

We would like to thank the anonymous reviewer for the comments that significantly improve the clarity and readability of the manuscript. Our point-by-point responses are found below in blue ink. The revised content is highlighted in yellow.

The authors report results from an aerosol sampling campaign in a rural cloud forest during December 2018. Different size fractions were sampled on filters taken during daytime and night-time and during some days fog events impacted the aerosol composition. The most important measured aerosol components were ammonium, nitrate, sulphate, and black carbon. Ammonium and nitrate were also analysed for stable isotopes of nitrogen and oxygen.

The study nicely shows local dynamics of aerosols and their partitioning into different size fractions. Differences in stable oxygen isotopes of nitrate during foggy conditions revealed a possible oxidation pathway involving peroxy radicals.

1. My major concern is the performed source apportionment using the stable nitrogen isotopes and a mixing model (MixSIAR). Many aspects of the procedure are insufficiently described (e.g. what is posterior in this context, and how should probabilities be interpreted). Table 4 seems to list the results of the source apportionment. I see mostly values around 20 with standard deviations around 15. A threshold of 20 is applied, but the choice of this value is not motivated. Overall, most values do not seem to be significantly different. I fail to see how any conclusions can be drawn from this model. Therefore, I suggest to remove this part.

A: The MixSIAR is a Bayesian mixing model to infer the probable sources of a mixture using given prior information. In this study, the mean values and standard deviation of stable isotope from different sources in a previous study was applied as the prior data and assumed to have a Gaussian distribution. After applying the Bayes' theorem, the posterior probability is the conditional probability based on these observation data. The following description in Lines 146-150 is revised as follows for clarification: "MixSIAR is a statistical model applying Bayesian Inference to infer the posterior probability of mixture sources by analyzing its tracer composition, such as stable isotope or fatty acids (Stock et al., 2018). The studied tracers are assumed to transfer from sources to the mixture through a conserved mixing process integrating the observed variability. In this study, the observed mass-weighted $\delta^{15}\text{N}$ of NH_4^+ and NO_3^- for each sampling period was used as prior information of the mixture."

The similar isotope values for some applied source data (i.e., traffic, industries and fertilizers for NH_4^+ $\delta^{15}\text{N}$, and CFPP and urban for NO_3^- $\delta^{15}\text{N}$) can lead to comparable posterior probabilities. However, the results can differentiate the sources with significantly different isotopes, such as relatively lower probabilities of feedlots and traffic in NH_4^+ $\delta^{15}\text{N}$, and fertilizers in NO_3^- $\delta^{15}\text{N}$. With the source and sample variability, the results of MixSIAR provide broader probabilities for source contribution, which might reflect the uncertainty of the ambient conditions. However, the possible differentiation among the similar $\delta^{15}\text{N}$ sources might require the integration of the back trajectory and model simulation with the known emission sources. In the content, the following information is added in Lines 282-

286 (section 3.3) to address this issue; “The similar posterior probabilities among some sources are due to the comparable source isotope values as stated above. However, with the source and sample variability, the results of MixSIAR provide a broader probability for source contribution and reflect the uncertainty of the ambient conditions simply using the mixing rule. The possible differentiation among the similar $\delta^{15}\text{N}$ sources might require the integration of the back trajectory and chemical transport model simulation with the known emission sources.”

Minor comments:

2. Language needs to be improved. Several issues... already in the first sentence of the abstract (aerosol components NOT compositions).

A: Thanks for the reviewer’s comment. We went through the content to correct the word and grammar for clarification. Some examples are shown as follows:

Line 9. “Ammonium and nitrate are major N-containing aerosol components.”

Line 36. “Ammonium and nitrate are the primary N-containing cation and anion species, respectively, ...”

3. Figure 1: extend the figure to also indicate how daytime and night-time chemistry results in different stable isotope composition. A good description is given in the supplement. Maybe some of this can be incorporated in Fig 1.

A: Fig. 1 and the figure caption are revised with the isotope values from fresh and aged gas precursors via different chemical pathways as follows:

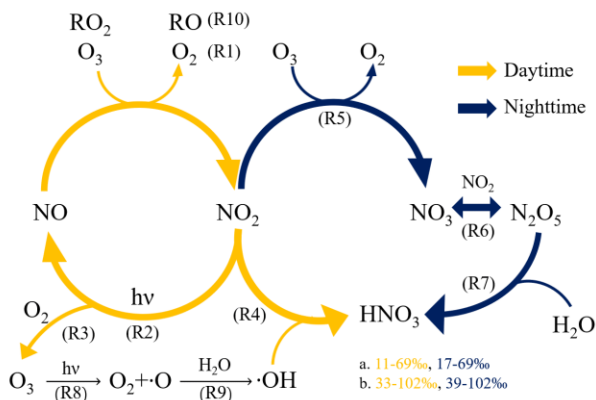


Figure 1. The formation pathway of nitric acid to form aerosol nitrate during daytime (orange color) and nighttime (blue color) with the predicted $\delta^{18}\text{O}$ range of NO₃⁻ based on

(a) freshly emitted NO and (b) NO cycled from NO₂, fully reacted with O₃ (detail can be found in Figures S6 and S7).

4. L132/133: I am not sure if organic nitrogen can be neglected. There are several papers out reporting organic nitrates and other organic nitrogen compounds in aerosols. The authors should at least discuss how their results would change if there are significant fractions of other nitrogen compounds.

A: The presence of organic nitrogen in aerosols is undeniable. However, the water-soluble reduced nitrogen, e.g., ammonium and organic nitrogen, can be estimated as the difference between total nitrogen and nitrate as WS(TN-NN). WS(TN-NN) shows a good correlation (slope is close to 1 with a small interception as shown in Figure S4) with the estimated ammonium determined using FT-IR. The result suggests that ammonium is the significant component of the reduced nitrogen for this studied case. However, the presence of organic nitrogen might lead to some deviation of the determined $\delta^{15}\text{N}$ NH₄⁺. Organic nitrogen might be related to NO_x and was reported a lower $\delta^{15}\text{N}$ than nitrate (Wu et al. 2021), less than -5‰. If organic nitrogen with a lower $\delta^{15}\text{N}$ than nitrate is taken into account, we can expect a slightly higher $\delta^{15}\text{N}$ NH₄⁺ than the current reported values. We added the following sentence to Lines 141-143 to address this issue, “If organic nitrogen is considered, a slightly higher $\delta^{15}\text{N}$ of NH₄⁺ than the current reported values can be expected because organic nitrogen might be related to NO_x and was reported a lower $\delta^{15}\text{N}$ ($\leq -5\text{‰}$) than nitrate (Wu et al., 2021).”

Wu, L., Yue, S., Shi, Z. *et al.* Source forensics of inorganic and organic nitrogen using $\delta^{15}\text{N}$ for tropospheric aerosols over Mt. Tai. *npj Clim Atmos Sci* **4**, 8 (2021). <https://doi.org/10.1038/s41612-021-00163-0>

5. L214-216: This sentence has language issues. The argumentation does not seem to be logical.

A: The sentence is revised in Lines 225-228 to clarify the argument as follows: “As stated in section 3.1.2., nitrate significantly contributes to the submicrometer particles during foggy daytime in addition to the usual peak over the supermicrometer particles for all conditions (Fig. 2). The nitrate can be divided into two groups, PM₁₋₁₀-NO₃⁻ for particle size in the range of 1 to 10 μm and PM₁-NO₃⁻ for particle diameter less than 1 μm , for further discussion.”

6. L228-230: Was there any evidence for agricultural activity during that period? What was different compared to other periods?

A: Since the sampling site was in a nursery of the experimental forest, some agricultural activities happened during the observation period. We recorded that fertilizers were applied on the field on December 18th, and the scheduled mowing activities nearby the sample collection site were on the daytime of December 20th and 21st. The sentence is revised as

“ The sample of 21D is a special case with higher $\delta^{15}\text{N}$ values. It might result from the recorded agricultural activities nearby,...”.

7. L281-282: “The posterior probability of PM1 and PM1-10 nitrate sources has difference slightly.” This seems to be a mixture of poor English with lab/model-slang.

A: The sentences are revised in Lines 296-300 as follows: “The difference in posterior probability between PM₁ and PM₁₋₁₀ nitrate sources is not significant: the PM₁₋₁₀ NO₃⁻ was more likely from CFPP, industries, or urban sources, while industries had the majority of PM₁-NO₃⁻ formation. However, the inferred source difference might suggest that the coarse mode aerosols came from the coastal sea salt particles mixing with the emission of coal-fired power plants or the Taichung-Changhua metropolitan during the inland transport.”