Responses to Anonymous Referee #2

We thank the reviewer for the insightful and valuable comments. Our specific responses are addressed below and colored by blue. Changes made to the manuscript are in quotation marks.

General comments: This manuscript reports the measurements of characterization of severe haze episodes and the influences of long-range transport in the Seoul metropolitan area using a high-resolution aerosol mass spectrometer (HR-AMS) and PMF analysis. The authors identified seven organic aerosol (OA) factors, including a HOA, a COA, a SFOA, two LO-OOAs, and two MO-OOAs. Their results present that nitrate was the major component of PM1 and the source of nitrate was originated from regional transport. The Pb was identified by the HR-AMS measurement and was associated with the long-range transport of polluted areas during haze events. This manuscript presents lots of scientific results and figures to support the findings of the regional transport-influenced. However, some parts of the AMS measurement results seem to be too detailed and not concisely presented. In general, this manuscript presents lots of scientific results and data analysis, and it's publishable on Atmospheric Chemistry and Physics with a major revision. Some concerns and comments are listed.

Specific comments:

1. The authors present 7-factor solution in this PMF analysis for this study. However, three primary organic aerosol (POA) related factors and four secondary organic aerosol (SOA) factors could lead to a confusion to readers. From the Figure 2(d-q) and Figure S6(d), the results show the R values in mass spectra for 11 pairs of factors are greater than 0.8. The 7-factor solution has 11 pairs of factors with similar mass spectra. Have you evaluated the combination of similar factors and conduct correlation analysis with tracers? The factor 3 and factor 4 share similar time series profiles. The 6-factor solution still has a similar time series of mass spectra of factor 2, factor 3, and factor 4. The solid-fuel OA(SFOA) is a new factor in this study but has similar mass spectra with previous BBOA. When you create a new factor, which is different from previous studies, you should provide strong evidence to support the naming of a new factor. The SFOA and LO-OOA1 are both burning influenced (from Line 338-339). If you combine both factors, will you see the correlations with biomass burning tracers?

The two LO-OOA and two MO-OOA factors have similar mass spectra and time series profile, which could lead to an unclear result to most readers. In line 408-409, the authors also claimed that the MO-OOA1 has secondary features but was influenced by burning sources, which was lined to SFOA. In this study, you have three factors with burning influenced factors (SFOA, LO-OOA1, and MO-OOA1). Besides, from Table 1, the LO-OOA2 and MO-OOA2 have similar correlation results. Both of them are regional transport-influenced and correlate highly or moderately with nitrate, ammonium, CHN fragments, MSA fragments, and Pb. I suggest that the LO-OOA2 and MO-OOA2 factors can be combined and rename the factor as regional transport-influenced OOA or any specific name for this important source.

Thank you for the comments. We selected the 7 factor solution based on extensive and systematic evaluation of the PMF solutions varying from 2 – 9 factors, especially from 6-8 factors. While it is true that the number of SOA factors we resolved is higher than the typical number reported in other AMS field studies, each of the 4 SOA factors identified in this study shows distinct features that give us confidence about their validity. First of all, MO-OOA1 and LO-OOA1 are very different than MO-OOA2 and LO-OOA2. Both MO-OOA1 and LO-OOA1 are only distinguishable during certain periods rather than over the entire study. Specifically, LO-OOA1 is more pronounced during haze period and MO-OOA1 is more pronounced during burning period. For MO-OOA2 and LO-OOA2, while it is true that their time series are similar, their mass spectra are very different (O/C 1.11 vs 0.65, N/C, 0.084 vs 0.017). As the reviewer suggested, we evaluated the correlations between the sum of MO-OOA2 and LO-OOA2 and SOA tracer ions and the correlation coefficients have now been included in Table 1. The combined time series indeed showed a better correlation with SOA tracer ions. Since they both represent regionally transported SOA, it is possible that the emission source regions for both types of SOA are similar but their processes in the atmosphere are different. Because of lack of supporting data, we weren't able to specify what exactly those processes are. However, given LO-OOA2's high N/C ratio and good correlation with MSA, we hypothesize that chemical processing occurring after long-range transport might have some influence on the feature of LO-OOA2. Thus, we chose to keep those factors separated for the future investigation. We referenced the sum of LO-OOA2 and MO-OOA2 as RSOA (regional transport influenced SOA) in case we need to clarify the discussions.

For SFOA, we didn't specify typical tracer for SFOA, however, we mentioned that it showed the mixture feature of BBOA and CCOA. Indeed it showed the tracer of BBOA (m/z 60, 73) and C COA (m/z 115, PAH), thus we call them as SFOA. Hence, triangle plot show the difference betw een the previous BBOA and the current SFOA. To clarify it, in the revised version, we further sh ow the mass spec up to 120 to show the signal at 115.

Furthermore, in this study, we identify the two burning related factors – MO-OOA1 and SFOA. We found that their combined time series showed better correlations with biomass burning tracer ions such as $C_2H_4O_2^+$ (r = 0.90) and $C_3H_5O_2^+$ (r = 0.86), but had less well correlations with coal burning tracers (e.g. PAH (r = 0.51) and $C_9H_5^+$ (r = 0.70) compared to the correlation only with SFOA (e.g., PAH vs SFOA (0.75), $C_9H_5^+$ (r = 0.77)). The correlation coefficients have now been added in Table 1. Since $C_2H_4O_2^+$ and $C_3H_5O_2^+$ may enhance slightly in coal-combustion OA, to a significantly lesser degree than in BBOA, their better correlations with the sum of MO-OOA1 and SFOA than with the individual factors support the associations of MO-OOA1 and SFOA with burning related activities. The lower correlation between MO-OOA1 + SFOA vs. $C_9H_5^+$ (r = 0.7) and vs PAH (vs 0.51) than between SFOA vs. $C_9H_5^+$ (r = 0.77) and SFOA vs. $C_9H_5^+$ (r = 0.75) supports the separation of SFOA and MO-OOA1 with SFOA more closely related to coal combustion or PAH emitting burning sources. MO-OOA1 is only distinguishable during certain period rather than over the entire study. This suggest that coal and biomass burning combined burning activities generally emit as a form of SFOA and $C_2H_4O_2^+$ and $C_3H_5O_2^+$ strongly emitted burning source (e.g., BBOA) seem to influence during certain period as a form of MO-OOA1. We have rewritten the relevant section to make these points clear.

"The SFOA were found to be another important POA source (7% of the total PM, Fig. 1c) in the SMA in March in addition to vehicle and cooking emissions. The mass spectrum of the SFOA showed typical features of biomass burning OA (BBOA), with dominant peaks at *m/z* = 60 and 73 and strong signals of oxygenated ions ($C_xH_yO_1^+$: 34.7% of the total SFOA signal; $C_xH_yO_2^+$: 14.5% of the total SFOA signal) (Fig. S15). Also, it showed the intense peak of the typical feature of coal combustion OA (CCOA) at *m/z* =115 (mainly $C_9H_7^+$), showing a mixed characteristics of biomass burning and other fuel burning, not pure biomass burning OA. Indeed, the time series of the SFOA correlated with biomass burning tracers, i.e., $C_2H_4O_2^+$ (r = 0.85), $C_3H_5O_2^+$ (r =0.74) potassium (r = 0.63), the CHN family of ions such as $C_2H_5N^+$ (r = 0.59) and $C_3H_7N^+$ (r = 0.70) and BC (r = 0.82), but also it exhibited a good correlation with Pb (r = 0.60), PAH (r = 0.75) and alkyl fragments ($C_nH^+_{2n+1}$ and $C_nH^+_{2n-1}$), including $C_9H_7^+$ (r = 0.81), which were likely emitted from other burning activities, such as fossil fuel combustion (Hu et al., 2013) (Table 1). Hence, when SFOA is combined with MO-OOA1, a SOA influenced by burning event, the correlations with biomass burning tracers were enhanced (e.g., $C_2H_4O_2^+$ (r = 0.90), $C_3H_5O_2^+$ (r = 0.86,) whereas the correlations with coal burning tracers were decreased (e.g. PAH (r = 0.51) and C_9H_5 (r = 0.70), implying that biomass burning OA is probably separated into SFOA and MO-OOA1 and coal burning is significantly impacting on SFOA, another evidence of the mixture feature of SFOA during this study. Furthermore, the scatter plots of *f*44 versus *f*60 indicate high *f*60 and low *f*44 values (i.e., toward the center of the triangular area of the biomass burning plumes) with increasing relative importance of biomass burning to the total OA (Fig. S13). The *f*44 and *f*60 values of the SFOA in this study were much higher than the values of the COA and HOA; in contrast, the *f*60 values of SFOA were somewhat lower than the previous BBOA values observed in the SMA (Kim et al., 2017), further verifying that the SFOA are influenced by the impacts of other burning activities such as pulverized coal combustion (Wang et al., 2013). Furthermore, BBOA is typically prevalent at locations where wood is used for residential heating (Crippa et al., 2013; Ge et al., 2012a; Young et al., 2016); however, residential wood burning is not the main heating source in the SMA. For these reasons, this factor was indicated as part of the SFOA and not purely BBOA. Given that the polar plot of the SFOA revealed high concentrations at both low and high WSs (Fig. S17), the sources of the SFOA in the SMA likely include both local and regional burning activities. The local burning activities possibly occurred for the purposes of open and public area heating (e.g., construction areas and markets), disposal of leaves and woody trash in the city, and residential heating, which can include all types of burning. The regional sources of the SFOA are possibly the open biomass burning activities in the agricultural areas near Seoul (Heo et al., 2009) and the transport emissions from North Korea or farther away from Mongolia (Jung et al., 2016), where biomass and coal burning is a major heating source during the cold season (Batmunkh et al., 2013; Jung et al., 2010). Indeed, back-trajectory analysis indicated a high fraction of the SFOA in the plumes originating from the north, including North Korea and the Mongolian area (Fig. 1d). The more oxidized features than those of the BBOA observed in the SMA (O/C ratio, i.e., 0.53 vs 0.34 (Kim et al., 2017) further supports that there is some influence of regional transport (Fig. 2f)."

2. Line 338-339: “The SFOA and LO-OOA1 contribute 32.9 and 29.7%, respectively, to the C2H4O2 + (m/z = 60) and C3H5O2 + (m/z = 73) signals (Fig. S16).” However, Table 1 shows the r is -0.10 between LO-OOA1 and C3H5O2+. Please explain it.

Thank you for the comments. LO-OOA1 was the typo and it was supposed to be MO-OOA1 which is burning related sources and also showed the good correlation with $C_2H_4O_2$ and $C_3H_5O_2$. Fig. S16 also showed that both SFOA and MO-OOA1 mainly contributed not LO-OOA1. That part has been corrected and moved to the section when we discussed about the MO-OOA1 and that section reads;

“Indeed, the MO-OOA1 contribute 29.7 and 26.5 %, respectively, to the biomass burning signal of $C_2H_4O_2^+$ (*m/z* = 60) and $C_3H_5O_2^+$ (*m/z* = 73) sharing with SFOA (32.9 and 26.6 %, respectively) (Fig. S16).”

3. From Figure 6 during the EP3-S3, we can see the strong wind and higher fraction of MO-OOA1. What’s the reason for the high MO-OOA1? Is the burning source from local or remote region? In line 514, “the enhancement of the burning-related sources (SFOA and MO-OOA1) was observed”, which didn’t show the enhancement of SFOA from Figure 6.

Thank you for the comments. Figure 6 show the enhancement of both SFOA and MO-OOA1 although MO-OOA1 enhancement looks more significant. The differences from other enhancement is that, MO-OOA1 enhanced only this time whereas SFOA enhanced several other times as well. Thus we suspect that there’s different types of plums than rest of other enhanced cases. We didn’t specify the local or remote burning sources. Instead we mentioned that the MO-OOA1 and SFOAs appeared to be intrinsically linked since, diurnal patterns of both MO-OOA1 and SFOA appeared similar during the high-loading period, but a small afternoon peak of the MO-OOA1 was observed (discussed in section 3.2.1), suggesting that that the MO-OOA1 is the SOA formed by the impacts of the burning activities on March 22. This discussion indirectly suggest that MO-OOA1 is the SOA of burning source of remote region. In order to clarify this, the revised version reads;

“ ~during this period, with strong winds from the north, enhancement of the burning-related source, i.e., MO-OOA1 (section3.2.1) was observed, suggesting that the aged burning plums in remote region might influence during this period.”

4. Line 531-535: Do you have evidence of planetary boundary layer height diurnal pattern from modeling result or from previous studies to support your hypothesis? Ask Benjamin

We do not have measurement and/or shared data but there were several papers to discuss about the enhanced boundary layer during daytime in Seoul, which are;

Lee, H., Jo, H., Kim, S., Park, M., Kim, C. : Impacts of atmospheric vertical structures on transboundary aerosol transport from China to South Korea. *Sci Rep* **9,** 13040, https://doi.org/10.1038/s41598-019-49691-z, 2019.

Lee, J., Hong, J., Lee, K., Hong, J., Velasco, E., Lim, Y.J., Lee, J.B., Nam, K, Park, J.: Ceilometer Monitoring of Boundary-Layer Height and Its Application in Evaluating the

Dilution Effect on Air Pollution. *Boundary-Layer Meteorol* **172,** 435–455, https://doi.org/10.1007/s10546-019-00452-5, 2019.

We add those references here.

5. Line 533: ". . ..due to the evaporative of semivolatile species at high air temperature. . .", this sentence is not explained clearly. Do you mean the HNO3 loss leads to the nitrate decrease?
Nitrate is the volatile species, thus it can be evaporated in the high air temperature. HNO3 also could be volatilized but unfortunately, we don't have measurement. Since we are discussing this based on the diurnal profile of NO3, for the clarification, we revised this sentence to;
Decrease trend → Decrease trend of NO_3~

6. Most main figures have multiple figures, which did not present the most important result but showed many detailed figures in the main figures. I suggest that the main figure just shows the most important figure and move other detailed figures to supplemental figures. For example, in Figure 3(a) is the main figure, and the Figure 3(b-e) can be moved to supplemental material.
Thanks for the suggestions. In the current revised version, all the figures of resolution is highly enhanced. Also font size was increased. Also Fig. 3(b-e) has moved to supplementary.

Technical corrections:
1. Figure 2(c) is semivolatile oxygenated OA(SV-OOA), which is not mentioned in the main text.
Thanks, it has been removed.

2. Figure 4 (b) : "PM1 gaseous species" should be "PM1 mass species" in the caption.
Thanks, it has been corrected.

3. Figure S3 is blurry. Please replace the figure with a higher resolution figure.
Thanks, it has been replaced.

4. Figure S3 on p.8 (line 127) should be Figure S4. This figure is not clear but it presents the m/z 103, 103.5 and 104, which is not mentioned in the main text.
Thanks, figure numbering and figure resolution has been replaced. For the explanation in the main text, m/z 103, 103.5 and 104 was mentioned in the main text such as ;
"The signals corresponding to the ions of the other main lead isotopes ($^{207}Pb^{+}$ and $^{206}Pb^{+}$) (Fig. S3), as well as to the doubly charged ions of the three main lead isotopes ($^{208}Pb^{++}$, $^{207}Pb^{++}$ and $^{206}Pb^{++}$), were also observed (Fig. S4). "
For the clarification, the figure caption of Figure S4 has been updated;
"2.5 minute averaged open V mode mass spectra at (a) m/z 103, (b) m/z 103.5 and (c) m/z 104 for $^{208}Pb^{++}$, $^{207}Pb^{++}$ and $^{206}Pb^{++}$, during Haze period at KIST site.~"

5. Figure S5 is blurry. Please replace the figure with a higher resolution figure.
Thanks for the suggestions. In the current revised version, all the figures of resolution is highly enhanced. Also font size was increased.

6. Figure S8 is blurry. Please replace the figure with a higher resolution figure.

Thanks for the suggestions. In the current revised version, all the figures of resolution is highly enhanced. Also font size was increased.

7. Figure S10 is blurry. Please replace the figure with a higher resolution figure.
Thanks for the suggestions. In the current revised version, all the figures of resolution is highly enhanced. Also font size was increased.

8. Figure S17 is blurry. Please replace the figure with a higher resolution figure.
Thanks for the suggestions. In the current revised version, all the figures of resolution is highly enhanced. Also font size was increased.

9. Table S3 line 58 (Figs. Sx and x): Please label the Figure number.
Thanks, it has been revised.

10. Table 1 : CH2SO2 + (79) should be CH2SO2 + (78).
Thanks, it has been revised.

11. Table 2: “BBOA” should be “SFOA”.
Thanks, it has been revised.

12. Figure 6: “BBOA” should be “SFOA”.
Thanks, it has been revised.

13. The name of episodes should be consistent in the main text and figures. For example, the EP3_S1, and EP3_S2, EP3_S3 are labeled in Figure 6, but in Line 505 and 507 they are S3. Line 264 : Ep1, Ep2, and Ep3 should be EP1, EP2, and EP3.
Thanks for the comments. Ep1, Ep2, and Ep3 in line 264 has been replaced with EP1, EP2, and EP3.
The S1, S2, S3 in line505-507 explained what those indicate for. For example, for EP1-S1 shown in figure, the line 505-507 guide how to interpret S1. It designate the event of haze episode_stage of episode. For the clarification, we add following sentence;
“In each figure and relevant discussions, haze stage denote followed by the haze event, i.e., EP1-S1, EP1-S2, etc.”

14. Line 193: HRMS should be HR-AMS.
Thanks, it has been revised.

15. Line 508: RSOA is not defined.
Thanks, it was defined as regional transport-influenced SOAs (RSOA) at line 382.

16. Line 539 : NOR is not defined.
Thanks, it has been defined as nitrate oxidation ratio.

17. Line 553 : SOR is not defined.
Thanks, it has been defined as sulfate oxidation ratio.

18. Line 969: the “PMA” analysis should be corrected as “PMF” analysis.
Thanks, it has been corrected.

19. Please use a consistent mass unit (ug m-3 or ug/m3) throughout the main text, figures, and tables.
Thanks, it has been revised throughout the manuscript.