

Interactive comment on “Measurement report: Characterization of severe spring haze episodes and influences of long-range transport in the Seoul metropolitan area in March 2019” by Hwajin Kim et al.

Anonymous Referee #1

Received and published: 15 July 2020

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 spectrome-

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ter (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 (NO_3^-) to particle nitrate plus gaseous NO_2 . 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.

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

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 winter-time 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.*,

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

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.

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

Lines 411-412: You state, “Furthermore, we also observed that the evolution of the MO-OOA1 and SFOAs appeared to be 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.

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.

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).

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.

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.

Main Text:

Line 20: Change “organic mass-to-carbon” to “organic mass to organic carbon”

Line 33: Change “AMS” to “HR-AMS”

Line 38: Add a comma after “haze period” and place “was” between “local burning” and “also important”

Line 60: Change “KORUR-AQ” to “KORUS-AQ”

Line 110: Change “Fig. 1” to “Fig. 1a”

Line 121: Change “9 h earlier than” to “9 h ahead of” and “1 h earlier than” to “1 h behind”

Line 124: Change “AMS” to “HR-AMS”

Line 125: Remove space between “HR-“ and “AMS”

Line 136: Change “Fig. S1a” to “Fig. S1c”

Lines 137-145: Change all five instances of “AMS” to “HR-AMS”

Line 139: Change “Figs. 1c and S1d” to “Figs. S1c and S1d”

Line 141: Change both instances of “size distribution” with “concentration”

Lines 168-170: Change both instances of “AMS” to “HR-AMS”

Line 188: Change “would be” to “is”

Line 228: Add “ m” after “691”

Line 228: What is the starting height for cluster 2? Add that value to the sentence.

Line 228: Change “Fig. 1” to “Fig. 1d”

Line 230: Add “(Fig. S19)” after “Beijing” at the end of the sentence.

Line 232: Add “(Fig. S17)” after “species” at the end of the sentence.

Line 240: Change “Ox” to “O3”. Note: Figure S8 in the SI is illegible, but the caption mentions an O3 time series.

Line 249: Change “Fig. S8” to “Fig. S9”

Line 259: Change “AMS” to “HR-AMS”

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.”

Line 278: Add a superscripted “-1” after “ppbv”

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

Line 301: Change “Fig. 2” to “Fig. 2b”

Line 307: Vehicle emission control measures are not marked in Figure 5 as indicated.

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.

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Line 324: You say “during the 2016 winter”, do you mean “during the 2015 winter”?

Line 325: Change “Fig. S12” to “Fig. S15”

Line 371: Move “(Fig. 2g)” to just after “MO-OOA2” in line 370.

Line 373: Place “(Fig. 2n and o)” after “LO-OOA2”

Line 375: Change “Fig. 2n and o” to “Fig. 2p and q”

Line 376: Add “during the haze episode” after “RSOA concentration”

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.

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.

Line 402: Place “the following” before “typical features.”

Line 404: Remove “the” before “photochemical production”

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.

Line 428: Remove “, respectively” from the parentheses.

Line 437: Remove the “s” from the end of “enhancements”

Line 438: Change “(Figs. 4 and S11, respectively)” to “(5.2 and 4.7, respectively) (Figs. 4 and S11)”

Line 440: Change “Fig. S8” to “Fig. S9”

Line 458: Remove the “s” from “Figs. 4” and remove “, respectively” from the parentheses

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Line 484: Replace “high” with “large”

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.

Line 526: Replace “selected based on” with “highlighted in”

Line 539: Remove “the” before “RH”

Line 543: Remove “, respectively” from the parentheses

Line 553: Change “Fig. 7b” to “Fig. 7”. Note: there are no alphabet labels in figure 7.

Line 554: Place “(Fig. S10)” after “SO₄ formation” at the end of the sentence.

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

Line 585: Change “5-7 March, 11-13” to “March 5-7,”

Line 596: Add the percent contributions to Pb of MO-OOA₂ and LO-OOA₂ in parentheses just after they are mentioned.

Line 612: Add an “s” to the end of “event” and replace “was strongly” with “were strongly”

Line 616: Replace “decreased” with “shifted”

Line 617: Change “AMS” to “HR-AMS”

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”.

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.”

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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.

Table 1: Replace “K (AMS)” with “K (HR-AMS)”

Figure 1: Add “HYSPLIT” before “clusters” in section (d) of the caption.

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?

Figure 5: In the legend, change “BBOA” to “SFOA”

Figure 6: In the legend, change “BBOA” to “SFOA”

Figure 7: In the last sentence of the caption, add “the dashed line indicates the” before “high-loading periods”.

Figure 8: In section (e) of the caption, replace “components” with “factors”

Supplement:

Line 57: Add a space between “Expected” and “(deLeater”

Line 58: Replace “(Figs. Sx and x)” with the figures you meant to reference.

Table 3: Replace “Natual” with “Natural” in table heading.

Line 120: Replace “HRAMS” with “HR-AMS”

Line 122: Remove “s” from end of “individuals”

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Figure S3: What do A, B, and C correspond to in the caption? The colors don't match the description in the caption.

Line 127: Change "Figure S3" to "Figure S4"

Line 128: Replace "HRRAMS" with "HR-AMS"

Line 130: Remove "s" from end of "individuals"

Figure S4: Same issues as figure S3.

Figure S5: Replace "(red), closed" with "(red) and closed"; replace "(terquid)" with "(turquoise)"; bold the "(c)".

Figure S9: Replace "NOR" with "nitrogen oxidation ratio (NOR)"

Figure S10: Replace "SOR" with "sulfur oxidation ratio (SOR)"

Figure S11: Add label to figure with the names of the different pie chart components.

Line 311: Replace "colored by the time of the day" with "colored by date"

Lines 315-316: In several places, the subtraction sign "-" was accidentally subscripted. Need to remove subscripting.

Line 317: Remove "the" before "comparison"

Line 354: Remove the "8" and replace "families" with "family"

Figure S17: Illegible figure. Cannot read the axes. Increase font size and improve figure resolution.

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

Figure S20: Label images with the EP#s and S#s.

Line 417: Change "Figure 21" to "Figure S21"

Figure S21: the $[\text{NO}_2]$ times solar radiation data is not shown in the figure, but it is mentioned in the caption. Also, add “rad” to the legend.

Figure S22: Replace “nitrate” with “sulfate” in the caption, and subscript the 2 and 4 in “ H_2SO_4 ”

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-382>, 2020.

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