

We thank the reviewers for their valuable comments. Our point-to-point responses are provided below and colored by blue. Changes made to the manuscript are in quotation marks.

### **Responses to Anonymous Referee #1**

I thank the authors for considering my comments further. While I would have personally expressed more caution in associating the COA factor with cooking (because it is inconsistent with previous urban studies), at least the caveats are in place now. Similarly with my issue regarding the haze processing, however I notice that the authors copied and pasted some of my review text. While flattered, I would insist that they use their own words here.

We have rewritten the section based on this comment as well as the comments from another reviewer who suggested adding the cases observed in China which also showed the weak diurnal patterns of COA. The revised section now reads;

“As widely reported in highly populated urban cities, a COA factor was resolved in SMA as well. The COA showed good correlations in time series with the key tracer ions commonly used to justify the presence of aerosols from cooking related activities (He et al., 2004; Adhikary et al., 2010; Mohr et al., 2009; Zhao et al., 2007; Ge et al., 2012; Sun et al., 2011; Young et al., 2016; Allan et al., 2010; Huang et al., 2010; Sun et al., 2013; Hayes et al., 2013; Mohr et al., 2012; Dall'Osto et al., 2013), such as  $C_3H_3O^+$  ( $m/z$  55;  $r = 0.75$ ),  $C_3H_5O^+$  ( $m/z$  57;  $r = 0.61$ ),  $C_5H_8O^+$  ( $m/z$  84;  $r = 0.89$ ),  $C_6H_{10}O^+$  ( $m/z$  98;  $r = 0.99$ ) and  $C_7H_{12}O^+$  ( $m/z$  112;  $r = 0.70$ ) (Fig. S18 and Table S2). It was also a major contributor to the signals of  $C_5H_8O^+$ ,  $C_6H_{10}O^+$ , and  $C_7H_{12}O^+$ , accounting for 62%, 94%, and 67%, respectively (Fig. S16). The mass spectrum of the COA determined in this study was almost identical to the COA spectrum determined in winter 2015 – 2016 at the same site (Fig. 8b and S15) (Kim et al., 2017). In addition, as shown in Figure S19b, the ratios between  $f_{55}$  and  $f_{57}$  for OA in Seoul increased proportionally as the fractional contribution of COA to total OA increased, forming a “V” shape with two edges defined by the COA and the HOA factors from several urban AMS data sets (Mohr et al., 2012). Furthermore, the diurnal profile of the COA displayed a large enhancement at evening starting between ~ 18:00 – 19:00, corresponding to dinner time, and a small noon-time peak at ~ 12:00, corresponding to lunch time, although the features do not reflect fully the expected cooking activities. Nevertheless, these observations generally corroborate the association of the COA factor with cooking activities. The relatively weak noon-time enhancement of the COA was probably due to strong dilution of primary emissions caused by convection and elevated boundary-layer height. It is also possibly related to the Korean cooking habits. Previous AMS studies reported COA factors with less pronounced patterns at noon or evening as well (Hayes et al., 2013). In addition, the COA of this study on average accounted for ~ 18% of the total OA mass during lunch time and ~ 37% during dinner time, which are within the values observed for COA in Beijing (Sun et al., 2016 and references therein). Nevertheless, it is important to caution that the noon-time and nighttime peaks could be influenced by photochemical formation of SOA and evening POA emissions coupled with reduced boundary layer height, respectively. In addition, while the COA tracer ions mentioned above tend to represent fatty acids which are usually a significant component of cooking aerosols (He et al., 2004; Mohr et al., 2009; To et al., 2000; Zheng et al., 1997), they can also be contributed by other sources such as plant wax, fossil fuel, soil particles, and SOA (Wang et al., 2006; Zhao et al., 2014). For these reasons, it is possible

that the COA factor identified in this study might contain significant contributions from sources other than cooking activities, similar to situations reported in several other studies (Dall'Osto et al., 2013; Mohr et al., 2009; Mohr et al., 2012; Hayes et al., 2013). Furthermore, a recent study reported that the relative ionization efficiency (RIE) of cooking OA tends to be higher than the typical RIE of 1.4 used for organic aerosol quantification (Reyes-Villegas et al., 2018), suggesting that cooking aerosol concentrations were likely overestimated in this study.

I still find the trailing sentence regarding gas phase processing ("However, observed gradual increase of  $f_{\text{SO}_4}$  could be at least partly due to the gas phase oxidation of  $\text{SO}_2$ ." ) to be completely unsupported by any evidence. Most of the gradual increase during the spring occurs at night, when the OH mechanism cannot occur, so it is simply not plausible here. There is a slight rise before sunset during the winter, but the gradient (I estimate about a fractional increase in excess of 1 per day) is far greater than what would be expected from a reaction with a lifetime of 11 days (less than 0.1 per day, but even this is probably an overestimate). Unless the authors are proposing a different gas phase mechanism, I would suggest they remove this sentence.

We thank the reviewer for spotting the inconsistency and have removed the sentence accordingly. Now the section reads;

"Also,  $f_{\text{SO}_4}$  increased gradually from 11:00 till 6:00 of the next day. For the daytime increase from 11:00 to 18:00, the life time of  $\text{SO}_2$  was calculated to be ~11 days, using the temperature dependent daytime reaction rate constant,  $k$  ( $8.75\text{-}9.24 \times 10^{-13} \text{cm}^3 \text{s}^{-1}$ ) for  $\text{SO}_2 + \text{OH} + \text{M} \rightarrow \text{H}_2\text{SO}_4$  (Burkholder et al., 2015) and corresponded OH concentration ( $1.2 \times 10^6 \text{molecules cm}^{-3}$ ) which is in the range of other urban cities such as Tokyo (Wang et al., 2006) and Beijing (Rao et al., 2016). This suggests that the localized photochemical reaction of  $\text{SO}_2 + \text{OH}$  is not the major process for controlling  $\text{SO}_4^{2-}$  concentration during this period."

## Responses to Anonymous Referee #2

Accepted as is

## Responses to Anonymous Referee #3

I think the authors have addressed the other reviewer's comments and added sufficient discussions and modifications. I have a few more suggestions that I would like the authors to take before the manuscript get published.

( 1 ) For the COA problem, in other places like megacity Beijing, the PMF COA does not always show clear patterns at noon or evening. I think adding discussion about the caveats of lacking strong evidence to support explanation of unique COA is a proper way to solve the problem. However, saying that "The reasons for the small lunch time peak was attributed to strong secondary formation affecting to the peak of COA emissions. And the large and broad nighttime peaks are due to lower boundary layer height" is too strong. The authors need to be careful when stating hypothesis and possible explanations as cause and outcome. Instead, I suggest something like "Reasons of lacking characteristic peaks at noon and evening are perhaps strong dilution at noon due to convection and boundary-layer changes in the evening, which overwhelms the emissions". The authors should also summarize the "range" of previous findings on diurnal profiles of COA to support their statements of large uncertainty remaining in COA diurnal profiles.

Thank you for the good suggestions. We revised our discussions on COA diurnal patterns and added a reference and relevant discussions to discuss about the uncertainties of diurnal patterns of COA. Now the section reads;

"As widely reported in highly populated urban cities, a COA factor was resolved in SMA as well. The COA showed good correlations in time series with the key tracer ions commonly used to justify the presence of aerosols from cooking related activities (He et al., 2004;Adhikary et al., 2010;Mohr et al., 2009;Zhao et al., 2007;Ge et al., 2012;Sun et al., 2011;Young et al., 2016;Allan et al., 2010;Huang et al., 2010;Sun et al., 2013;Hayes et al., 2013;Mohr et al., 2012;Dall'Osto et al., 2013), such as  $C_3H_3O^+$  ( $m/z$  55;  $r = 0.75$ ),  $C_3H_5O^+$  ( $m/z$  57;  $r = 0.61$ ),  $C_5H_8O^+$  ( $m/z$  84;  $r = 0.89$ ),  $C_6H_{10}O^+$  ( $m/z$  98;  $r = 0.99$ ) and  $C_7H_{12}O^+$  ( $m/z$  112;  $r = 0.70$ ) (Fig. S18 and Table S2). It was also a major contributor to the signals of  $C_5H_8O^+$ ,  $C_6H_{10}O^+$ , and  $C_7H_{12}O^+$ , accounting for 62%, 94%, and 67%, respectively (Fig. S16). The mass spectrum of the COA determined in this study was almost identical to the COA spectrum determined in winter 2015 – 2016 at the same site (Fig. 8b and S15) (Kim et al., 2017). In addition, as shown in Figure S19b, the ratios between  $f_{55}$  and  $f_{57}$  for OA in Seoul increased proportionally as the fractional contribution of COA to total OA increased, forming a "V" shape with two edges defined by the COA and the HOA factors from several urban AMS data sets (Mohr et al., 2012). Furthermore, the diurnal profile of the COA displayed a large enhancement at evening starting between ~ 18:00 – 19:00, corresponding to dinner time, and a small noon-time peak at ~ 12:00, corresponding to lunch time, although the features do not reflect fully the expected cooking activities. Nevertheless, these observations generally corroborate the association of the COA factor with cooking activities. The relatively weak noon-time enhancement of the COA was probably due to strong dilution of primary emissions caused by convection and

elevated boundary-layer height. It is also possibly related to the Korean cooking habits. Previous AMS studies reported COA factors with less pronounced patterns at noon or evening as well (Hayes et al., 2013). In addition, the COA of this study on average accounted for ~ 18% of the total OA mass during lunch time and ~ 37% during dinner time, which are within the values observed for COA in Beijing (Sun et al., 2016 and references therein). Nevertheless, it is important to caution that the noon-time and nighttime peaks could be influenced by photochemical formation of SOA and evening POA emissions coupled with reduced boundary layer height, respectively. In addition, while the COA tracer ions mentioned above tend to represent fatty acids which are usually a significant component of cooking aerosols (He et al., 2004; Mohr et al., 2009; To et al., 2000; Zheng et al., 1997), they can also be contributed by other sources such as plant wax, fossil fuel, soil particles, and SOA (Wang et al., 2006; Zhao et al., 2014). For these reasons, it is possible that the COA factor identified in this study might contain significant contributions from sources other than cooking activities, similar to situations reported in several other studies (Dall'Osto et al., 2013; Mohr et al., 2009; Mohr et al., 2012; Hayes et al., 2013). Furthermore, a recent study reported that the relative ionization efficiency (RIE) of cooking OA tends to be higher than the typical RIE of 1.4 used for organic aerosol quantification (Reyes-Villegas et al., 2018), suggesting that cooking aerosol concentrations were likely overestimated in this study.

( 2 ) Please change “bulk neutralized” to something like “bulk balanced in observed cations and anions” when discussing ion balance.

Thank you for the suggestions. Done as suggested

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