Response to Reviewer #3

This manuscript deals with an episodic model simulation and evaluation with PM2.5 and its composition measurements. the relationship among sulfate, nitrate and ammonium and the neutralization degree are well explained and discussed with the observations and simulation results. The manuscript provides interesting and scientific information on PM2.5 formation and transport in Northeast asia. However it is not clear what the authors want to tell in the manuscript. The contents are good enough, but it would be better reconsider the discussion points. Specific comments are listed below.

Dear Reviewer #3,

Thank you for taking the time to review our manuscript for *Atmospheric Chemistry and Physics*, and providing helpful comments.
We have revised our manuscript according to the reviewers' comments and suggestions. We believe that these revisions address all points raised by the reviewers. We also provide a point-by-point response below. The revisions are indicated in blue in the revised manuscript.

Sincerely,

Syuichi Itahashi

Abstract, Lines 29-35, "Analyzing the gas ratio, which is an indicator of the sensitivity of NO3- to changes in SO42- and NH4+, showed that the air mass over China was super NH3-rich for type N, but was almost NH3-neutral for type S."

1) Is it because of low SO2 to Sulfate conversion rate in Type N? It should be also noted that the observed levels of total NH3 in Figure 6 are almost identical for Type N and Type S.

   In the two cases analyzed in this study, the decomposition of NO_3^- was determined by the production of SO_4^{2-}. The decomposition rate of NO_3^- and the production rate of SO_4^{2-} during transport from China to Fukuoka are marked in Fig. 13.

2) Why are NH3 conditions for Type S and Type N different when we consider the target modeling period is quite short?

   Because the backward trajectories were similar in both types, NH_3 emission intensity is not a factor in governing the NH_3 conditions. Due to the short lifetime of NH_3 in the atmosphere, NH_3 condition can change within the target period.

3) What determines Type N and Type S? Is it due to meteorology, different origins of air plumes, or pathways of back-trajectories? It seems that outflow from Shanghai area affects the monitoring sites for Type N, and air plume is transported from Hebei and Beijing for the Type S.

   The meteorological conditions that affect SO_4^{2-} production are one factor in determining type N and S in our case. Because the backward trajectories were similar in both types, the origins of the air mass and the related emission intensity would not be important factors.

   Although we can find a high concentration around Shanghai in Fig. 9, the air masses from Hebei and Shandong provinces were mainly transported along the backward trajectory of T_N from China to Fukuoka, as shown by the analysis of temporal variation of spatial distribution.

Figure 5, Why domestic influence does not appear on January 13 and 14 when dominant wind direction shown on Figure 4(c) is easterly?

On 13 and 14 January, domestic contributions for nitrate were found at Fukuoka (Fig. 6). With regard to SO_4^{2-}, the local contribution at Fukuoka was small through the year, as we showed in a previous study (Itahashi et al., 2017) and as was also suggested from observation (Kaneyasu et al., 2014). Over the remote islands of Goto and Tsushima, and the remote site of Tottori, no local contribution was found, which we attribute to the low amount of SO_2 emissions.

Page 9, Lines 10: "the main component of SNA was NO3- during the first episode and SO42- during the second episode." –> It would be helpful to indicate quantitative portions of nitrate and sulfate for the episodes.

Quantitative portions have been added in the revised manuscript. We have added the following sentences (P9, L33-P10, L1).

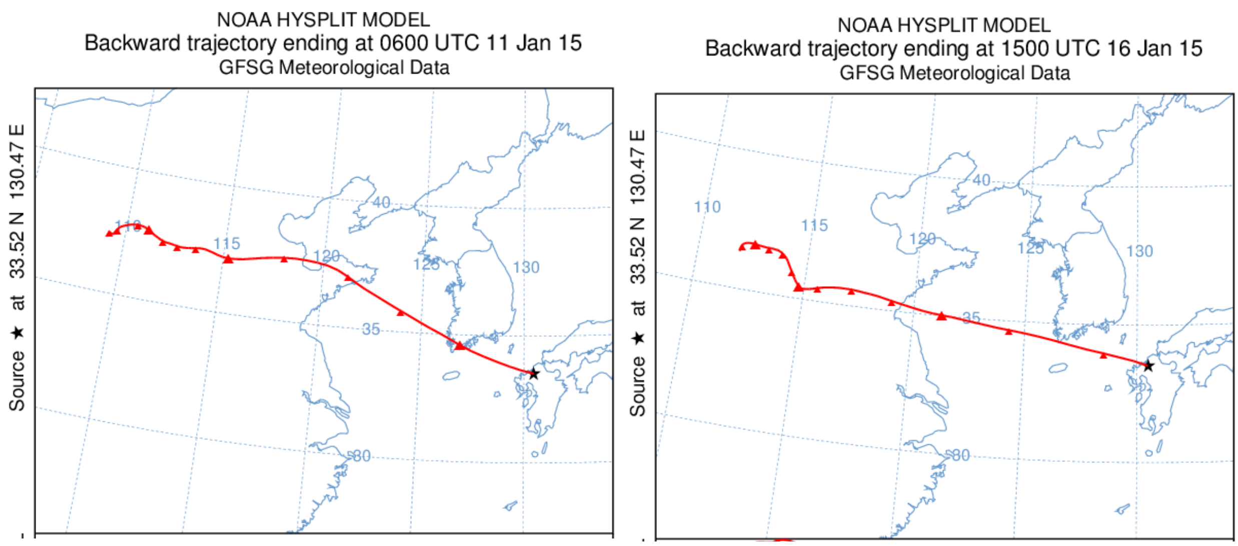
“At Fukuoka, the relative portions of SO_4^{2-}, NO_3^-, and NH_4^+ within $PM_{2.5}$ were respectively 18%, 20%, and 14% during the first episode, and 27%, 6%, and 14% during the second episode.”

Page 9, Line 20: “ However, 20 the sensitivity simulation confirmed that the transboundary NO3- air pollution was dominant for types N and S.” –> The sentence is not clear. Does it mean that Type N and Type S are determined by sulfate concentration or Fs rather than nitrate concentrations?

For the two cases analyzed in this study, nitrate was directly transported from China and decomposed into gas-phase during transport from China to Fukuoka, which was clarified in Fig. 13. The decomposition rate can be determined from the sulfate concentration.

Trajectory analyses: It seems trajectory results should be checked. For the Tn case in Figure 9, the trajectory and wind vectors are relatively well matched. However, in Figure 10, more northerly wind is dominant, and the trajectory origin should move more northward. But, tracjectoies in Figures 9 and 10 are almost the same.

Because the wind fields on Figs. 9 and 10 were thinned for better visualization, some differences between Type N and S are displayed. We have checked the backward trajectories again and confirmed that the transport pattern is similar between types. The original figures for trajectory analysis are shown as Supplemental Figure 3-1.



Supplemental Figure 3-1. Backward trajectory for (left) Type N, and (right) Type S.