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 #1

General Comments: This work presents an analysis of size-resolved aerosol and gaseous species concentration data from a mixed forest canopy near Tokyo using a multi-layer atmosphere-soil-vegetation model with aerosol dynamics and dry deposition. In particular, the authors hope to explain observed apparent fluxes of NO_3^-, NH_4^+ and HNO_3 above vegetative canopies by within-canopy evaporation of ammonium nitrate (NH_4NO_3).

In general, I believe this to be important work and interesting, valuable data, and I also agree with their major conclusion that 3-D chemical transport models need to better incorporate the within-canopy aerosol dynamic/equilibrium processes that are the focus of this work.

Response: We sincerely appreciate your interests and positive comments on our manuscript.

However, I believe that this article needs additional polishing to make its presentation more effective and the results more accessible to ACP readers. I offer my suggestions for this presentation enhancement below. I recommend that the article be published after additional measures are taken to enhance its presentation.

Response: Thank you so much for helpful suggestions; we revised the manuscript as follows. We hope that the manuscript is drastically improved.

Specific Comments:

○One concern is that the model used in this study is not adequately described. The paper does reference other published articles (Katata et al., 2013; Katata et al., 2014) where portions of the model are described in some detail; however, the full model used here seems to have been described in a gray literature document (Katata and Ota, 2017). It would be better if the authors included more model description in this paper, especially providing information on model setup for this particular application (e.g., model inputs, number of layers, time resolution, model outputs, etc.), referring to the other publications (or an Appendix) for details.

Response: Since a large part of description in Katata and Ota (2017) consists of our two published articles (Katata et al., 2013; 2014), the model has been basically reviewed in scientific journals. As suggested by the reviewer, we added the summary of simulation settings as supplemental table (Table S1) in addition to subsection 3.2.

A second major concern is the presentation of some of the Figures, as follows:

○Figures 2 & 3 – The figures are very small and cannot be adequately evaluated, especially with respect to

the agreement between the measurements and model results. Some way needs to be found to present the figures in a larger, clearer way.

Response: As you suggested, these figures were too small for evaluation. We increased axis fonts of old Figs. 2 and 3, and separate to two figures as new Figs 2-5.

○Figure 5 – The colors chosen for the vertical profiles are very difficult to distinguish between some species. Bolder color differences would be a great improvement in understanding this very important figure.

Response: The figure (new Fig. 7) was revised with different colors which enable readers to understand.

○Figure 9 – The y-axis title is so small as to be illegible. Please increase.

Response: As you suggested, the font of both axes of new Fig. 11 was increased with a modification of alignment.

○p. 4, lines 90-91, line 104, line 111: There seem to be two definitions of "a", one which I believe is the leaf area density, and the other a constant in Eq. (4).

Response: Those confused the reviewer. We defined "a" as the leaf area density in Eqs. (1) and (2), and new "b" as the constant for Eq. (4).

○p. 4, lines 90-91: The term "R'" is not defined.

○p. 4, line 104: "T_c" is not defined.

Response: We defined the above variables.

○p. 4, Eq. (4): A reference should be provided for this formula, which is Massad et al. (2010), ACP, 10, 10359-10386.

Response: We added the reference as suggested.

○p. 6, line 148: Was it really a "grass" fiber filter or was this supposed to be a "glass" fiber filter?

Response: It was typo; we corrected as "glass" (L.171, p.6)

○p. 8, Section 3.3: The description of the simulation scenarios is somewhat confusing on first reading. The phrases "NH_4NO_3 equilibrium" or "no NH_4NO_3 equilibrium" might lead someone to believe that nonequilibrium thermodynamics is being modeled here, when actually it's just that no NH_4NO_3 gas-particle exchange is being allowed in the "no NH_4NO_3 equilibrium" scenario. A possible suggestion might be something like "NH_4NO_3 gas-particle conversion" and "no NH_4NO_3 gas-particle conversion".

Response: Since our wording was confusing, all of NH4NO3 equilibrium was replaced to "NH4NO3 gas-particle conversion".

○p. 4, line 96: Should be "perfect absorption".

○p. 6, line 150: If I understand the intention of the sentence, both instances of "reading" could (and should)

be deleted – “We obtained 5 daytime data sets and 6 nighttime data sets.”

○p. 8, line 235: Should read “… on both NH_3 and fine NH_4^+ concentrations …”

○p. 8, line 239: Should be “… competing shrinkage mechanism, …”.

○p. 9, line 252: Should be “… evaporation has less impact on

○p. 9, line 269: Should be “among”, not “amoung”.

○p. 11, line 334: There are two instances of “typically” in this sentence, which is awkward.

○p. 12, in “Author contributions”: Should be “… developed the model with support from MK,”.

Response: All items were revised. Thank you for your suggestions.

○p. 11, first paragraph in Section 5.3: This discussion here (and Figure 10) is very confusing. How can the ratios be plotted as a function of RH, but distinctions still made between “high RH conditions” and “low RH conditions”? Whatever the subject is here, it is needs to be more clearly explained.

Figure 10 – As mentioned below, this figure (and the discussion that goes with it) is very confusing. I don’t really understand what point is being made with this figure (ratios as a function of RH, but under “high RH conditions” and “low RH conditions” – this doesn’t make sense as explained).

Response: We should explain about the background of the figure in more detail. We added the sentence about “In this study, since water uptake of aerosols, typically represented as the hygroscopic growth factor defined as the ratio between the humidified and dry particle diameters, is almost negligible under RH < approximately 80 % and increases over RH > 80 % (e.g., Fig. 6 in Katata et al., 2014), we defined the threshold of 80 % for high and low RH conditions.” (L.362-365, p.12).