

# **Title: Aerosol dynamics and gas-particle conversion in dry deposition of inorganic reactive nitrogen in a temperate forest**

**Authors: G. Katata et al.**

## **Author response to reviewer comments**

### **Response to Anonymous Referee #3**

General The author present the development of an advanced deposition model, here for the inorganic reactive nitrogen gas phase species  $\text{HNO}_3$  and  $\text{NH}_3$  and particle species  $\text{NO}_3^-$  and  $\text{NH}_4^+$ . I suggest to use not use the term ‘aerosol’ when solely particles are addressed. Use the term ‘aerosol’ when you address particle together with the gas phase where they are dispersed in. Otherwise use ‘particle’.

*Response:* Thank you for your suggestion. As you suggested, we replaced most of words of “aerosol” into “particle” throughout the manuscript.

Surely, deposition much deserves a better treatment in many atmospheric model, so in principle an improvement in deposition schemes is highly welcome.

*Response:* We appreciate for your positive comments on our work.

Overall, I feel the model can deliver useful results but there are many approximations it its set-up. This should be treated most carefully. In my view, the paper needs a huge amount of improvement but I rate this as still doable and not recommend rejection. I would therefore like to recommend major revision according to all reviewer comments with external re-review necessary.

*Response:* Thank you for your interests and many suggestions on the manuscript. As you suggested, we should have emphasized that this was first application by clearly stating the model description and setup for numerical experimental.

### **Details**

○Abstract: I feel the abstract should give more information, at best, in a quantitative manner. It now reads too much like an introduction. What is the main numerical outcome? What is better than before? The abstract should clearly state what is treated.

*Response:* As you suggested, the information was not enough for readers. We totally revised the sentences and added more sentences in Abstract as follows: “Although dry deposition has an impact on nitrogen status in the forest environments, the mechanism for high dry deposition rates of fine nitrate particles ( $\text{NO}_3^-$ ) observed in forests remains unknown and is a potential source of error in chemical transport models. Here we modified a multi-layer land surface model coupled with dry deposition and aerosol dynamics processes for a temperate mixed forest in Japan, so that we carried out its first application to the ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ) gas-particle conversion (gpc)

and aerosol water uptake of reactive nitrogen compounds. The processes of thermodynamics, kinetics, and dry deposition for mixed inorganic particles are modeled by a triple-moment modal method. The data of inorganic mass and size-resolved total number concentrations measured by filter-pack and electrical low pressure impactor in autumn was used for model input and numerical analysis. The model overall reproduces observed turbulent fluxes above the canopy and vertical micrometeorological profiles as our previous studies. The sensitivity tests with and without gpc demonstrated inorganic mass and size-resolved total number concentrations clearly changed within the canopy. The results also revealed that the within-canopy evaporation of  $\text{NH}_4\text{NO}_3$  under dry conditions significantly enhances deposition flux for fine  $\text{NO}_3^-$  and  $\text{NH}_4^+$  particles, while reducing deposition flux for nitric acid gas ( $\text{HNO}_3$ ). As a result of evaporation of particulate  $\text{NH}_4\text{NO}_3$ , the calculated daytime mass flux of fine  $\text{NO}_3^-$  over the canopy were 15 times higher in gpc" scenario than "no gpc" scenario. This increase caused high contribution of particle deposition flux to total nitrogen flux over the forest ecosystem ( $\sim 38\%$ ), while the contribution of  $\text{NH}_3$  was still large. A dry deposition scheme coupled with aerosol dynamics may be required to improve the predictive accuracy of chemical transport models for the surface concentration of inorganic reactive nitrogen.”.

○Line 13: Maybe a word should be added after ‘nitrogen’? Like ‘input’?

Response: The word of “input” was inserted into the sentence (L.19, p.1)

○Line 15, 16: Why is these only Japanese references, please check other deposition work.

Response: We should describe the background more in detail. To our knowledge, the references for “direct measurement of nitrate dry deposition” are rare except for Japanese work (c.f., Nakahara et al., 2019). To emphasize this, we revised the sentences with several references of dry deposition estimate studies in East Asia as “In East Asia, where air pollutant emissions continue to increase (EANET, 2016), although the importance of dry deposition of inorganic reactive nitrogen is suggested by prior studies by indirect estimate studies (e.g., Pan et al., 2012; Li et al. 2013; Xu et al., 2015), direct measurement studies are still limited (Nakahara et al., 2019). Recent observational studies at forests revealed that dry deposition flux of inorganic reactive nitrogen of fine  $\text{NO}_3^-$  was markedly higher than that expected from theory (Takahashi and Wakamatsu, 2004; Yamazaki et al., 2015; Honjo et al., 2016; Sakamoto et al., 2018; Nakahara et al., 2019).” (L.19-24, p.1-2).

○Line 29: This is not only known from/for deposition studies but also for myriad of particle characterization studies. Are there more recent references?

Response: We referred the textbook for well-known  $\text{NH}_4\text{NO}_3$  volatilization process in atmosphere as “although the process itself has already been known in the atmospheric chemistry community for a long time (Seinfeld and Pandis, 2006)” (L.39, p.2). Regarding to the recent papers for deposition modeling, to our best knowledge only two papers are available as follows: Nemitz and Sutton (2004) (appearing from L.41 in p.2) and Ryder (2010) suggested by other reviewer (L.309, p.10).

Ryder, J., 2010. Emission, deposition and chemical conversion of atmospheric trace substances in and above vegetation canopies. PhD Thesis, University of Manchester, UK. Available from the University or via <https://nora.nerc.ac.uk>

○Line 51: What does SOLVEG mean ?

Response: Our model “SOLVEG” was defined as “a multi-layer atmosphere-SO<sub>i</sub>L-VEGetation model” in the last manuscript (currently L.54, p.2)

○Line 74: Reference Genuchten’s concept

Response: We referred the paper for unsaturated snow modeling as “Hirashima et al., 2010; Katata et al., recently accepted) (L.83, p.3).

○Line 99: How does Eqn (3) relate to the Henry Constant? Can you clarify more what if written in the text?

Response: Eq. (3) was the equation for calculating  $\chi_s$  (stomata) and was not related to Henry’s law. Since this paragraph was not organized and confused the reviewer, we insert the word “Meanwhile,” before the sentence for  $\chi_d$  which follows Henry’s law and dissociation equilibrium with the atmospheric concentration of NH<sub>3</sub> at each canopy layer.” (L.111-112, p.4).

○Line 104ff: Where does Eqn (4) come from? ‘Affinity’ is a strange term. Better justify the approximation for SO<sub>2</sub> deposition.

Response: Following the suggestion from other reviewer, we revised the sentence as “the following empirical formula for  $r_d$  is applied” with the reference (Massad et al., 2010) (L.113, p.4). Although “affinity” was used in the original reference van Hove et al. (1989), we added the word as “(such as solubility on water)” to make its meaning clear (L.117, p.4).

○Line 117 ff: See initial remark on nomenclature and revise this whole treatment consistent with clear naming.

○Line 296: This headline must be revised. The size distribution does not have a formation mechanism, only the particles have

Response: The headlines were inappropriate; the word of “aerosol(s)” were revised to “particle(s)” when we referred solely particles, based on your suggestion.

○Line 139, end: ... of the Tokyo ...

○Line 231: ... fine particles.

Response: We corrected the sentences as you suggested.

○Line 148: What is a ‘grass fiber filter’?

Response: This was a typo; the sentence was now revised as “glass fiber filter”.

○Line 200ff: There seem to be a lot of approximations for the particle size distribution initialization. How critical can this be for the overall study?

Response: This sentence was inappropriate and confusing the reviewer. In fact, we made manual fitting independently using our ELPI+ data, which was compared with literature values as one of other examples at urban environment in autumn (Salma et al., 2011). To avoid this confusion, we revised this sentence as “The lognormal parameter sets of ( $D_{g3}$ ,  $\sigma_g$ ) for fine and Aitken modes at the upper boundary condition were respectively set at (0.089  $\mu\text{m}$ , 2.1) and (0.26  $\mu\text{m}$ , 2.0) based on manual fitting of ELPI+ measurements at 30 m height (Fig. 1a). These parameter sets were applied to both the early and late autumn periods.” (L.226-229, p.8). This uncertainty somewhat influences inorganic mass concentration profiles, but sensitivity tests with changing mean diameter showed that our important findings such as “vertical gradients of  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations drastically increased due to  $\text{NH}_4\text{NO}_3$  evaporation” (L.278-279, p.10) were consistent.

○Line 361: I think feasibility might be the wrong term.

Response: We deleted a part of the sentence “with high feasibility” since it was inappropriate.

○Figures: It would be great to show correlation plots for some key properties rather than only time-series plots.

Response: On the basis of the other reviewer, this study should not demonstrate model improvement and performance test based on scatter plots due to very limited datasets, so that we should much focus on comparisons as first application of the model to the  $\text{NH}_4\text{NO}_3$  gas-particle conversion. This is emphasized in “In this study, we made a simulation with basic and less time-resolved datasets as very first application of the model to the  $\text{NH}_4\text{NO}_3$  gas-particle conversion and aerosol water uptake of reactive nitrogen compounds. The above uncertainties associated with input data such as number concentration and particle size distribution should be improved in future.” (L.239-241, p.8)