Authors' Response to Referee #1 Comments

Thank you for the careful review and valuable comments on our manuscript. We address all the referee's comments in the revised manuscript. Here we provide our responses to each comment as follows:

Anonymous Referee #1

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General comments: The measurement of aerosol during the whole year is important especially over East Asia, due to the lack of such continuous monitoring. Owing to the rapid economic growth of East Asian countries, the atmospheric aerosols increase not only over East Asia, but also the outflow region. In fact, transpacific air pollution sometimes arrives at the west coast of the United States. Sapporo is the one of the suitable place to monitor the amount of the outflow from East Asia and Siberia, as mentioned by the authors. Therefore, the in-situ observations are useful as well as satellite obser- vations and aerosol-transport models. One of the most important ionic species among inorganic compounds is sulfate, which is often the most abundant in the atmosphere. Therefore, I (and probably most readers) like to expect the authors to show what is the major source of the sulfate compounds at Sapporo. Unfortunately, I did not find clear answers from the current manuscript, even though I found several independent suggestions. This is the first major comment. I recommend that the authors divide the results into those for the abundant compounds and for the other trace metals. Second, the main conclusions in this study come from the results using the factor analysis, i.e., "varimax rotated factor analysis", as mentioned in the manuscript. However, I cannot find any description of the analysis including a range of the uncertainties. Therefore, I am not convinced the conclusions from the manuscript. I strongly recommend the author to describe the method of the factor analysis in detail in the manuscript. Third, although the authors showed the correlations among various species, each point used in this analysis represents 13-18 days mean value. In Fig. 4, for example, although the authors showed the correlation coefficients in each season, the number of the points in each season is small, with the value of 8 (autumn), 3 (winter), 5 (spring), and 5 (summer). I feel that the time resolution is too coarse to investigate the seasonal variation of the sources. The results during 13-18 days probably include different air masses; therefore at least the authors explain the reason and the validity of the use in the manuscript. Basically, the authors often suggest very important conclusions, but some of them are unclear for the readers. In conclusion, I cannot recommend publishing the manuscript in the current form.

Response: *(i) Based on factor analysis-multiple linear regression (FA-MLR) of ionic species, we found that anthropogenic emissions including biomass burning followed by the subsequent photochemical oxidation is the major source of sulfate in Sapporo aerosols. We also estimated the biogenic fraction of SO_4^{2-} that accounted up to 22% by assuming the $MSA^-/nss\text{-}SO_4^{2-}$ molar ratio is a function of temperature, as reported by Bates et al. (1992) and using measured MSA^-. We include these results and discussion in the revised MS.*

(ii) Following the referee's suggestion, we divided the results of ionic components and trace metals and conducted the FA-MLR for source identification and apportionment. The results and discussion of the FA in ACPD paper will be replaced with those of FA-MLR in the revised manuscript. We provide the description of FA-MLR and the uncertainty in model estimation of each species in the revised MS.

(iii) We collected each sample for about 2-week period in order to obtain the sufficient amount of carbon content for radiocarbon (^{14}C) analysis of organic molecular species, another objective of this research. We include this point in the revised MS. Although the time resolution is not high, the backward air mass trajectories conducted for every 48 hours during each sample period suggest that they were originated from the same source region. This point also be included in the revised MS. In fact, we found a clear seasonal pattern in the percent modern carbon (pMC) and the concentrations

of biomarkers as well as their contributions to organic carbon (OC) and water-soluble organic carbon (WSOC) in the Sapporo aerosols (Pavuluri et al., 2013). Therefore, we believe that the investigation of seasonal sources is reasonable that could provide reliable conclusions, although the number of the samples in each season is small.

Specific comments:

1. P6590, L20-21: This statement is interesting, but I cannot find clear evidence in the manuscript.

Response: *The FA-MLR model estimations showed that the Sapporo inorganic aerosols are influenced by mixed sources and the estimated fraction of biogenic SO_4^{2-} is up to 22% only. Hence, we reconsider our claim and modify this statement in the revised MS.*

2. P6602, L8-12: Although the Sapporo and the ocean are not so far, is the depletion of Cl- through formation during the transport from the ocean?

Response: *The depletion of Cl^- during atmospheric transport is a potential reason. The other possibility is an enhanced dry/wet deposition and/or a potential loss during sampling. However, we remove this point in the revised MS. We also include the discussion on potential sampling artifacts in the revised MS.*

3. P6602, L19-24: This description should be moved to section 2. In addition, more explanation should be inserted, as mentioned in my general comments. Especially, please explain the meaning of the values in Table 2 and clarify the uncertainties of secondary species including SO42-, NO3-, and NH4+.

Response: *We provide the description of FA-MLR model used in this study in the revised MS. We also note the meaning of factor loadings and provide the uncertainties in model estimation of all the species in the revised MS.*

4. P6605, L1-14: The anthropogenic species shown here are secondary aerosols. However, I think a factor analysis generally includes a large uncertainty especially for the secondary aerosols. Originally, how confidant does this result? This comment is related to the above comment #3.

Response: *The uncertainty in model estimation of secondary species (SO_4^{2-}, NO_3^- and NH_4^+) is within 32% and comparable to those reported in the literature (Guo et al., 2004; Rout et al., 2013). Therefore we consider that the performance of the FA-MLR model used in this study is reasonable. We discuss these points in the revised MS.*

5. P6607, L15-18: The correlation coefficients in winter and summer are very low. The values are ranging from 0.05 to 0.12. Statistically, these values represent no relation.

Response: *We remove this statement in the revised MS.*

6. P.6610, L15-16: More evidence is needed. To understand the transport, why does the authors analyze the air mass in detail?

Response: *(i) We reconsider our claim that marine/coastal biota emission of dimethyl sulfide (DMS) is the major source of MSA^- and biogenic fraction of SO_4^{2-} in Sapporo aerosols. However, our results suggest that the emission of DMS from higher plants could possibly contribute for some extent. We include this discussion in the revised MS.*

(ii) Since we collected the samples for about 2-week period each, we preferred to conduct the air mass trajectories for every 48 hours in order to understand that whether the sample was influenced by mixed air masses or not. We note this point in the revised MS.

7. P6622, Fig. 2: For me, the variation of the SO42- is smaller than that of the MSA, even though the authors stated the similarity of the variation.

Response: *Yes, SO_4^{2-} and MSA^- are increased by a factor of about 2 and 3, respectively, but their trends are similar each other. We note this point in the revised MS to avoid any misunderstanding.*

We revise the discussion by interpreting such comparability for mainly photochemistry rather than origins in the revised MS.

Technical comments:

1. P6589, Title: The term 'ionic' can include organic compounds, but they are not treated in this study. Therefore, the term 'ionic' may be suitable. The terms like 'inorganic' will be better.

Response: *We modify the title to better represent the content of the paper by using "inorganic" in the revised MS.*

2. P6590, L13: Table 3 shows 32.47%, which is about 32%.

Response: *We correct such errors in the revised MS.*

3. P6590, L20-21: This study only analyzes inorganic aerosols, therefore the term 'Sapporo inorganic aerosols' rather than 'Sapporo aerosols' is preferred.

Response: *Based on the comment, we modify this phrase in the revised MS.*

4. P6624, Fig. 4: If the authors like to add some of r2 value in the panels, please make a table to show the all of r2. The randomness to show the r2 value in the panels is not good.

Response: *Following the referee's suggestion, we provide the r^2 values for all the species in the Table in the revised MS.*

References

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