Responses to Anonymous Referee #1

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 measurement report titled "Characterization of severe spring haze episodes and influences of long-range transport in the Seoul metropolitan area in March 2019" submitted by Hwajin Kim, Qi Zhang, and Yele Sun reports on aerosol characteristics in Seoul Korea during haze events in early spring with comparisons to similar measurements taken simultaneously in Beijing China. The manuscript meets the requirements of a measurement report, i.e., it reports substantial new results from field measurements with conclusions of more limited scope than in research articles. The authors utilized measurements from the high resolution aerosol mass spectrometer (HR-AMS), meteorological measurements, other aerosol and gas phase measurements, and various analysis techniques to arrive at the main conclusion of the report, that long-range transport is a substantial (but not main) contributor to severe haze episodes in the Seoul metropolitan area. The overall quality of the manuscript is good, although some areas will need improvement. I recommend that the manuscript be published after making the following revisions.

Specific Comments:

Line 248: You mention a poor correlation between RH and nitrogen oxidation ratio (NOR), but you do not say what NOR (or later SOR) describe. You could say that NOR is the molar fraction of particle nitrate (NO3-) to particle nitrate plus gaseous NO2. Or, you could show an equation and cite a reference. It would also be nice for you to explain why we are looking at the correlation between RH and NOR. What is the typical trend for regional transport or local oxidation? What does good correlation between RH and NOR mean? Same questions for SOR.

Thanks for the suggestions. As reviewer suggested, we describe the NOR and SOR and also add the equations to calculate them. We are looking at the correlation of NOR and SOR with RH since RH can be a key factor to facilitate nitrate and sulfate formation, although many other factors (e.g., pH and T) may play a role as well. The correlations between RH and NOR (or SOR) thus suggest the possibility of the relevant reactions, not the exact evidence. With the suggestions of the reviewer, we rewrite the relevant section, which now reads;

"The relationships of nitrate and/or nitrogen oxidation ratio (NOR, molar fraction of NO3 in total N, i.e., NO3 + NO_2) and sulfate and/or sulfate oxidation ratio (SOR, molar fraction of SO_4 in total S, i.e., SO_4 + SO_2) with RH were often analyzed to understand the role of heterogeneous aqueous phase formation of secondary inorganic aerosols (Sun et al., 2013; Li et al., 2017; Xu et al., 2014). Both NO_3 and NOR showed an increasing trend with RH, but in a rather scattered relationship (i.e., r = 0.40). The correlation between RH and NOR is also positive but relatively weak (r = 0.48)). These results suggest that heterogeneous aqueous-phase processing likely contributed to some degree to the formation of inorganic nitrate and sulfate during this period (Fig. S9)."

In terms of the typical trend for regional transport or local oxidation, there are no specific numbers for each cases. For nitrate, generally, the concentration of its precursor (i.e., NO2) depends on local sources (although some are possibly from regional sources). Thus if NOR and NO3 enhanced, we can "suspect" an enhanced regional impact of nitrate. For sulfate, SO2 is mostly from regional sources and SO4 formation primarily occurs on a regional scale with some local contributions (mostly through aqueous phase reaction). Thus even if there is a significant regional impact, it is possible that SOR wouldn't enhance (since both sulfate and SO2 enhance). We've already discussed about it in section "3.3.3 Diurnal patterns of the PM1 composition during haze".

Line 257: What is your source for the assumption that PM1 represents approximately 80% of the PM2.5 mass?

Thanks, Mention the following reference to explain the assumptions.

Lim, S., Lee, M., Lee, G., Kim, S., Yoon, S., and Kang, K.: Ionic and carbonaceous compositions of PM10, PM2.5 and PM1.0 at Gosan ABC Superstation and their ratios as source signature, Atmospheric Chemistry and Physics, 12, 2007-2024, 10.5194/acp-12-2007-2012, 2012

Lines 293-294: You mention that nitrogen-to-carbon (N/C) ratio gradually increased overnight until 10:00 and indicated that both primary and secondary factors might influence the N/C ratio. Perhaps the increase of N/C ratio over the night is due to nighttime reactions of amines with nitrate radical, which has been observed in previous wintertime studies (Silva et al., 2008; Chen et al., 2016) and has been shown to increase during fog events (Chen et al., 2016). This might be worth mentioning in the manuscript.

Chen, C.; Chen, S.; Russell, L. M.; Liu, J.; Price, D. J.; Betha, R.; Sanchez, K.; Lee, A. K. Y.; Williams, L.; Collier, S. C.; Zhang, Q.; Kumar, A.; Kleeman, M.; Zhang, X.; Cappa, C. D. Organic aerosol particle chemical properties associated with residential burning and fog in wintertime San Joaquin Valley (Fresno) and with vehicle and firework emissions in summertime South Coast Air Basin (Fontana). J. Geophys. Res. - Atmos., 2018, 123, DOI:10.1029/2018JD028374

Silva, P. J.; Erupe, M. E.; Price, D.; Elias, J.; Malloy, Q. G. J.; Li, Q.; Warren, B.;Cocker III, D. R. Trimethylamine as precursor to secondary organic aerosol formation via nitrate radical reaction in the atmosphere. Environ. Sci. Technol. 2008, 42, 4689-4696. DOI:10.1021/es703016v

Good suggestions. Enhanced N/C ratio during night could be due to amines formed by nitrate radical reaction and/or fog processing. Although we do not have other evidence other than higher N/C ratio, we can suggest the possible night time reaction. Also RH during night was higher than during daytime. Now the relevant section reads;

"Or this enhanced N/C ratio overnight is possibly due to the night time reactions of amines with nitrate radical (Silva et al., 2008; Chen et al., 2016) which can be more increased during fog events (Chen et al., 2016). High night time RH during this study (Fig. 7) further suggest the possibility of night time reaction of nitrogen containing species. More investigations will be needed to confirm this night time processes in SMA."

Line 372: You say, “However, the mass spectrum of the LO-OOA2 is somewhat different from the less oxidized mass spectrum.” Do you mean the less oxidized mass spectrum determined by Sun et al., 2014? If so, place the reference at the end of the sentence.

It was meant that the LO-OOA2 is somewhat different from the mass spectrum of ROSA (sun et al., 2014). Relevant section has been clarified;

“However, the mass spectrum of the LO-OOA2 (Fig. 2n and o) is somewhat different from the mass spectrum of RSOA (Sun et al., 2014).”

Line 374: Which haze episode is being referred to in this sentence? Is it an average of all haze events? Please specify.

Yes, it was an average of all haze events. It has been specified as below;

“Compared to the clean period, the averaged RSOA (LO-OOA2+MO-OOA2) concentration during the three haze episodes had increased by a factor of ~8 (Figs. 2, 5, 6 and Table 2) ~”

Lines 411-412: You state, “Furthermore, we also observed that the evolution of the MO-OOA1 and SFOAs appeared to by intrinsically linked.” Upon first reading, it took me a while to realize that the following three sentences described how this was observed. I suggest making the following changes to the next sentence to clarify this better: Change “Overall, both diurnal patterns . . .” to “Overall, the diurnal patterns of both MO-OOA1 and SFOA . . .” in line 412.

Thanks. We make this clear in the paragraph and now reads;
“Overall, diurnal patterns of both MO-OOA1 and SFOA appeared similar during the high-loading period~”

Line 427: You report the range of aerosol concentration increases from low- to high loading periods of all aerosol components and OA sources as 1.7 – 8.6 and reference Table 2 and Figures 4 and S11. However, the upper range of 8.6 is not shown in any of the references. According to Table 2, the high value is 10.7. Please check these numbers.

Thanks, the numbers are from the value of RSOA (averaged value of LO-OOA2 + MO-OOA2). Since we didn’t use that value in both table and figure, the relevant section has been corrected;

“the average concentrations of all aerosol components and sources of the OA were 1.31 – 10.7 times higher during the high-loading periods than those during the low-loading periods (Table 2, Figs. 4 and S11). ”

Lines 436-438: In comparing the enhancement factors for the regional transport factors LO-OOA2 and MO-OOA2 with the local source factors HOA and COA, you state that the enhancements are considerably higher for the regional transport factors. This may be true for LO-OOA2 (10.7), but MO-OOA2 (5.4) is similar to HOA and COA (5.2 and 4.7, respectively). They are both higher, but only LO-OOA2 is considerably higher (in my opinion).

Yes, as reviewer mentioned, both HOA and COA are also higher and it is possibly due to lower concentration during clean period since both showed the higher ratio of overall/low loading period, shown in Table 2. But higher ratios are also possible due to there are some effect of local accumulation which we also consider as a possible reason and mention about this in section 3.3.4. For the clarification, follow sentence has been added;

"Note that HOA and COA, which are locally emitted OA also showed the enhancement with a factor of 5.2, and 4.7, respectively, indicating that not only regional impacts but also local accumulation might impact on the haze episode. However, relatively higher ratio between overall- and low-loading periods (Table2) suggest that lower concentration during clean period also possibly enhance the ratio between high- and low-loading periods. Details on the haze evolution will be discussed in section 2."

Line 599: You state that, "The HOA, COA, and LO-OOA1 did not contribute to Pb."

However, figure 8 shows a significant contribution to Pb from LO-OOA1. Am I reading the figure correctly? It is difficult to tell because the text in the figure is very small and the figure itself is of low resolution (see my comments in the technical corrections section on figure resolution and readability) and the OOA colors are so similar.

Thank you for the comments. First, Figure is revised to make the text easier to read. According to linear regressions, the variability of Pb concentration is explained strongly by the variations of LO-OOA1. However, the linear correlation between Pb and LO-OOA1 was somewhat low (r = 0.24). In order to make this point clear, we have rewritten the relevant section;

"The contribution of the Pb sources analyzed using a linear decomposition algorithm further showed that the airborne Pb measured at SMA can be freshly emitted from burning sources and transported in aged air masses along with other unknown species. Figure 8 reveals that a major source mixed with Pb was the SFOA (40%). The other mass fractions of the Pb-associated OA sources were MO-OOA2 (22%), LO-OOA1 (17%), and LO-OOA2 (13%). In particular, the contributions of the MO-OOA2 and LO-OOA2 were greatly enhanced during the high-Pb period, which is also consistent with the haze periods, i.e., March 5-7, March 11-12 and March 19-21, thus further supporting that haze formation was indeed impacted by regional transport. Note that overall contribution of LO-OOA1 is higher than that of LO-OOA2 (Fig. 8e), suggesting that there could be another source of Pb in local scale in SMA. As discussed in previous, LO-OOA1 is the locally formed SOA mostly enhanced during the first haze episode, thus it showed the significant impacts on the Pb concentration during the first haze episodes together with MO-OOA2 and LO-OOA2. However, the fraction of LO-OOA1 is high during the low loading period as well resulting the third highest significance to Pb (Fig. 8e) although the correlation with Pb is low (r = 0.24). This might be because the variation of Pb, specially the high concentration of Pb is mostly caused by the regional transport related with MO-OOA2 (r=0.62) and LO-OOA2 (r=0.73) (Table 1). The HOA and COA rarely contribute to Pb. These results suggested that 40% of the Pb-containing

particles in the SMA originated from combustion sources of the OA, whereas the rest was associated with aged and transported sources of the OA (RSOA) and locally formed SOA. ”

Technical Corrections: There were a number of minor technical corrections in both the main text and supplement, as well as some important corrections for the figures and tables.

Thanks. We have looked through all main text and supplement and did the technical correction. Also figures and tables has been updated as suggested.

Main Text:
Line 20: Change “organic mass-to-carbon” to “organic mass to organic carbon”
Thanks, it has been corrected.

Line 33: Change “AMS” to “HR-AMS”
Thanks, it has been changed throughout the manuscript.

Line 38: Add a comma after “haze period” and place “was” between “local burning” and “also important”
Thanks, it has been corrected.

Line 60: Change “KORUR-AQ” to “KORUS-AQ”
Thanks, it has been corrected.
Line 110: Change “Fig. 1” to “Fig. 1a”
Thanks, it has been corrected.

Line 121: Change “9 h earlier than” to “9 h ahead of” and “1 h earlier than” to “1 h behind”
Thanks, it has been corrected.

Line 124: Change “AMS” to “HR-AMS”
Thanks, it has been changed throughout the manuscript.

Line 125: Remove space between “HR-“ and “AMS”
Thanks, it has been corrected.

Line 136: Change “Fig. S1a” to “Fig. S1c”
Thanks, it has been corrected.

Lines 137-145: Change all five instances of “AMS” to “HR-AMS”
Thanks, it has been changed throughout the manuscript.

Line 139: Change “Figs. 1c and S1d” to “Figs. S1c and S1d”
Thanks, it has been corrected.

Line 141: Change both instances of “size distribution” with “concentration”
Thanks, it has been corrected.

Lines 168-170: Change both instances of “AMS” to “HR-AMS”
Thanks, it has been changed throughout the manuscript.

Line 188: Change “would be” to “is”
Thanks, it has been corrected.

Line 228: Add “ m” after “691”
Thanks, it has been corrected.

Line 228: What is the starting height for cluster 2? Add that value to the sentence.
The starting height for cluster 2 was the same with cluster 1.
The relevant sentence has been clarified;
“The average starting height for the back trajectories over the entire study period was approximately 225m for clusters 1, 2 and 691 m for cluster 3, respectively”

Line 228: Change “Fig. 1” to “Fig. 1d”
Thanks, it has been corrected.

Line 230: Add “(Fig. S19)” after “Beijing” at the end of the sentence.
Thanks, it has been added.

Line 232: Add “(Fig. S17)” after “species” at the end of the sentence.
Thanks, it has been added.

Line 240: Change “Ox” to “O3”. Note: Figure S8 in the SI is illegible, but the caption mentions an O3 time series.
Thanks, it has been changed and figure has been replaced with the updated one.

Line 249: Change “Fig. S8” to “Fig. S9”
Thanks, it has been changed.

Line 259: Change “AMS” to “HR-AMS”
Thanks, it has been changed throughout the manuscript.

Lines 260-262: Change sentence to say, “Using a global standard, 70% of the days (30 days) violated the WHO daily PM2.5 standard (25 ug/m3), thus indicating how significant the haze was during the measurement period.”
Thanks, it has been changed.

Line 278: Add a superscripted “-1” after “ppbv”
Thanks, it has been added.

Line 278: Rearrange parentheses to read, “The moderate correlation of the daytime (10:00~16:00) SOA/Ox ratio (r=0.60, 0.19 ug m-3 ppbv-1) suggests . . .”

Line 301: Change “Fig. 2” to “Fig. 2b”
Thanks, it has been changed.

Line 307: Vehicle emission control measures are not marked in Figure 5 as indicated.
The Figure 5 is to compare the time series of chemical species from SMA and Beijing during the haze period, thus it seems to be complex to mark the emission control measures, thus Fig. 5 has been erased from the text and only the period was mentioned in the text.

Line 322: Remove “s” from “winters”, and add “spring of” before “2016”. Also, winter of 2015 is not shown in either Fig. 2d or Fig. S14.
Thanks, it has been corrected.

Line 324: You say “during the 2016 winter”, do you mean “during the 2015 winter”?
That measurement has been done from 2015 Dec to 2016 Jan. For the clarification, the relevant sentence corrected to “2015~2016 winter (Kim et al., 2018)~”

Line 325: Change “Fig. S12” to “Fig. S15”
Thanks, it has been changed.

Line 371: Move “(Fig. 2g)” to just after “MO-OOA2” in line 370.
Thanks, it has been moved.

Line 373: Place “(Fig. 2n and o)” after “LO-OOA2”
Thanks, it has been moved.

Line 375: Change “Fig. 2n and o” to “Fig. 2p and q”
Thanks, it has been changed.

Line 376: Add “during the haze episode” after “RSOA concentration”
Thanks, it has been added.

Line 387: Add an “s” to the end of “fragment” and move the superscripted “+” to after the subscripted “2n+1” (and “2n-1”) in the parentheses.
Thanks, it has been corrected.

Lines 397-398: “Diurnal Patterns” should be placed third in the list (after “mass spectra” and “time variations”) because the diurnal patterns are shown in Figure 7 only.
Thanks, it has been corrected

Line 402: Place “the following” before “typical features.”
Thanks, it has been corrected

Line 404: Remove “the” before “photochemical production”
Thanks, it has been removed.

Line 408: According to Figure 2q (MO-OOA1 factor time series) there is a large concentration spike on March 22, not March 23 as indicated in the text. Please check these dates. Similar comment for line 415.
Thanks, it has been corrected.

Line 428: Remove ", respectively" from the parentheses.
Thanks, it has been removed.

Line 437: Remove the "s" from the end of "enhancements"
Thanks, it has been removed.

Line 438: Change "(Figs. 4 and S11, respectively)" to "(5.2 and 4.7, respectively) (Figs. 4 and S11)"
Thanks, it has been changed.

Line 440: Change "Fig. S8" to "Fig. S9"
Thanks, it has been changed.

Line 458: Remove the "s" from "Figs. 4" and remove ", respectively" from the parentheses
Thanks, it has been removed.

Line 484: Replace "high" with "large"
Thanks, it has been replaced.

Lines 506-507: The dates mentioned in this sentence do not seem to match the dates in the axis of figure 6. Please check / correct these dates.
Thanks, it has been corrected. Now the sentence read;
"The PM concentration in Beijing started to increase on March 18 when no haze was observed in the SMA (S1), while the SMA haze started to intensify starting on March 19 12:00 (S2)."

Line 526: Replace "selected based on" with "highlighted in"
Thanks, it has been replaced.

Line 539: Remove "the" before "RH"
Thanks, it has been removed.

Line 543: Remove ", respectively" from the parentheses
Thanks, it has been removed.

Line 553: Change "Fig. 7b" to "Fig. 7". Note: there are no alphabet labels in figure 7.
Thanks, it has been changed.

Line 554: Place "(Fig. S10)" after "SO4 formation" at the end of the sentence.
Thanks, it has been changed.

Lines 558-560: These two sentences are redundant. I suggest removing the second sentence.

Thanks, it has been removed.

Line 585: Change “5-7 March, 11-13” to “March 5-7,”
Thanks, it has been changed.

Line 596: Add the percent contributions to Pb of MO-OOA2 and LO-OOA2 in parentheses just after they are mentioned.
Thanks, it has been added.

Line 612: Add an “s” to the end of “event” and replace “was strongly” with “were strongly”
Thanks, it has been added and replaced.

Line 616: Replace “decreased” with “shifted”
Thanks, it has been replaced.

Line 617: Change “AMS” to “HR-AMS”
Thanks, it has been changed.

Lines 621-622: Replace “has investigated” with “have been investigated”; change “AMS” to “HR-AMS”; change “2015 Dec.” to “December 2015”; replace “transport” with “transported”; and replace “features” with “NO3”.
Thanks, it has been all changed.

Lines 624-625: Rearrange sentence to read, “Due to the current emission control policies in China, SO4 and SO2 did not considerably change during the haze period compared to the low loading period.”
Thanks, it has been rearranged.

Tables and Figures: A general comment about figures. All figures in the manuscript, including the supplement, NEED to be of a high resolution (DPI 300 or greater). All font NEEDS to be large enough to be read easily (e.g., Time New Roman 10 or greater). Some of the figures in your main text (notably Figure 8 and the pie charts in other figures) were too low resolution with small text. Almost all of the figures in the supplement were too blurry (with text too small), which rendered them illegible (e.g., Figs S8 and S17). IMPORTANT: The main and supplement figures will need to be corrected before the paper is published.
Thanks for the suggestions. In the current revised version, all the figures of resolution is highly enhanced. Also font size was increased.

Table 1: Replace “K (AMS)” with “K (HR-AMS)”
Thanks, it has been changed.

Figure 1: Add “HYSPLIT” before “clusters” in section (d) of the caption.
Thanks, it has been changed.

Figure 4: In section (b) of caption, add a "," after "PM1" and place ", ratios," between "meteorological parameters" and "and tracers". Also, what do the dotted lines mean in 4b?
Thanks, it has been changed. Also the dotted lines are for the guiding eye. Follow sentence has been added in figure caption;
"The dotted lines in Fig. 1(b) are guide for the eye."

Figure 5: In the legend, change "BBOA" to "SFOA"
Thanks, it has been changed.

Figure 6: In the legend, change "BBOA" to "SFOA"
Thanks, it has been changed.

Figure 7: In the last sentence of the caption, add "the dashed line indicates the" before "high-loading periods".
Thanks, it has been added.

Figure 8: In section (e) of the caption, replace "components" with "factors"
Thanks, it has been changed.

Supplement:

Line 57: Add a space between "Expected" and "(deLeater"
Thanks, it has been corrected.

Line 58: Replace "(Figs. Sx and x)" with the figures you meant to reference.
Thanks, it has been removed.
Table 3: Replace "Natual" with "Natural" in table heading.
Thanks, it has been corrected.

Line 120: Replace "HRAMS" with "HR-AMS"
Thanks, it has been corrected.

Line 122: Remove "s" from end of "individuals"
Thanks, it has been corrected.

Figure S3: What do A, B, and C correspond to in the caption? The colors don't match the description in the caption.
A, B and C correspond to (a) m/z 206, (b) m/z 207 and (c) m/z 208 for $^{206}Pb^{+}$,$^{207}Pb^{+}$ and $^{208}Pb^{+}$.
More descriptions has been added to Figure captions and color description also has been corrected.

Line 127: Change "Figure S3" to "Figure S4"
Thanks, it has been corrected.

Line 128: Replace "HRAMS" with "HR-AMS"
Thanks, it has been corrected.

Line 130: Remove “s” from end of “individuals”
Thanks, it has been corrected.

Figure S4: Same issues as figure S3.
A, B and C correspond to (a) m/z 103, (b) m/z 103.5 and (c) m/z 104 for $^{208}Pb^{++}$, $^{207}Pb^{++}$ and $^{206}Pb^{++}$. More descriptions has been added to Figure captions and color description also has been corrected.

Figure S5: Replace “(red), closed” with “(red) and closed”; replace “(terquid)” with “(turquoise)”; bold the “(c)”.
Thanks, it has been corrected.

Figure S9: Replace “NOR” with “nitrogen oxidation ratio (NOR)”
Thanks, it has been corrected.

Figure S10: Replace “SOR” with “sulfur oxidation ratio (SOR)”
Thanks, it has been corrected.

Figure S11: Add label to figure with the names of the different pie chart components.
Thanks, the legend has been corrected.

Line 311: Replace “colored by the time of the day” with “colored by date”
Thanks, it has been replaced

Lines 315-316: In several places, the subtraction sign “-“ was accidentally subscripted. Need to remove subscripting.
Thanks, it has been corrected.

Line 317: Remove “the” before “comparison”
Thanks, it has been removed.

Line 354: Remove the “8” and replace “families” with “family”
Thanks, it has been removed.

Figure S17: Illegible figure. Cannot read the axes. Increase font size and improve figure resolution.
Thanks, it has been replaced with new updated figure.

Figure S19: Add more description to the caption. What are the different lines in the figure?

We add detailed figure caption;

Forward trajectory from Beijing measurement site. Each vertical and horizontal blue dotted line indicate longitude and latitude, respectively. Each black point indicate the endpoint of air parcel movement during 12 h.

Figure S20: Label images with the EP#s and S#s.
Thanks, the image has been labled and relevant figure caption has been added;
“Corresponded haze episode and stages are labeled left top of the figure”

Line 417: Change “Figure 21” to “Figure S21”
Thanks, it has been corrected.

Figure S21: the [NO2] times solar radiation data is not shown in the figure, but it is mentioned in the caption. Also, add “rad” to the legend.
Thanks the figure captions has been corrected and now reads;
“Figure 21. One-hour averaged diurnal profiles for nitrate and various parameters and proxies for formation pathways in (a-c) entire period (d-f) low loading period and (g-i) high loading period during 2019 spring. One-hour averaged diurnal profiles of NO_2, NO_3, NOR (nitrate oxidation ratio) are shown in top row; $[NO_2][O_3]$ as a proxy for nighttime formation of HNO3, RH and one-hour averaged O_3 are shown in middle row; and KAN as the equilibrium constant for gas-to-particle partitioning for ammonium nitrate and solar radiation are shown at the bottom row.”

Figure S22: Replace “nitrate” with “sulfate” in the caption, and subscript the 2 and 4 in “H2SO4”
Thanks, it has been changed.

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.