

## Response to Anonymous Referee #1

The paper by Seo et al. summarizes observations at a background and an urban site (Deokjeok Island and Seoul) during a multi-day haze episode in east Asia. The measurements are focused on aerosol composition and auxiliary gases. Meteorological conditions and weather systems associated with this event are also discussed. The main conclusion of the paper is that in Seoul, both regional transport and local emissions contribute to haze formation when meteorological conditions are favorable. The paper is overall well organized and well written. I support publishing the paper after the following minor comments are addressed:

We appreciate the reviewer for careful reading and helpful comments that improve quality of the manuscript. As indicated in the following point-by-point responses, we have incorporated the reviewer's comments and suggestions into the revised manuscript. We have conducted additional analyses, modified texts, figures, and tables, and added several new figures (Figs. 2 and 5) and references in the revised manuscript. Each response to reviewer colored in blue and changes in the manuscript colored in red.

1. p. 3, L 16: PM<sub>10</sub> was also measured at both sites, correct?

Yes, it was. We used measured PM<sub>10</sub> mass concentrations for (i) comparisons with the PM<sub>10</sub> data from the air quality monitoring network (beta attenuation monitoring) and (ii) comparisons of PM<sub>2.5</sub>/PM<sub>10</sub> ratios between haze and clean periods. We replaced "PM<sub>2.5</sub>" in L16 on p.3 with "PM" in the revised manuscript.

2. p. 4, L7: what precautions were taken to reduce gas-phase artifacts in the OC filter measurements? any back-up filters used? If not, please comment on the possible effects on the reported results.

In this study, we did not use both organic denuder and backup filters in the high-volume air sampler, which help to correct positive and negative OC artifacts. In addition, as the other reviewer pointed out, using the different-volume air samplers for inorganic and carbonaceous species could result in biases in estimation of particle concentrations. Considering these two factors, we added a new paragraph that addresses probable artifacts resulted from the measurement at the end of Sect. 2.1, as follows:

Note that the PM<sub>2.5</sub> sampling conducted in this study could result in artifacts of carbonaceous species. Firstly, we used the high-volume air sampler for carbon analyses, while the total mass and ion concentrations were obtained by the low-volume air sampler. Secondly, we did not employ preceding organic denuder and backup filters in the high-volume air sampler for correction of both positive artifacts (by adsorption of organic vapor) and negative artifacts (by volatilization of semivolatile materials) of measured OC. Although the positive artifacts are thought to be larger than the negative artifacts (Chow et al., 2010; Kim et al., 2016), the high-volume air sampler tends to underestimate particle concentrations (Lagler et al., 2011; Aggarwal et al., 2013) and thus the measurement by the high-volume air sampler may partly reduce such positive artifacts as recently estimated by Kim et al. (2016). However, aging and oxidation properties and source characteristics of measured aerosols can still be altered by potential loss of the semivolatile organic compounds. In this study, therefore, we used organic compounds more for qualitative comparisons between different places and periods rather than for quantitative analysis.

3. p. 4, L 21: what was the total volume of the solvent mixture? Also, rephrase as "...and methanol (3:1; v/v) twice, each for 30 min."

The total volume of the solvent mixture was 100 mL because we conducted ultrasonication of the one filter sample for two times in each of two 50 mL solvent mixture. To clarify the procedure, we modified the sentence as follows:

To identify and measure concentrations of individual organic compounds, one-half of the quartz fiber filter was used and ultrasonicated twice in each of two 50 mL mixtures of dichloromethane and methanol (3:1; v/v) in sequential order, each for 30 min (total volume of the solution: 100 mL).

4. p. 4, L23: Please comment on the possible evaporation artifacts when the samples are heated to 40 C.

We added a sentence to address the unavailability of light compounds that resulted from the probable evaporation in the analytic process, as follows:

This process induces low recovery rates (~70%) of relatively low molecular weight compounds (e.g. *n*-alkanes lighter than C<sub>20</sub> and PAHs lighter than anthracene), and we excluded these compounds from further analyses.

5. p. 7, L 10: Considering the distance between Deokjeok Island and Seoul and the wind speed of 2 m/s during the haze period, it seems there would be a 12 hr transport time for plumes to travel from Deokjeok Island to Seoul. Couldn't secondary formation of aerosols during this time also contribute to higher pollution levels in Seoul compared to Deokjeok?

The "local contribution" that we mentioned in this sentence intrinsically includes both contributions by (i) the local primary emissions and (ii) the secondary formation "at local" from both transported and locally emitted precursor gases. To clarify this, we modified the sentence as follows:

On the other hand, the tendency of the lower mass concentration of each aerosol component in Deokjeok than that in Seoul (Table 1) implies the local contributions of both primary emissions from the SMA and secondary productions from local and transported precursor gases to the long-lasting haze in Seoul.

6. p. 7, L26: I don't think comparing CO and PM<sub>2.5</sub> is correct since CO is a primary pollutant and PM<sub>2.5</sub> is predominantly a secondary pollutant.

We agreed with the reviewer's comment and removed "similar characteristics to total PM<sub>2.5</sub>," from the sentences, as follows:

CO, which is an incomplete combustion product of fossil fuels or biomass/biofuels, is affected by both local emissions in the SMA and the regional transport from China and thus shows higher concentration in Seoul than in Deokjeok and also in the haze period than in the clean period (Table 1).

7. p.7, L30: PM<sub>2.5</sub>/PM<sub>10</sub> values are not significantly different at the two sites, so I would remove the 2nd sentence in section 3.2.3.

We agree with the reviewer's comment. Instead of removing this sentence, we modified it to describe significantly high PM<sub>2.5</sub>/PM<sub>10</sub> values during the haze period, as follows:

The larger PM<sub>2.5</sub>/PM<sub>10</sub> during the haze compared to the clean period in both places (Table 1) suggests more influence of the secondary aerosol formation during the haze period (Irei et al., 2015).

We also deleted the second row (fine mode particles related to PM<sub>2.5</sub>/PM<sub>10</sub>) from Table 3.

8. p.8, L7: Contribution of HNO<sub>3</sub> to NOR is not considered (possibly because the measurements were not available). Since temperature and RH affect partitioning of gas phase nitric acid to aerosols, without HNO<sub>3</sub>, NOR as defined is not that useful. I suggest removing NOR from the discussions in the text and tables.

We agree with the reviewer’s point that contribution of  $\text{HNO}_3$  should be considered when we discuss the oxidation of  $\text{NO}_x$ . We removed every words related to “NOR,” following the reviewer’s suggestion. L7 on p.8 was now changed as follows:

The sulfur oxidation ratio ( $\text{SOR} = n \text{SO}_4^{2-} / [n \text{SO}_4^{2-} + n \text{SO}_2]$ ) ( $n$  refers to the molar concentration) represents the atmospheric conversion of  $\text{SO}_2$  to sulfate aerosol through the oxidation and partitioning (Squizzato et al., 2013).

Also, the “NOR” related rows in Tables 1 and 5, as well as related descriptions in the text were deleted in the revised manuscript.

9. p.8, L10: SIA fraction in Deokjeok is 51%, not 57%.

Thanks for the correction. It was now corrected.

10. p.8, section 3.2.3 and 3.3: Although it’s true that Seoul measurements include contributions from local emissions, I think it’s also important to indicate that differences in boundary layer heights in Seoul vs. Deokjeok can also contribute to some of the observed differences in Fig. 4-5.

The higher concentrations of pollutants in Seoul during the haze could be contributed not only by the more local emissions and secondary formation but also by the shallower boundary layer, if the boundary layer height (BLH) in Seoul was lower than BLH in Deokjeok. However, as shown by Fig. S1, the BLH in Seoul was rather higher than that in Deokjeok during the haze, and thus the contribution of different boundary layer condition in Seoul and Deokjeok to the observed differences in Figs. 4 and 5 (Figs. 6–7 in the revised version) seems to be limited. We added average BLH for haze and clean periods in Deokjeok into Table 1 for additional information.

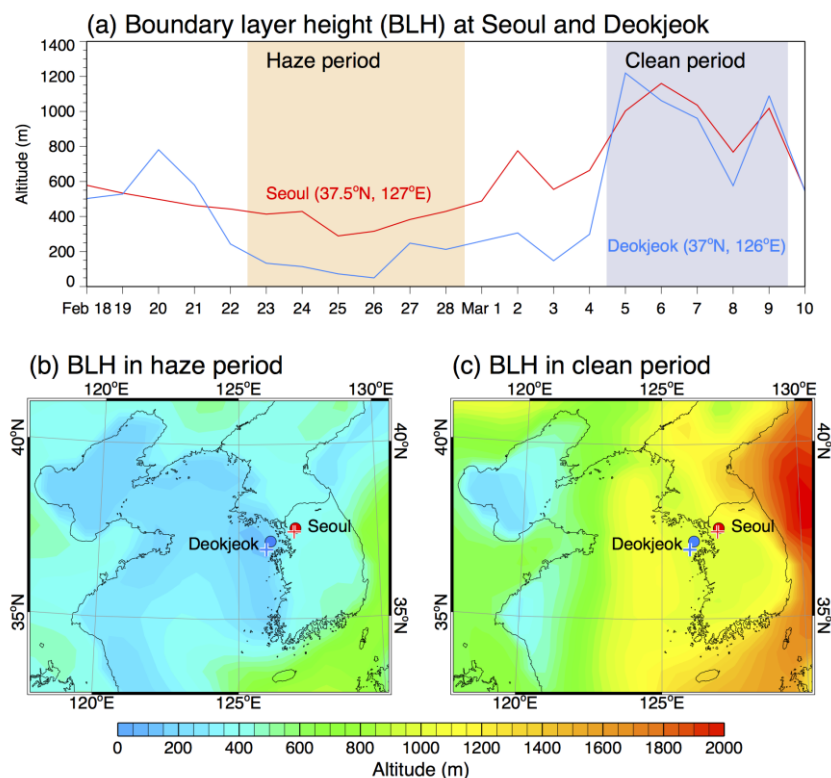


Figure S1: Boundary layer height (BLH) from ERA-Interim reanalysis data. (a) Time series of BLH near Seoul (37.5°N, 127.0°E) and Deokjeok (37.0°N, 126.0°E). (b and c) Spatial distribution of BLH averaged for (b) haze period (February 23–28) and (c) clean period (March 5–9). Filled circles and plus symbols indicate locations of the measurement site and nearest ERA-interim grid points from the sites, respectively.

11. p. 9, L4: what are petrogenic sources specifically and how are they different than petroleum combustion processes?

Petrogenic sources of PAHs are related to spilled or leaked petroleum products (crude oil, fuels, lubricants, etc.), while petroleum combustion is one of the pyrogenic sources of PAHs (fossil fuels combustions or biomass burning) (Stogiannidis and Laane, 2015).

12. p.9, the paragraph on the fractions of PAHs and emission sources is lengthy and at the end, it seems each pair of PAHs suggest one type of source impact. I suggest rephrasing and shortening this section.

To shorten the second paragraph of Sect. 3.3.1, we created a new figure set, Fig. 5 to summarize the PAH ratios from the present study and previously reported PAHs diagnostic ratios with references. Together with Fig. 5, the lengthy paragraph was shortened as follows:

PAHs are combustion byproducts of all types of organic matters, especially related to the incomplete combustion of fossil fuels and biomass burning (Nisbet and LaGoy, 1992; Bi et al., 2003). Various diagnostic ratios with individual PAH species are helpful to search their emission sources (Tobiszewski and Namieśnik, 2012). The average PAH ratios in Seoul and Deokjeok for the haze and clean periods are summarized in Fig. 5 with previously reported diagnostic ratios (Yunker et al., 2002; Pies et al., 2008; De La Torre-Roche et al., 2009; Akyüz and Çabuk, 2010; Oliveira et al., 2011). Interpretation of these PAH ratios all together seems ambiguous and contradictory due to mixing of various emission sources. However, relative comparisons of each ratio between different places and different periods reveal more pyrogenic sources such as fossil fuel combustion and vehicular emissions in Seoul than in Deokjeok (Figs. 5a–c), in the overall influence of coal combustion and/or biomass burning during the haze period (Figs. 5a–b, and 5d).

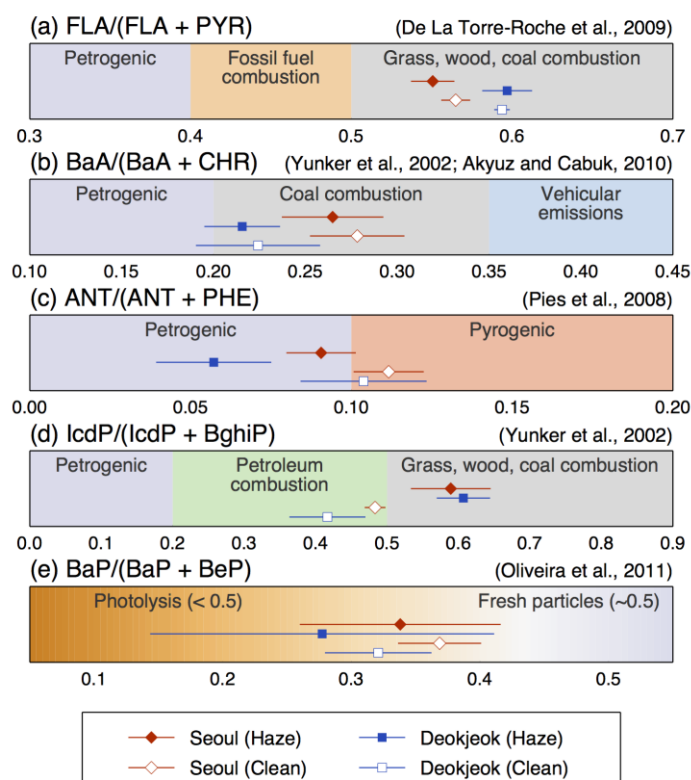


Figure 5: Various PAH ratios of (a)  $FLA/(FLA + PYR)$ , (b)  $BaA/(BaA + CHR)$ , (c)  $ANT/(ANT + PHE)$ , (d)  $IcdP/(IcdP + BghiP)$ , and (e)  $BaP/(BaP + BeP)$  in Seoul (red diamonds) and Deokjeok (blue squares) during the haze (filled symbols) and clean (open symbols) periods. Horizontal bars indicate standard deviation.

13. p. 9, L22: As somebody who's not familiar with seasons in Seoul, it's surprising that measurements in Feb. would indicate biogenic alkane emissions from plant waxes. Aren't trees dormant in Feb in Seoul? Also related to the biogenic alkanes... it seems only C<sub>20</sub>-C<sub>36</sub> alkanes that are found in biogenic emissions are characterized here. This certainly skews the results since midsize semivolatile alkanes are mostly associated with vehicular emissions (see e.g. Gentner et al., EST, 2016). This needs to be addressed.

In South Korea, coniferous forests are about 40% of the total forest area (Lee et al., 2017). Since conifers are not dormant during the cold season, epicuticular wax-related alkanes could be emitted from conifer needles by sloughing and wind abrasion even in winter. In the previous studies on PM<sub>10</sub> organic compounds in Seoul, concentrations of typical biogenic *n*-alkanes (C<sub>29</sub>, C<sub>31</sub>, and C<sub>33</sub>) in winter was comparable to those in other seasons (see Table 1 in Choi et al., 2016).

In terms of the second point, we agree with the reviewer. Sampling and analytic artifacts that commented previously by the reviewer actually resulted in the low recovery rate of light alkanes, and we excluded these compounds (which may associate with vehicular emissions) from the present study.

Thus, we modified and extended the second sentence of the paragraph (L23–24 on p.9) as follows:

Although the measurements in this study were conducted in the cold season, conifers that occupy ~40% of forest in South Korea (Lee et al., 2017) can be a biogenic source of organic compounds. Whereas short-chain (light) *n*-alkanes are mostly associated with incomplete combustion or vehicle exhaust (Gentner et al., 2017), biosynthetic processes result in long-chain high molecular weight (C<sub>27</sub>–C<sub>33</sub>) *n*-alkanes with distinctive odd-to-even carbon number preference (Simoneit, 1991; Rogge et al., 1993). Note that *n*-alkanes in this study reflect less anthropogenic sources because we analyzed only high molecular weight *n*-alkanes (C<sub>20</sub>–C<sub>36</sub>). The carbon preference index (CPI<sub>odd</sub>) defined by a concentration ratio of odd-to-even numbered *n*-alkane homologues is higher than 3 for more biogenic sources while that is close to 1 for more anthropogenic combustion sources (Simoneit, 1989).

14. p. 10, L4: which carboxylic acid can originate as a primary aerosol component from fossil fuel combustion? The low O/C content of POA in many urban environments suggests primary OA does not contain such oxygenated compounds like carboxylic acids. This is again repeated in p. 12, L3-5. Higher acidic components in Seoul would suggest local SOA production rather than contribution from primary emissions.

Although the major source of dicarboxylic acids is believed to be secondary production (Kundu et al., 2010), their primary sources also have been reported by previous studies: motor exhaust (Kawamura and Kaplan, 1987; Grosjean, 1989), wood combustion (Rogge et al., 1998; Oros and Simoneit, 2001), forest biomass burning (Narukawa et al., 1999; Falkovich et al., 2005), and meat cooking (Rogge et al., 1991). For example, Kawamura and Kaplan (1987) reported that the major dicarboxylic acid compounds in vehicular emission except the most abundant one, oxalic acid, were maleic and phthalic acids, which were also measured in this study. Since L4–6 on p.10 is a general description, we didn't remove the text related to primary source of dicarboxylic acids but slightly modified it with additional references, as follows:

Dicarboxylic acids could originate from the primary sources like fossil fuel combustion or biomass burning (Kawamura and Kaplan, 1987; Rogge et al., 1998), but more from the secondary sources like gas-particle partitioning of semivolatile products from the photooxidation of anthropogenic or biogenic precursors and aqueous chemistry in aerosol waters (Rogge et al., 1993; Kundu et al., 2010; Zhang et al., 2010; Zhang et al., 2016).

Also, L3–5 on p.12 is modified as follows:

As briefly mentioned in Sect. 3.3, dicarboxylic acids mainly originate from the secondary sources. Concentration differences for dicarboxylic acids in Seoul and Deokjeok are small during the clean period, but large during the haze period (except on February 26) probably due to influences of local precursor emissions and secondary formation in Seoul (Fig. 7f).

15. p. 11, L18: To fully understand aerosol nitrate formation, some exercise with a thermodynamic model is needed because equilibrium partitioning of  $\text{HNO}_3$  to  $\text{NO}_3^-$  is RH and acidity dependent. Therefore, the high correlation of  $\text{NO}_3^-$  with RH doesn't necessarily mean  $\text{NO}_3^-$  was produced aqueously by uptake of  $\text{N}_2\text{O}_5$ . If the authors mean uptake of  $\text{HNO}_3$  instead (L20) then the process is not aqueous chemistry, but rather shifts in equilibrium partitioning and dissolution of  $\text{HNO}_3$ .

We basically agree with the reviewer's points. However, the term "aqueous chemistry" we used here was actually organonitrate formation by non-radical reactions of glyoxal and  $\text{HNO}_3$  in aerosol liquid water (Lim et al., 2016). Lim et al. (2016) showed that photochemical formation of  $\text{HNO}_3$  through reactions of peroxy radicals,  $\text{NO}_x$ , and OH in gas-phase and  $\text{HNO}_3$  uptake into the aerosol liquid water can enhance the organonitrate formation via reactions of glyoxal and  $\text{HNO}_3$  in aqueous-phase. Organonitrates can also give  $\text{HNO}_3$  by hydrolysis, and aerosol liquid water helps aerosol partitioning of  $\text{HNO}_3$ . Recent study by Lim et al. (manuscript in preparation, 2017) shows the hygroscopic growth and acidification of  $\text{PM}_{2.5}$  in Seoul through the  $\text{HNO}_3$  uptake–organonitrate formation–water uptake cycles, by using the combined kinetic and thermodynamic model simulation with humid chamber experiments and measurement data.

Instead of removing L17–22 on p.11, therefore, we modified the sentences as follows:

The high correlations of RH with nitrate ( $r = 0.53$ ) (Table 5) implies a role of aqueous chemistry in nitrate aerosol formation in Seoul. A recent thermodynamic model simulation with smog chamber experiments and measurement data (Lim et al., in preparation, 2017) reveals that  $\text{NO}_x$  photochemistry under the high RH condition facilitates hygroscopic growth of aerosols through water and  $\text{HNO}_3$  uptake cycles with organonitrate formation in aerosol liquid water (Lim et al., 2016) and thus contributes to the high nitrate proportion to  $\text{PM}_{2.5}$  in Seoul (Fig. 4a).

16. p. 11, L26: it was indicated on L15 (P11) that  $\text{NO}_2$  decreased during 2/27-3/2 with high wind speeds, but that's not similar to the observations in CO that showed little difference with wind speed. Please clarify.

CO in Seoul showed smaller difference with wind speed compared to  $\text{NO}_2$  in Seoul because it is also affected by regional transport as shown as CO in Deokjeok. Therefore, the difference of CO between Seoul and Deokjeok can be more like  $\text{NO}_2$  in Seoul rather than CO in Seoul itself. To clarify this, L25–27 on p.11 was modified as follows:

On the other hand, the concentration differences of CO between Seoul and Deokjeok are large during the haze period but small on the high wind speed days (e.g. February 27 and March 2) like  $\text{NO}_2$  concentrations, and this indicates local influence on the high CO levels in Seoul.

17. p. 11, L31-33: It's unclear why trends in organic tracers suggests regional transport of  $\text{SO}_2$ .

L31–33 on p.11 are now modified as follows:

These are the local characteristics as shown in  $\text{NO}_2$ . Higher concentrations of *n*-alkanes, monocarboxylic acids, and sugars during the haze period than those during the clean period in Deokjeok show the regional transport characteristics as shown in  $\text{SO}_2$ .

18. p. 12, L7: Why is the O:C of carboxylic acids only explored? The conclusions drawn are acceptable if O:C represented values for all components of OA.

We newly calculated O/C for the total identified OM (using analyzed mono- and dicarboxylic acids, sugars, *n*-alkanes, and PAHs all together, of which mass is ~5% of total estimated OM, Fig. 7a) and represented its daily time series in Fig. 7h. Day-to-day variation of the O/C of total identified OM is mostly contributed by that of dicarboxylic acids O/C. On the other hand, small variability of O/C in monocarboxylic acids and sugars (Fig. S2) implies that the sources of these two organic compound groups are mainly primary rather than secondary production.

We modified L7–8 on p.12 as follows:

The O/C of dicarboxylic acids and total identified OM that stayed high during both early and late stages of the haze period in both two places indicate secondary production as a source of the organic compounds during the long-lasting haze (Figs. 7g and 7h).

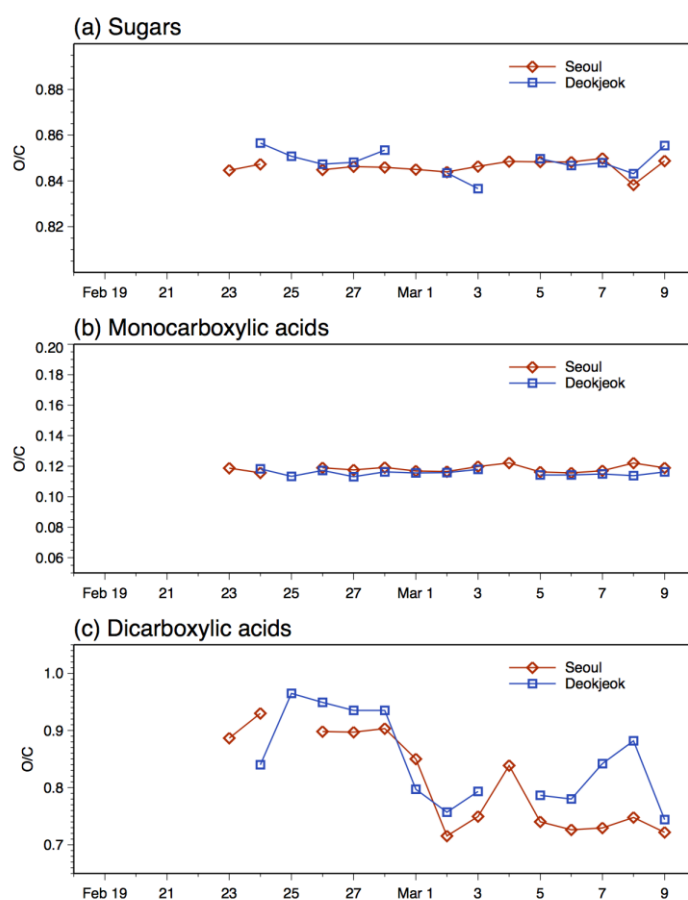


Figure S2: Daily time-series of O/C of (a) sugars, (b) monocarboxylic acids, and (c) dicarboxylic acids at KIST site in Seoul and Deokjeok site for the analysis period.

19. p. 12, L12-13: I disagree with the conclusion here. If RH and OC are not well correlated while RH and OC/EC is, this suggests to me that EC and RH are somehow correlated, but still EC is solely a primary tracer, I think the correlation is merely due to meteorology and cannot suggest anything about aq-phase production of OC.

Total OC can be regarded as the sum of primary OC (POC) and secondary OC (SOC). POC can be represented with a constant intrinsic OC-to-EC ratio of local primary emission ( $[OC/EC]_{pri}$ ) (e.g. Castro et al., 1999) as follows:

$$OC = POC + SOC = [OC/EC]_{pri} \times EC + SOC$$

Thus, the variability of OC/EC represents that of SOC/EC, and correlations of them with RH should be same ( $r = +0.51$  in this study). This value is much higher than correlations of EC (and POC,  $r =$

+0.03), OC ( $r = +0.33$ ), and SOC (approximate SOC calculated using the minimum OC/EC of 2.9 as  $[\text{OC/EC}]_{\text{pri}}$ , see the modified Table 1,  $r = +0.39$ ) with RH.

Weak correlation of EC with RH is natural because EC is a primary tracer, and anthropogenic emissions do not depend on the meteorological condition. The correlation of SOC with RH could also be lower than that of SOC/EC (or OC/EC) because the amount of SOC not only depends on chemistry but also depends on the amount of precursors. Since the precursor emissions could be approximately inferred from the primary pollutant marker, EC, the SOC (or OC) weighted by EC (SOC/EC or OC/EC) shows the degree of secondary production of OC. Therefore, the high correlation between OC/EC and RH shows probable influence of RH on the degree of SOC production. Since O/C (and OM/OC) increase by photochemical processing and secondary organic aerosols (SOA) production (Aiken et al., 2008), and hygroscopicity increases with increasing O/C (Jimenez et al., 2009), the higher correlations of RH with O/C ( $r = +0.54$ ) and OM/OC ( $r = +0.51$ ) compared to that of other meteorological variables (see the modified Table 5) support that aqueous process played a role in the SOA production during the measurement period.

Therefore, instead of removing L12–13 on p.12, we added a sentence at the end of the paragraph as follows:

Hygroscopicity of OA can be enhanced with increasing O/C and OM/OC by SOA production and aging (Aiken et al., 2008; Jimenez et al., 2009). The higher correlations of RH with O/C of total identified OM ( $r = 0.54$ ) and OM/OC ( $r = 0.51$ ) compared to that of other meteorological variables implies hygroscopic properties of SOA produced during the long-lasting haze in Seoul.

## Figures

Fig. 2- please add the sampling site locations to one of the panels.

We marked locations of Seoul and Deokjeok sampling sites in the panels, as the reviewer suggested.

Fig. 3- I understand that OC was the parameter directly measured, but since the pie charts represent total aerosol mass from each species, why not convert OC to OM, using appropriate, representative ratios of OM/OC?

We modified the pie charts as following the referee's suggestion. Average OM concentrations in the pie charts (Fig. 4) were derived from daily measured OC and calculated OM/OC ratios of identified OM (~5% of estimated OM).

Fig. 6- I suggest marking the political boundaries with a different colors than the contours. Also, the wind arrows don't show up well. Consider making the panels larger. It would also help if the locations of the sampling sites are marked on one panel.

We enlarged each panel in Fig. 8 and modified figures following the reviewer's suggestion.

## References

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