

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

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

#### **General comments**

While this paper constitutes part of a large, multinational study, it must be judged on its own merit. The methods employed here can't exactly be described as new and the findings aren't exactly unexpected; most of the results (but not all; see cooking comment below) would come as no surprise to a seasoned AMS user. However, given the diverse nature of pollution in large Asian cities, there is a need to provide a knowledge base for large but relatively poorly characterised cities such as this, so that aspect of its novelty means that this is appropriate for ACP. In addition, from a technical perspective it is also interesting to note the performance of the combined organic-inorganic PMF in this instance; while success in applying this to AMS data has (to this reviewer's knowledge) been inconsistent, it is important to document the cases where this works and most importantly, the authors report the corresponding organic only version in the SI. That said, beyond resolving the contribution of  $m/z=30$  to the organics, it is questionable how dramatic a contribution to the science this makes.

I would question the suitability of ascribing the COA factor to cooking. Normally, when this has been reported in urban areas, it has been accompanied by a very characteristic diurnal profile (e.g. <https://www.atmos-chem-phys.net/10/647/2010/>). However, this is not present here and the reasoning for assigning the factor as such is based on mass spectral similarity alone. However, this particular mass spectral profile can be seen as indicative of monocarboxylic acids generally and there have been studies where the association of this profile with cooking has been questioned (e.g. <http://pubs.acs.org/doi/10.1021/acs.est.5b02922>). Without any further supporting data here, it would be far safer to refer to this as 'COA-like' and caveat the discussion accordingly.

Thank you for the comments. However, the support for the association of the COA factor with cooking activities in this study is quite strong, including having characteristic mass spectrum that is similar to those of cooking OA observed in a large number of studies in urban areas, the good correlations with tracer ions for cooking aerosol, and the excellent agreements between the COA factor of this study and the COA identified in winter at the same site for both mass spectral pattern and diurnal profile. The small lunchtime peak could be explained by atmospheric dilution associated with high boundary layer height and photochemical oxidation around noon time. The evening peak corresponds to dinnertime and the high concentration was likely influenced by reduced dilution due to stagnant nighttime condition (low mixing height and low wind speed). Nevertheless, we agree that the COA factor we observed may be different than POA emitted directly from food cooking activities and thus now refer it as food cooking-influenced organic aerosol when the "COA" acronym is first introduced (see Page 2, line 10 in the revised manuscript). Now the new paragraph on COA reads;

"A COA factor was resolved during this study and showed a mass spectrum almost identical to the COA spectrum determined in winter 2015 – 2016 at the same site (Fig. 8b and S15) (Kim et al., 2017). As shown in Fig. S18 and summarized in Table S2, the key HR-AMS tracer ions for COA,

such as  $C_5H_8O^+$  ( $m/z$  84),  $C_6H_{10}O^+$  ( $m/z$  98) and  $C_7H_{12}O^+$  ( $m/z$  = 112) (Sun et al., 2011), all showed good correlations with COA in time series. The correlation between COA and  $C_6H_{10}O^+$  is particularly good with Pearson's  $r$  values of 0.96 and 94% of the signal in this ion was attributed to COA (Fig. S16). In addition, using the approach reported in Mohr et al. (2012), we examined the ratios between  $f_{55}$  and  $f_{57}$  for POA (i.e., OA subtracted of contributions from LV-OOA and SV-OOA) and found that the ratio increased proportionally as the fractional contribution of COA to total OA increased (Fig. S19b), consistent with the behaviors of COA and HOA reported for several urban AMS data sets (Mohr et al., 2012). The diurnal pattern of the COA factor in this study displayed a large enhancement in the evening around 19:00, due to dinnertime cooking emissions coupled with lower boundary layer height, and a small peak at  $\sim$  12:00 corresponding to lunchtime emissions. These observations confirm the identification of COA in this study. On average, COA accounted for 22% of the OA mass during this study and 20% in winter (Kim et al., 2017), indicating that cooking related activities are an important source of air pollution in the SMA area. This finding is consistent with observations made in a large number of urban locations, where cooking emissions have been frequently identified as a significant contributor to fine particle mass (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)"

Generally, the overall size of the manuscript would be considered too large for the scientific findings that are presented, with some areas (e.g. the peak by peak discussion of the HR mass spectra) I would consider trivial, given the current state of the knowledge. Given what is already known from the preceding decade's worth of AMS papers, it would be better if the authors could focus better on what is genuinely new and different about this work.

In response to this comment, we have shorten the paper and cut the length by  $\sim$ 20%. Major changes were made at section 3.2 Characteristics and source apportionment of organic aerosol organic source, by shortening the discussions on the characteristics of individual OA factors and placed more general discussions in the supporting materials. Also some redundant discussions were removed and Tables 1 and 2 in the original manuscript have been moved to the supplementary.

### Specific comments

1) Page 12, line 15: Given the kinetics of the  $SO_2+OH$  reaction are very well known, does the measured trend in  $f_{SO_4}$  correspond to a reasonable atmospheric OH concentration? If not, then a different mechanism must be responsible.

Thanks. Unfortunately, we do not have OH measurement during this campaign.

In addition, a major source of  $SO_4^{2-}$  observed in SMA is regional transport from the southwest and the diurnal profile of sulfate showed almost no enhancement during daytime. However, a gradual increase of  $f_{SO_4}$  can be observed during daytime, for which gas phase oxidation of  $SO_2$  was likely an important mechanism. It is true though daytime sulfate formation could happen due to mechanisms other than  $SO_2 + OH$ , so we have revised the text so it now reads:

"Also,  $f_{SO_4}$  increased gradually from 11:00 till 6:00 of the next day, which can be explained by daytime photochemical formation of  $H_2SO_4$  (Fig. 4c) ~"

2) Page 12, line 15: Which aqueous-phase oxidation process are the authors referring to? The peroxide pathway (generally thought to be the most important) will not be available at night and while O<sub>3</sub> and NO<sub>2</sub> are possible, they are both pH-limited, so it is questionable whether they are important in haze (as opposed to cloud) droplets.

It is true that SO<sub>2</sub> oxidation by O<sub>3</sub> and NO<sub>2</sub> are both pH-dependent and the reaction rates increase with pH. We did not measure/or calculated the pH of aerosol in this study, however, NH<sub>3</sub> concentration in SMA is likely high since aerosol particles in this region are bulk neutralized (see Fig. S10), suggesting that oxidation by both O<sub>3</sub> and NO<sub>2</sub> could occur actively. In addition, a recent study reported that NO<sub>2</sub> could contribute to sulfate formation in aerosol water under polluted conditions. Thus, by considering elevated PM concentration during nighttime, NO<sub>2</sub> reaction is also possible.

For clarification, sentence below is added;

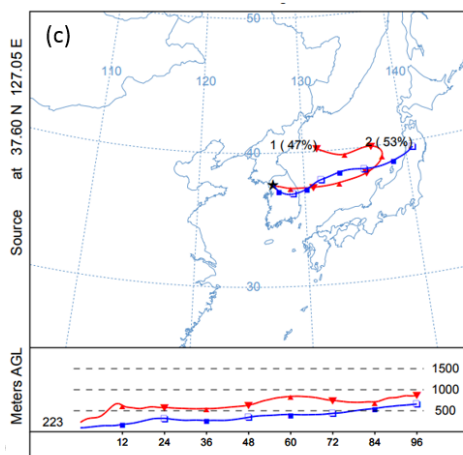
“Possible oxidants during night are NO<sub>2</sub> and O<sub>3</sub> since particles appeared to be bulk neutralized (Fig. S10), where SO<sub>2</sub> oxidation by both oxidants could occur actively (Seinfeld and Pandis, 2006). Furthermore NO<sub>2</sub> has been investigated as an important oxidant in aerosol water under hazy conditions (Cheng et al., 2016).”

3) Table 1: Minimums and maximums are probably not the best statistics to report, as these will capture spikes. 5th and 95th percentiles would probably be better.

Done as suggested

4) Figure 10c: The sheer number of overlaying trajectories mean that this figure is impossible to interpret. Please revise with a smaller number of trajectories.

Thanks for the comment. As we showed at the original version, all trajectories were from the EAST during the investigated period. And we got two clusters, which are all from EAST as shown at the figure below. As reviewer suggested, we have provided trajectories with two clusters of all calculated trajectories during the organic dominant events.



5) Page 15, line 30: The similarity in N:C and O:C diurnal profiles merely tells me that there could be a source of aerosols with a high H:C ratio that is prevalent at night. It would be inappropriate to speculate on secondary mechanisms based on this result.

We agree with the reviewer that the discussions on secondary source of organic nitrogen aerosol is quite speculative. Thus, we removed the related discussions. Now the section reads;

“we found that O/C and OM/OC ratios had similar patterns but the pattern of H/C was different, due to variations in the relative contributions of POA and SOA. Also, nitrogen-to-carbon (N/C) ratios showed a distinct diurnal profile with a bimodal feature peaking at 10:00 and 16:00, similar to the O/C diurnal profile. “

#### **Technical comments**

6) Page 3, line 26: Suggest rephrasing ‘mainland China’ to ‘mainland Asia’. Some of the emissions affecting Korea can originate from other areas (e.g. yellow sand from Mongolia).

“Thanks for the suggestion. Mainland Asia is historically known as Indochina, comprising Vietnam, Laos, Cambodia, Thailand, Myanmar and West Malaysia. Thus, we rephrase the mainland China to Asian Continent which is used in referred paper.”

7) Page 4, line 23: Change ‘specially’ to ‘specifically’  
Done as suggested

8) Page 11, line 12: Change ‘outskirt’ to ‘outskirts’.  
Done as suggested

9) Page 13, line 10: Presumably, the use of the product of NO<sub>2</sub> and O<sub>3</sub> is a measure of the production of NO<sub>3</sub> and in turn, as a proxy for the formation of N<sub>2</sub>O<sub>5</sub> and consequently HNO<sub>3</sub>. This should be explicitly stated because there are a number of other factors at work in this chain that ensure that while it is probably sound as a qualitative measure of nocturnal nitrate production, it is unlikely to be strictly quantitative.

Thanks, the text has been revised and it now reads:

“However, compared to springtime, the product of NO<sub>2</sub> and O<sub>3</sub> ([NO<sub>2</sub>][O<sub>3</sub>]) during winter was ~ a factor of 2 lower during night (Figs. 4a,b), indicating that nighttime nitrate formation is more significant in spring. [NO<sub>2</sub>][O<sub>3</sub>] is a proxy for nighttime formation rate of particulate nitrate, since the reaction between NO<sub>2</sub> and O<sub>3</sub> produces N<sub>2</sub>O<sub>5</sub> and nitrate radical ( $\cdot$ NO<sub>3</sub>), which can react heterogeneously to form HNO<sub>3</sub> and subsequently particulate nitrate (Young et al., 2016). “

10) Page 14, line 4: Should be ‘the southwest’  
The sentence has been corrected accordingly.

11) Figure 4: The figure caption shouldn’t really include a formula. This should be worked into the text that refers to it and numbered accordingly.

Thanks, the figure caption has been corrected accordingly in the revised manuscript. Also, the equation and relevant explanations are moved to pg 13, line 16.