F_{\mathrm{unc,meas}}$) to their corresponding flux values (F_{meas}). The uncertainty of the measured fluxes was estimated after 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 point is only implemented for NH_3. This sentence was added to line 153.

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 added 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. 20 m. By assuming a constant flux and using the stability parameters, the concentrations can be estimated for the canopy top and the typical observation height (2.5 m above roughness length (z_0)) in air quality networks. We added the highlighted information to line 188.

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: We rephrased the corresponding line as follows: On hourly basis, the land-use specific total dry deposition was calculated in LOTOS-EUROS by applying DEPAC for NH_3, NO, NO_2, and HNO_3. Dry deposition of pNO_3^- and pNH_4^+ was calculated according to Zhang et al. (2001) (see Manders-Groot et al. (2016, Sect. 5.2)). Please note lines 185-188.

PAN is of course in LOTOS-EUROS included but is not considered in DEPAC. 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 diameter of 0.7 µm was used. For the coarse fraction of NO_3^-, 8 µm was taken as diameter (Manders-Groot et al. (2016, Sect. 5.2)). We added this sentence to line 206.

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 of the original submitted version were shortened and moved to Sect. 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 for in the wet deposition measurements (l243f of the original submitted version). The CBT approach focuses on inorganic N species. The „contribution of the dry deposition of organic N in the total N_r dry deposition“ is not directly measured by CBT. Dry deposition of ΔDON could have been occurred, but it may be also caused by microbial conversion of inorganic N_r or by leaching from plant tissues. We are not able to draw a firm conclusion about the contribution of 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 Sect. 3.1 makes the reading difficult since the level of detail in the results section was rather high and thus it was challenging to take conclusions from Sect. 3.1. We appreciate your suggestions to the structure of Sect. 3.1 and changed the way of presenting facts according to your suggestions.

First, we made a comparison N_r species, which were available at half-hourly resolution taking measured concentrations as the basis (Sect. 3.1.1). Therefore, we revised Fig. 1, which shows now a comparison of NH_3, NO_X, and ΣN_r only. Due to the new figure, we deleted lines 250-259 of the original submitted version. Afterwards, we made the comparison of passive samplers and DELTA with LOTOS-EUROS (Sect. 3.1.2)

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: Please note the response to R1.16. For the other figures, we adjusted the y-axis limits of corresponding figures like 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 both Referees to change the structure of the sections making a section for comparisons on v_d and fluxes.
In Section 3.2, only deposition velocities are described starting with the N_r compounds of DEPAC-1D and LOTOS-EUROS (Sec 3.2.1). Therefore, we removed lines 304-327 of the original submitted version and shorten the description of Fig. S4 as follows:

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. For NO_2, deposition velocities of LOTOS-EUROS and DEPAC-1D agreed well in their temporal pattern and the median deposition velocities, but the variability in DEPAC-1D deposition velocities was slightly higher during summer. In both model applications, NO deposition velocities were practically zero (medians always < 0.06 cm s^{-1}). For pNH_4^+, deposition velocities of DEPAC-1D and LOTOS-EUROS agreed well with median deposition velocities close to zero, but a large disagreement was found during winter. Deposition velocities of pNO_3^- were close to zero during the entire campaign in DEPAC-1D, but LOTOS-EUROS showed a large scattering of v_d in the winter months. For HNO_3, a discrepancy in v_d was also found during winter, and, similar to NH_3, deposition velocities of DEPAC-1D were generally larger from May to September. The comparison of the deposition velocities for each N_r compound modeled by DEPAC-1D and LOTOS-EUROS is shown in Fig. S4.
Please note lines 304 to 315 of the revised version.

Afterwards, the comparison of modeled and measured ΣN_r deposition velocities followed (Sect. 3.2.2). We adjusted the y-limits of Fig. 3 changing them to -1 cm s^{-1} to 2 cm s^{-1} and removed lines, which are not related to deposition velocities in general or redundant, e.g., lines 333, 342-346 of the original submitted version. Furthermore, a panel showing diurnal patterns of ΣN_r deposition velocities of LOTOS-EUROS stratified by the same parameters as DEPAC-1D and TRANC measurements was added to Fig. 4. The following lines were added to line 357:

In case of LOTOS-EUROS, separating diurnal cycles of v_d led to similar observations made for DEPAC-1D regarding relative humidity and leaf surfaces. In addition, lower temperatures and concentration tend to increase v_d, which contradicts the results of DEPAC-1D. Generally, values of v_d are closer to TRANC deposition velocities, but the diurnal pattern differs from TRANC and DEPAC-1D showing maxima in the morning (~06:00 LT) and evening (~18:00 LT) and low values around noon except for high relative humidity and wet leaf surfaces.

In Section 3.3, the half-hourly fluxes of the N_r compounds and ΣN_r are described starting with the modeled fluxes of the N_r compounds (Sect. 3.3.1). We removed lines 368-405 of the original submitted version and replaced them by the following lines:

The statements made for v_d can be transferred to the flux predictions. Differences to the observations made for v_d (Fig. S4) are related to the concentration input data. For example, due to overestimations of modeled NH_3 concentrations in spring and autumn, differences in fluxes were higher during the same time. Modeled NO_2 and HNO_3 concentrations of LOTOS-EUROS were lower than their measured values resulting in flux underestimations by LOTOS-EUROS for NO_2 and HNO_3 during summer. High modeled input concentrations of particulate nitrogen led to substantial deposition fluxes in the LOTOS-EUROS simulations. Following the model predictions, NH_3 fluxes had the largest contribution to the modeled ΣN_r flux with an average flux of -12.5 and -13.0 ng N m^{-2} s^{-1} in the DEPAC-1D and LOTOS-EUROS applications, respectively, considering the entire campaign. Averaged fluxes of NO_2 and HNO_3 showed – although on a low level in absolute terms – higher deposition fluxes for DEPAC-1D, namely 2.0 and 1.3 ng N m^{-2} s^{-1}, respectively, compared to 1.2 and 0.3 ng N m^{-2} s^{-1} in case of LOTOS-EUROS. Substantial flux differences were found for particulate nitrogen. DEPAC-1D averaged fluxes were close to zero (0.9 and 0.1 ng N m^{-2} s^{-1} for pNH_4^+ and pNO_3^-, respectively), whereas LOTOS-EUROS showed substantial higher aerosol deposition with averaged fluxes of 3.7 and 2.2 ng N m^{-2} s^{-1} for pNH_4^+ and pNO_3^-, respectively. The comparison of fluxes for each N_r compound of LOTOS-EUROS and DEPAC-1D is shown in Figure S5. Please note lines 365 to 377 of the revised version.

Section 3.4 was changed to Section 3.3.2.

The section describing the cumulative N exchange and annual budgets was also modified. Repetitions from the previous sections and information about the compensation point of NH_3 were deleted (lines 464-483 of the original submitted version). The description of the annual dry nitrogen deposition was also shortened (lines 494-496 and 508-508 of the original submitted version)

Comment R1.19: 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 R1.19: As written in the responses above, changes to the results section were made including a reduction in details and a presentation of the most important messages. Please also note the responses from R2.22 to R2.28.

Comment R1.20: 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 R1.20: 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 is necessary to highlight these uncertainties making readers, developers, and potential users aware of them. Please note the response to R2.36.

Comment R1.21: Lines 819-824: No need to repeat what you have done in the conclusion section.
Response to R1.21: We deleted these lines and additionally rephrased parts of the conclusion, which appeared like a repetition of the results (lines 825-844 of the original submitted version).

Comment R1.22: 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 R1.22: We thank again for the suggestions and considered your comments in the revision of the manuscript, which hopefully led to a better readability of the text.

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 original submitted version needed improvements and the description of the results was too detailed, in particular Sect. 3.2 and 3.3, which also includes the description of supplementary figures. We shortened the description of the supplemental figures and kept the main messages of those figures in the text. We further agree that the comparison to literature was not always useful in the original version. Large parts of the discussion, in particular of Sect. 4.1 were too quantitative. We revised the discussion by focusing on the mechanisms, which were likely responsible for differences in model-measurement data. References were used to support the explanations. We considered your suggestions to the discussion section. The conclusion section was adjusted since parts did sound like a summary. We agree that the discussion on soil resistance should be extended. Your 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 added (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 was 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 72-75.

Comment R2.5: L95: redundant with sentence above.
Response to R2.5: We are very sure that the information has not been given before. No changes were made.

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 placed the numbers in front of each statement and added 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 was 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 as we decided to rephrase the whole section (see comments above). It should have been "… are responsible for carrying out the ICP IM protocol at the site."

Comment R2.9: L122: specify the sampling resolution for all variables.
Response to R2.9: We added the sampling resolution. Please note the response to R1.7 and the newly designed Table 1.

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: In DEPAC, soil resistance is set to a constant value depending on whether the soil is frozen, dry, or wet. In addition, the in-canopy resistance (as part of the effective soil resistance) is dependent on the inverse of u_*, surface area index (LAI+ area index of stems and branches (van Zanten et al., 2010)) and may lower the 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 beyond the scope of this manuscript. Still, we noted that the discussion on soil resistance should be extended, which was done in the revised version. Please note the rephrased discussion on modeling uncertainties (Sect. 4.2). We included a subsection dealing with the soil compensation point and soil resistance.

Comment R2.11: L168: is there a reference for the national emission inventory?
Comment to R2.11: We added the following reference to line 184. 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 aggregated at half-hourly resolution. All concentrations are based on available measurements. This information was given in lines 191-203 of the original submitted version and is now provided in Sect. 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 replaced instationarities by periods of low auto-correlation.

Comment R2.15: L221: avoid the use of etc…, not precise enough.
Response to R2.15: Please note that lines 221f of the original submitted version were deleted. In the revised version, all occurrences of etc. were deleted.

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 explains 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 of the original submitted version were integrated into Sect. 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 replaced total and dry nitrogen deposition in ecological field research based on inorganic nitrogen fluxes (NO_3^- , NH_4^+) only by total atmospheric deposition of dissolved inorganic nitrogen (DIN_t) based on wet inorganic nitrogen fluxes of NO_3^- and NH_4^+-ions estimated from open-site precipitation (bulk deposition) and throughfall of NO_3^- and NH_4^+-ions measurements […].

Comment R2.18: L243: what do you include in "deposite inorganic nitrogen into dissolved organic nitrogen?
Response to R2.18: The following addition was made to that sentence:

The biological conversion of deposited inorganic nitrogen into dissolved organic nitrogen (DON) in the canopy phyllosphere (bacteria, yeasts, and fungi) or the dry deposition of atmospheric DON onto the canopy or the exudation of DON from plant tissues […].

Comment R2.19: L246: Does total nitrogen deposition Nr include organic and inorganic species?
Response to 2.19: Yes, by adding ΔDON to DIN from throughfall measurements or to DIN_t inorganic and organic nitrogen are included in dry deposition estimates.

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 added 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: Sentences were combined accordingly.

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 shortened the description of the figures and provided the essential messages of the figures in a few lines. We agree that a detailed description of the individual fluxes and deposition velocities is not needed if it is not essential for further interpretation of the results.

Since the description of Figures S4 and S5 was shortened substantially, there was no need to place the figures in the manuscript. Please note the response to R1.18.

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 note the response to R1.18. While discussing the structure of the manuscript for the initial submission, we thought the separation by method would have made the manuscript more accessible. Methods, measurements, and model applications are relatively new and therefore a step-wise description may be needed. Thus, figure descriptions came with a high detail.

However, we agree that a structure referring to deposition velocities first and fluxes second makes the manuscript more accessible. In the revised version, Sect. 3.2 is about deposition velocities and Sect. 3.3 about flux measurements.

Comment R.2.24: L354: give the principle of the scheme in one sentence.
Response to R2.24: We replaced 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 was imprecise. We deleted lines 370ff of the original submitted version and replaced lines 803-807 of the original submitted version by the information given in this response:
The large contribution of aerosols to the total deposition (Fig. 7) modeled by LOTOS-EUROS was accompanied by unusually high deposition velocities of pNH_4^+, pNO_3^-, and HNO_3 from November 2017 to February 2018. Deposition of HNO_3 and particulate nitrogen is mostly driven by the aerodynamic resistance and quasi-laminar boundary resistance, R_a and R_b. Since v_d of those compounds was relatively high compared to measurements during that time, R_a and R_b were probably low or even close to zero. R_a and R_b depend on various parameters like u_*, the integrated stability corrections functions after Webb (1970) and Paulson (1970), surface roughness, and leaf area index. 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 snow cover in the parametrization affect the albedo of the surface and thus the prevailing stratification of the boundary layer, which probably leads to more occurrences of stable stratification. An implementation of snow cover in the parametrization of L may reduce the deviations of simulated vs. measured stability and u_*.

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 responses to R2.22 and R1.18.

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 were deleted.

Comment R.2.28: Line 500 to 510: please synthesize and shorten this paragraph

Response to R2.28: We deleted lines 500 to 507 of the original submitted version and added the following lines:

The difference to TRANC estimates until June 2018 was caused by the deposition fluxes in February 2018, which had an influence on the MDV method leading to significantly larger gap-filled fluxes. Hence, DEPAC-1D estimate was lowest among all methods for the first half of 2018. In 2016 and 2017, deposition estimates of DEPAC-1D were nearly identical due to similarities in micrometeorological and concentration input values. As expected from Fig. 7, annual LOTOS-EUROS estimates were highest in comparison to DEPAC-1D and TRANC. All deposition estimates were within the range of long-term lower and upper estimates of the CBT approach estimated from 2010 to 2018, with TRANC measurements close to the lower average and LOTOS-EUROS predictions to the higher average.

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 discussion of the submitted version, in particular Sect. 4.1 and partly Sect. 4.2, was too quantitative and the discussion on possible mechanisms causing differences between models and measurements was missing and can further be extended with regard to critical loads for example. The quantitative comparison to literature was removed, which included lines 525-600 of the original submitted version.

In the revised version, the differences in the contribution of N_r species to ΣN_r is part of Sect. 4.1. Therefore, we moved lines 770-779 of the original submitted version to Sect. 4.1 and discussed possible reasons which were responsible for the disagreements in the contribution of individual N_r species to ΣN_r concentrations between LOTOS-EUROS and measurements.

The second part deals with differences between modeled and measured fluxes. We discussed processes, which were likely responsible for the observed model-measurement differences. The discussion on critical loads was also extended (see R2.35). Appropriate literature references were kept if they were useful for the interpretation of the results.

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. The fertilizer application technique and type of fertilizer is not relevant for the discussion here, since the site was several kilometers away from nitrogen emission sources. From our comparison of measured and modeled NH_3 concentrations, we concluded that in the emission inventory of LOTOS-EUROS emissions of NH_3 are spatially not well allocated for the grid cell.

In the revised version, we removed this aspect from the discussion.

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 were deleted.

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 was deleted since we demonstrated with our measurement data that NH_3 concentrations were overestimated in the LOTOS-EUROS simulations. The chosen reference was 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. Please note lines 557 to 559.

Comment R2.34: L616 to 622: this sounds like a partial conclusion.
Response to R2.34: We agree and moved these lines 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 deleted the sentence Thus, the investigated forest ecosystem is in a potentially endangered state. and added the following information after line 606:

The state of tree physiological parameters suggested that the critical load concept, which indicated that the exposure of the forest to N deposition is still below critical limits, is a valuable tool to evaluate the functionality of an ecosystem. Long-term observations of nitrogen input to this ecosystem showed nitrogen concentrations in trees and water reservoirs, but ecosystem functionality was not impaired. According to leaf examinations done by Beudert and Breit (2014) at the site, balanced ratios of nitrogen to other nutrient concentrations in tree foliage were found, and usual tree growths were reported. Jung et al. (2021) found low nitrate concentrations in soil water, aquifers, and streams at the site showing an intact nitrogen retention and storage system. Moreover, green algae coatings on spruce needles usually indicating higher NH_x dry deposition (Grandin, 2011) were not found at the site.

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 NH_3 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 changed the title and discussed the potential uncertainty questions regarding their impact on the fluxes. First, we give an uncertainty discussion on DEPAC with regard to the influence of micrometeorology and the influence of soil resistance, soil compensation point, issues in the cuticular compensation point of NH_3, and the possible influence of emission of N_r species like NO_2 and HNO_3 on ΣN_r (Sect. 4.2).

Section 4.2.1 deals with uncertainties in DEPAC-1D (lines 715-750 of the original submitted version) and Sect. 4.2.2 (formerly Section 4.3) with uncertainties related to LOTOS-EUROS.

Comment R2.37: L687: "no distinct…": what do you mean?
Response to R2.37: No well-defined time lag of NH_3 was found. Thus, flux calculation using the EC method was not possible for NH_3. Please note that the sentence was deleted.

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 this is not relevant for the discussion of HNO_3 emission. We removed it from line 684.

Comment R2.39: L701 to 707: this is not a discussion topic.
Response to R2.39: We removed 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 is later mentioned in the conclusions, lines 752-758 of the original submitted version were deleted and lines 759-760 of the original submitted version were moved to the conclusions.

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 moved the information about the NH_3 emission to Sect. 4.1. Since the information on the range of these emission was not needed after merging, possible values were not added.

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: We deleted and overestimation was only partly related to other issues, for example, the grid cell size of 7x7 km^2. because the effect of the reduction in grid cell size was discussed before and did not reduce the dry deposition estimates of LOTOS-EUROS.

Summary and conclusion
Comment R2.43: In my opinion, should be conclusion only. Summary is abstract.
Response to R2.43: We agree and removed the term summary. As outlined in R1.21, we rephrased the conclusion as parts of the original submitted conclusion sounded like a summary.

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 only applied to gaps, which could not be filled with MDV. Please note that line 837 of the original submitted version was deleted.

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 replaced erroneous by uncertainties.

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