Response to Reviewer #1

This study analyzed two episodes (characterized as type N and type S according to the dominant compositions) with the high PM2.5 concentrations reaching around 100 μg/m3 during an intensive observation campaign in January 2015 at Fukuoka in western Japan. Several ground-based measurements and the CMAQ model as well as the path analysis of HYSPLIT model have been utilized to investigate the transboundary air pollution for both types. Authors addressed their results with the comprehensive methods and proved the importance of the transboundary air pollution dominated by NO3-, which will help refine our understanding of the transboundary heavy PM2.5 pollution in winter over East Asia. However, there are several rooms the paper can be much improved scientifically, such as the non-linearity effects of the sensitivity simulation to the secondary pollutions and the explanation of high speed of transboundary air pollution. If we take the 1000 km distance between coastline of China and western Japan, which is assumed by the authors, the transport of air mass speed will be almost 15 m/s while the traveling time is 18h. Is this reasonable for the wind speed reaching so high during the observation period? Overall, this is a nice piece of paper with clear objectives and methods and will provide valuable results. I recommended it for publication in Atmospheric Environment after minor revisions. Some comments and suggestions are listed as follows:

Dear Reviewer #1

Thank you for taking the time to review our manuscript for *Atmospheric Chemistry and Physics*, and providing helpful comments. To address your comments on the non-linearity of effects involved in this study, we have revised and added explanations. For the wind speed, we think we have fully addressed your concerns.
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 have also provided a point-by-point response below. The revisions are indicated in blue in the revised manuscript.

Sincerely,

Syuichi Itahashi

1) On Page 3, Line15. Observation and model simulation section. Authors should introduce their dealing methods for the different data. For example, the chemical compositions of aerosols measured by ACSA-12 and Denuder-filter pack method are 1 hour and 6-8 h, respectively. For CMAQ model, it is the hourly results. So, how could authors get the statistical parameters like R, MFE?

The statistical analysis for $PM_{2.5}$ discussed in Page 8, Lines 7-9 and Lines 25-26 were, respectively, based on ACSA-12 and BAMs. The measurement interval is 1 h. We have compared these observational data directly to CMAQ model output data.

2) On Page 6, Line12. Authors introduced the emission settings in the model simulation. They assumed the emissions in 2008 are similar with that in 2015. Although they issued the NO2 column in China from satellite observation is similar to those for 2009, the SO2 is complicated. How is the picture for SO2 emission? And How about the VOCs? At least, the emission amount for the primary air pollutants between China, Korea and Japan should be listed out.

SO_2 emission might be overestimated because of the assumption about the emission level in the year 2008. Because the appropriate reference for VOC is not available, the emissions amount of VOC is assumed to be at the 2008 level. In this study, our focus was on the behavior of sulfate-nitrate-ammonium (SNA). We have listed the emissions amounts for SO_2, NOx, and NH_3 in Table 1, which is newly included. We have added a brief comment about this issue in Section 2.2.

3) On Page 7, Line3. "Because the amount of emissions from China is larger than that from Japan, to avoid large nonlinearities in the atmospheric concentration response to emissions variation (e.g., Itahashi et al., 2015), the sensitivity simulation was designed to switch off the anthropogenic emissions in Japan." Why the anthropogenic emissions in Japan be switched off could avoid the nonlinearities? What is the amount of the anthropogenic emissions taken up in Japan, and how about the other sources, like biogenic and agriculture? Because, based on the previous study, the emission cut by 20-30% may decrease the nonlinearities in maximum in the sensitivity simulation.

We agree that the method with an emissions cut by 20-30% (e.g., Fiore et al., 2009, J. of Geophys. Res. 114: D04301) is a suitable approach, but for simplicity, we have applied the zero-out method in this study. We have fully revised the relevant sentence as follows (P7, L21-25).

"In terms of O_3, which is involved in complex nonlinear chemistry, larger nonlinearities in the atmospheric concentration response to emissions variation for China but not Japan were clarified due to the higher amount of emissions from China than from Japan (Itahashi et al., 2015). Therefore, the sensitivity simulation was designed to remove anthropogenic emissions in Japan instead of those in China."

The term 'anthropogenic' indicates that the emissions were taken from the REAS inventory; because of this, the agriculture category was included but the biogenic category was not. We have also added an explanation of the treatment of anthropogenic emissions as follows (P7, L19-20).

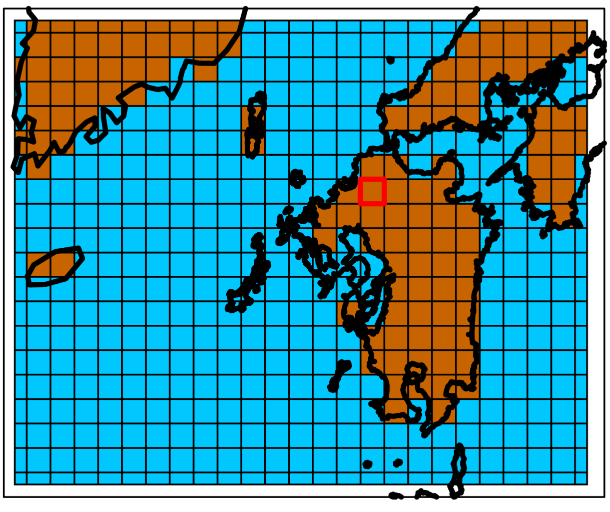
"Here, anthropogenic emissions were taken from the REAS inventory; because of this, emissions from agriculture were included."

4) On Page 7, Line9. It seems that Fig 2. should be Fig 4. and same as that Fig. 2a to Fig 4a.

This mislabeling has been corrected. We have also inserted the appropriate figure number for the other meteorological parameters.

5) On Page 7, Line20. Temporal variation of particulate matter. Authors presented very good simulation of particulate matters as well as their compositions in Japan and China during the period. It is curious to me, during the type N and type S episodes, the simulated wind speed is much higher than the observations, how the air pollution simulated well?

The wind speed at Fukuoka, Japan, is overestimated compared with observations, as we have shown in Fig. 4b. This is partly related to the land use mapping at 27-km resolution. The grid corresponded to Fukuoka is assigned to the land category; however, the surrounding north-west grid is assigned to the ocean category (Supplemental Fig. 1-1). Therefore, the observed slowed wind speed, which arises from the large effect of friction over land, might not be simulated well. Please also see reply 9).



Supplemental Figure 1-1. Mapping of land (brown) and ocean (light blue) categories around Kyushu island. The red square indicates the grid square of Fukuoka.

6) On Page 8, Line10. “therefore, the transboundary air pollution was dominant during January 2015”. First, similar as the above mentioned how the authors delimited the non-linearities just from switch off the anthropogenic emission in Japan? Second, how about the anthropogenic emission take up in the whole emission in Japan, what about the biogenic, such as ocean sources?

First, this sentence mentioned the transboundary air pollution status at Goto, Tsushima, and Tottori. We have added this point (P8, L30-31) as “at remote sites in western Japan”. At Fukuoka, we found domestic contribution in some cases. Because the zero-out method was applied to the emissions of Japan instead of those of China, nonlinearity will be smaller in the case of the zero-out method for Chinese emissions. To support a discussion about nonlinearity of the relevant chemistry, we further used a BC variation to investigate the local and transboundary contributions. Considering that BC variation, the dominance of the transboundary air pollution on both types can be assumed.

Second, only anthropogenic emissions were switched off in this case; the biogenic emissions were not switched off. As a biogenic source, dimethylsulfide (DMS) emissions from oceans were not included in the modeling system. We have explicitly mentioned the treatment of DMS (P7, L10-11).

7) On Page 10, Line14. “Based on the model results, because the domestic contribution for HNO_3 was observed on January 14”. It is confused to me that HNO_3 was observed since this the model results.

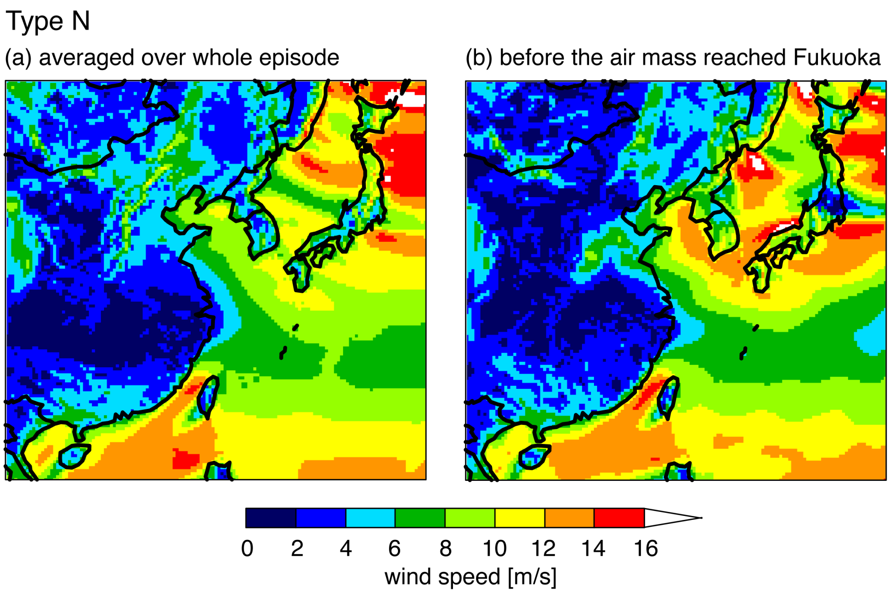
This discussion was based on model results. To avoid a misreading, we have changed the wording from “observed” to “found.”

8) On Page 10, Line23. BC section. BC is over estimated during both type N and type S episodes, while SO_4^{2-} and NH_4^+ is underestimated. Can the authors explain this? Since in the following sections, “the rates of decrease of total sulfate, total nitrate, and total ammonia were generally consistent with the rates of decrease of normalized BC and CO.” (On page 14, Line 26), and ”For SO_4^{2-}, the concentration was higher when the air mass arrived at Fukuoka compared with that in China, suggesting the fast production of SO_4^{2-} during the transport process.” (On page 12, Line 3), if the BC is over estimated, the SO_4^{2-} should be more overestimated. One exceptions, the BC or the transboundary has been overestimated in China.

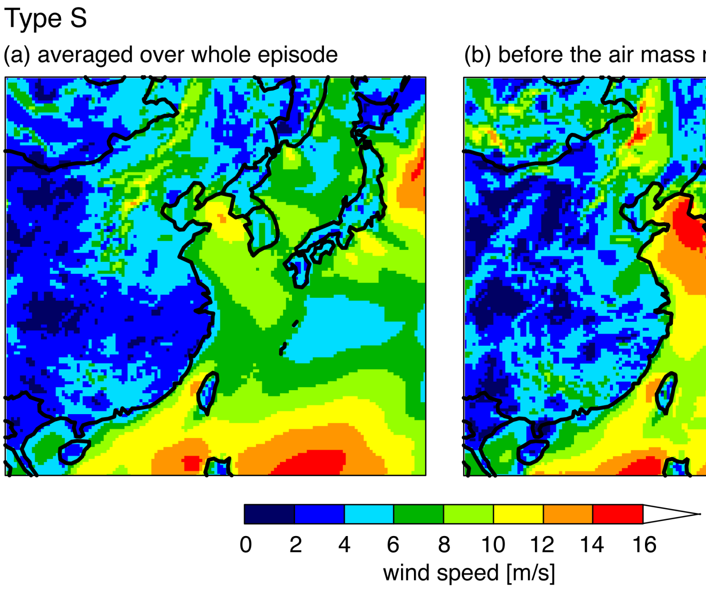
Because BC concentrations are changed via emission/deposition/transport processes, but SNA concentration are also involved in the chemistry, the model tendency to overestimate concentration is not necessarily related. The reason for model overestimation of BC at Goto might be related to the assumption about BC emissions in China.

9) On Page 11, Line25. "The traveling time from the coast of China to Fukuoka was about 18 h." As it is mentioned above, the traveling speed will be reached at 15 m/s, which means the wind speed should be 15 m/s. Is this reasonable? From the observations of meteorological conditions in Fukuoka, during the two episodes, the wind speed is 5-8 m/s, which is significant slow than 15 m/s.

As we mention in reply 5), the wind speed at Fukuoka is slowed by friction over land. In Supplemental Figures 1-2 and 1-3, wind speeds are shown for types N and S, respectively. In both types, episode-averaged wind speed over the Yellow Sea ranged from 8 to 12 m/s, which is greater than the observed wind speed at Fukuoka. Before the air mass arrived at Fukuoka, wind speed was further increased, beyond 12–16 m/s, over the eastern part of the East China Sea.



Supplemental Figure 1-2. Wind speed during type N episode (a) averaged over whole episode, and (b) averaged 3 h before the air mass reached Fukuoka.



Supplemental Figure 1-3. Wind speed during type S episode (a) averaged over whole episode, and (b) averaged 3 h before the air mass reached Fukuoka.