

Title: Using “remote region (area)” instead of “low polluted area” would sound much better to my opinion.

Line 5: ...at a mixed forest exposed to low air pollution levels.

Line 15: ... high solar radiation...

Line 19: No significant influence of temperature, humidity, friction velocity, or wind speed on ΣNr dry deposition sums were found.

➔ This is somewhat in contrast to what was mentioned in other places of the paper. These variables determine the deposition velocity and, hence, at the end also the total deposition. Maybe reformulate or delete.

Line 21: ...half-hourly value...

Line 25: ... to a remote forest ecosystem.

Line 37: nitric oxide (NO)

Line 39: ...aerodynamic gradient method (AGM), please change throughout

Lines 79-80: These few long-term micrometeorological measurements of Nr species above forests were made more than 20 years ago and no recent reports on long-term flux measurements of Nr are currently available.

Line 83-84: As stated above, the outstanding benefit of TRUNC is.....

Line 85-86: I would combine this sentence with the scientific objectives mentioned in lines 100-104: please reformulate / list points (1), (2), (3) as scientific objectives.

Lines 105-108: Please delete these lines as they are not required here and can be misleading. I would just mention one sentence that a follow up paper will deal with...

Line 196: Additionally, fast-response measurements.... (delete..., too).

Line 251: ... and associated descriptions are based on...

Line 293: ... replace “or nitrogen aerosols” with “or related aerosol compounds”...

Lines 305-306: Please delete: “Further details about the implementation of these resistances in surface-atmosphere models can be found in van Zanten et al. (2010).”

Line 312: replace “A breakdown...” by “The contribution of individual nitrogen compounds to the total ΣNr concentration pattern is shown in Fig. 2, which.....”

Line 315: NO_x also showed a... (delete “too”)

Line 318: The ΣNr concentration was 3.1....

Line 321: ...in the annual pattern was reasonable...

Line 322: ...with measurement height was observed.

Line 323-324: At 50 m the NH₃ concentration exceeded that at 30 m by 0.1 $\mu\text{g N m}^{-3}$.

Line 339:

I propose to use the expression diurnal cycles instead of daily cycles throughout the MS.

Line 325: The seasonal variations of the half-hourly ΣNr concentrations are represented by box-and-whisker plots including monthly medians in Fig. S3. (delete: Figure S3 shows monthly box plots of the concentrations.)

Line 327: Medians ranged between...

Figure 2 caption: ...Missing NH_3 values from the DELTA measurements.... Numbers above the bars indicate the relative coverage of TRANC measurements during each exposure period.

Line 349: NH_3 also featured seasonal variations with....

Line 353: As shown in Fig. 2, ΣNr ...

I would split the first results section in two parts:

3.1 Measured concentrations of individual reactive nitrogen compounds

Including Figures 1-3 and S1-S4

3.2 Measured exchange fluxes of total reactive nitrogen

Starting on page 14 (break at line 355)

Line 355-356: ...on a monthly timescale....

Line 359: on a half-hourly basis...., On a monthly basis...

Line 360: According to Langford et al. (2015), the limit of detection (LOD) is calculated by multiplying the random flux error (95% confidence limit) with 1.96.

Line 364: This indicates that emission fluxes....

Line 365: In general, median deposition was within the same range for the entire campaign with only small seasonal differences.

Line 367-368: Median deposition was significantly increased from June 2016 till September 2016 than for the same period in 2017 and IQR and whisker also covered a wider range in 2016.

Line 374: Fig. 5 shows averaged daily cycles of measured ΣNr fluxes for every month.

Figure 5: Mean diurnal cycle of ΣNr fluxes ($\text{ng N m}^{-2} \text{ s}^{-1}$) based on half-hourly measurements for every month from June 2016 to June 2018. The shaded....

Line 374-375: In general, the ΣNr diurnal cycle exhibited low deposition or fluxes close to zero during nighttime/evening and increasing deposition during daytime. Deposition fluxes were...

Line 378: ...with near-zero or small negative fluxes...

Line 379: ... months were comparable.

Line 381-382: ...was close to zero one year later.

Line 386: Again, the average standard error...

Line 390: The meaning of “From May to September, the curve was approximately bell-shaped.” is unclear. Please clarify.

Line 393: 3.3 Controlling factors...

Line 396: “leading to a constantly low v_d during the day (Fig. S10).” From Fig. S10 it is evident that v_d even strongly decreases during midday, this should be mentioned (and explained in the discussion).

Line 399: and **the** concentration of ΣNr , especially changes in the concentration of the **individual nitrogen compounds**....

Line 410: For visualizing the impact of **the** concentration on v_d (Fig. 6),...

Line 412: ...increments of **the** ΣNr concentration...

Line 413: ... on **the** ΣNr concentration...

Line 414: It demonstrates that **the** ΣNr concentration...

Line 419: ... v_d was **more influenced by** micrometeorological **variables than by the** ΣNr concentration.

Line 425-426: Combine 2 sentences, they should read as: **“During winter (December, January, and February), v_d was almost equal and even lower during the day, which resulted in a lower deposition of ΣNr .”**

Line 426-428: The sentence should read as:

The different shapes of the diurnal variations of v_d could be induced by micrometeorological variables, which change the composition of available ΣNr compounds during the day (~~Seinfeld and Pandis, 2006~~) and promote photosynthesis (e.g. stomatal uptake or release of NO_2 and NH_3).

Seinfeld and Pandis, 2006 is for sure not the appropriate literature here, please choose other more specific references (as in the discussion section).

Figure 7. Mean diurnal cycle of v_d from May to September for low and high temperature (a), relative humidity (b), and concentration (c). Median....

Line 435: ... lower relative humidity....

Line 437: During dawn/nighttime, deposition velocities exhibited no significant difference between the applied thresholds.... I can see a difference for the dry/wet leaf surface. Please double check this statement.

Line 439: ...compared to the May to September **period**.

Section 3.3 should be changed to:

3.4 Dependence of ΣNr dry deposition sums on micrometeorological variables

Figure 8. ... represented by box-and-whisker plots...

Eq. (3) (Pastorello et al., 2020) → please refer to the discussion section here.

Line 450: median deposition**s** of **the** ΣNr **fluxes**-with....

Line 451: median deposition**s**-... → please correct all instances

Figure 9. Annual Σ Nr dry deposition shown as bar graphs

Line 459: ... dry depositions sums...

Introduce new section after Line 466:

3.5 Wet and total nitrogen deposition

Line 473: In the second year, the contribution of dry deposition...

Line 476-477: Which was probably related to high NH_3 concentrations... For sure it was, you measured them, please refer to the corresponding Figure here.

Line 509: Thus, their influence on NO_x measurements was most likely small.

515-516: DELTA measurements further suggested that the Σ Nr concentration pattern was mainly influenced by gaseous Nr.

Line 521-522: Due to the reaction of NH_3 with HNO_3 and sulphuric acid particulate NH_4^+ is formed, available as NH_4NO_3 or $(\text{NH}_4)_2\text{SO}_4$.

→ I would change the order of compounds here:

Explanation:

In chemical systems composed of NH_3 , HNO_3 and H_2SO_4 , the formation of non-volatile $(\text{NH}_4)_2\text{SO}_4$ is preferred. Only when NH_3 is available in excess of H_2SO_4 and when favourable meteorological conditions (low to moderate T and/or high RH) prevail, neutralization of HNO_3 vapor with NH_3 occurs (Trebs et al., 2005).

*Trebs, I., Metzger, S., Meixner, F.X. et al., 2005. The NH_4^+ - NO_3^- -Cl $^-$ - SO_4^{2-} - H_2O aerosol system and its gas phase precursors at a pasture site in the Amazon Basin: How relevant are mineral cations and soluble organic acids? *Journal of Geophysical Research-Atmospheres*, 110(D07303): doi:10.1029/2004JD005478.*

Line 523: ... fine mode and associated with aerodynamic diameters....

Line 537: ..., but were probably...

Line 553-554: ...for instance by bidirectional exchange of NH_3 leading to both periods of net emission and deposition of Σ Nr.

Line 567: Also, the SO_2 concentration was much larger...

Line 577: ...resulting in a high vd, which is due to efficient turbulent mixing. Hence, even at low concentrations...

Line 578-579: In conclusion, particulate NH_4^+ was mainly responsible for the large Σ Nr deposition due to its excess over aerosol NO_3^- .

I propose that the section on Σ Nr emission and the influence of snow can be shortened. The English writing of this section must be improved (Lines 590-609).

Line 616: I think it is anyway highly unlikely that the concentration drives the deposition velocity.

However, the impact of increasing concentrations on....

Line 624: ... was nearly zero and emission...

Line 631: ... contribution of individually compounds do show a seasonal cycle. Since the Σ Nr compounds differentiate in their vd,...

Line 635: ...than **of** NO₂, but... than **of** NO₂ for woodland.

Line 636: ...and 2.2 cm s⁻¹ **for** NH₃ (see Schrader...

Lines 637-643: Rewrite to:

However, variations in the composition of ΣNr may correlate with micrometeorological parameters. For example, the formation of HNO₃ is correlated **with** Rg. The solar radiation responsible for the stomatal opening also promotes the formation hydroxyl radicals, which react with **NO₂ to form HNO₃** (Seinfeld and Pandis, 2006). Tair influences the diurnal pattern of NH₄NO₃, which may also volatilize close to the surface due to the depletion of its precursors and in case the temperature gradient is large enough (Wyers and Duyzer, 1997; Van Oss et al., 1998). Thus, **part of the NH₄⁺ and NO₃⁻ in the aerosol phase** may be converted to NH₃ and HNO₃, which **deposits faster to surfaces** than aerosols.

Line 646: In conclusion, **the** variability...

Line 648-649: Delete: ~~Definitely, ΣNr concentration had no influence on its deposition velocities.~~

Line 656: ...measured half-hourly values...

Line 657: ... low-quality **half-hourly values were** effectively...

Line 660: Was there any footprint analysis performed or required due to fetch limitations? Could you comment on that? Maybe refer to previous publications.

Line 669: of turbulent motions**s**...

Line 675: As **shown** in Fig. 8...

Line 679: ...a certain **half-hourly value** was...

Line 688: ...estimated dry deposition**s** for...

Line 693: ...has a distinct diurnal cycle.

Lines 701-702: Please delete: ~~The comparison of TRANC measurements with nitrogen throughfall measurements will be shown the second part of this study.~~

Line 705: ...total N deposition**s was**...

Lines 708-709: It suggests that the forest is currently not in a critical state in relation atmospheric N input.

→ I think this statement is incorrect. The N input was 10 and 12 kg N ha⁻¹ a⁻¹, which is within the range of the critical load.

According to the OECD, the critical load is defined as:

Critical Load is the quantitative estimate of the level of exposure of natural systems to pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur.

According to my understanding, the forest is just at the limit of receiving too much Nitrogen from the atmosphere. This implies that N inputs should not increase in the next years.

Line 713: ...above a protected **temperate** mixed forest, **that is located in a remote area**.

Line 721: ...throughout the year.

Line 726: ...periods of high solar radiation...

Line 727: seasonal changes in the concentrations of the ΣNr compounds,...

→ Before it was written that ΣNr does not influence v_d

Please double check.

Line 728: From May to September, ~~deposition velocity~~ v_d was....

Line 732-733: ~~Still, a comparison of measured and modeled deposition velocities of ΣNr with the latter being determined by inferential modeling with regard to micrometeorological controls, could hint on deficits in deposition modeling.~~

→ This sentence does not make sense, please delete.

Line 735: No significant influence of micrometeorological parameters on estimated dry deposition sums was found.

→ This sentence does not make sense and is in contrast to what was written before. (micrometeorology influences v_d and therefore also the total N deposition)

Line 736-737: ~~Using gap-filling approaches based on inferential modeling for long-term gaps, is an option which we investigate in the companion paper.~~

→ Please delete, note relevant here.

Please add information to the conclusion that dry deposition contributed 1/3 to the total N deposition.

Rephrase Supplement:

A1 Description of wet deposition measurements

Figure S3: ...shown as box-and-whisker plots....

Figure S4. Mean diurnal cycle of ΣNr concentrations ($\mu\text{g N m}^{-3}$) based on half-hourly measurements for every month from June 2016 to June 2018.

Figure S5: ... presented by box-and-whisker plots...

Figure S6. Mean diurnal cycle of $v_d(\Sigma\text{Nr})$ (cm s^{-1}) based on half-hourly measurements for every month from June 2016 to June 2018.

Figure S11. Diurnal cycles...

Figure S13.Wind direction corresponds to values measured in three-hourly intervals.