Interactive comment on “Nitrate transboundary heavy pollution over East Asia in winter” by Syuichi Itahashi et al.

Anonymous Referee #2

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This paper focuses on the nitrate transboundary heavy pollution over East Asia in winter. They developed a technique that could differentiate the nitrogen or sulfur dominated air pollution based on a regional chemical transport model and surface observations. They also highlighted the importance of the transboundary air pollution dominated by nitrate, which may refine our understanding of the transboundary heavy PM2.5 pollution in winter over East Asia. Overall, this represents an important work to document the sources-transport-deposition of air pollutants in a hotspot region. I recommend it to be accepted but with revision, to allow the authors to address my concerns below. Also, they could provide, if possible further analysis from another episode to see how comparable/consistent their results could be, especially for type nitrate episode.

Page 2, Line 18: The conversion of gas to particle is not the only way producing nitrate in the air. Besides, coarse particles like Gobi desert surface soil also involved in the nitrate production during the transport [Atmos. Chem. Phys., 14, 11571-11585].

Page 3: It is better to introduce “why the observation period from January 7–17, 2015 was selected”.

Page 4, Line 12: The citation (Pan et al., 2016) is not identical throughout the text. It is better to use (Pan X. et al., 2016) here, to avoid mixing from another EST paper by Pan Y. et al., 2016.

Page 4, Sect. 2.1.1, 2.1.2 and 2.1.3: All the measurements made have biases and in some cases they are significant. It was reported here that fine-mode aerosols were collected with a PTFE filter. It is well documented that positive artifacts of filter sampling are mainly caused by the adsorption of interference gases, such as acidic HNO3 gases, by the collected particles or the sampling filter [Atmospheric Environment 145, 293–298]. It is likely that nitrates have been overestimated due to the sampling filter itself, in addition to the difference in the cut-off diameter [Page 5, Line 1-4].

Page 5, Line 1-4: Is the difference in the cut-off diameter was the only reason for the systematic differences between ACSA and D-F method? It was documented that ultraviolet spectrophotometric method will overestimate the concentration of nitrate. Please also add some details about the comparison here, as most of the readers cannot follow references in Japanese.

Page 5, Line 13-14: It is easy to understand that the nitrate data from Goto Islands, Tsushima Island, and Tottori was not used due to volatilization. In addition to nitrate, ammonium was also affected by volatilization. So, why the data of ammonium was used?

Page 5, Line 5-22: How did PM2.5 measured by PM-712 compare to the Beta attenuation sampler? It is likely that PM2.5 have been overestimated, depending on temperature and relative humidity, and there could have been significant impacts.
Page 5, Line 22: Change the title of 2.1.5 to 2.1.6.

Page 9, Line 10-12: Even that I agree with the results in this study, I am unsure how reproducible those results are on the regular basis, since the authors only study one episode for "type N" and "type S", respectively. Such an issue needs to be critically addressed.

Page 9, Line 25-30: It is hard to believe the concentrations of ammonia are close to zero. As shown in Fig. 6, it seems the temporal variation of the NH₃ observation was mismatched with the ammonia simulation, especially for Type N. In a recent publication, enhanced values of NH₃ were observed within the Asian summer monsoon upper troposphere, where it might contribute to the composition of the Asian tropopause aerosol layer [Atmos. Chem. Phys., 16, 14357-14369].

Page 10, Line 20-21: Why precipitation on Jan 15-16 was not captured in the model [Fig. 4]?

Page 11, Sect. 3.2: Although the long-term range transport of air pollutant was discussed here, the readers are still wondering the contribution of nitrate production during the process/pathway of transport, as well as the contribution from local sources surrounding the investigated sites.

Page 16: The aforementioned contributions are encouraged to be quantified in the conclusion and abstract to highlight the importance of nitrate long-range transport.

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