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 ~ 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 ~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 SO2+OH reaction are very well known, does the measured trend in fSO4 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_{SO4} 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 SO2 + OH, so we have revised the text so it now reads:

"Also, f_{SO4} 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 O3 and NO2 are possible, they are both pH-limited, so it is questionable whether they are important in haze (as opposed to cloud) droplets.

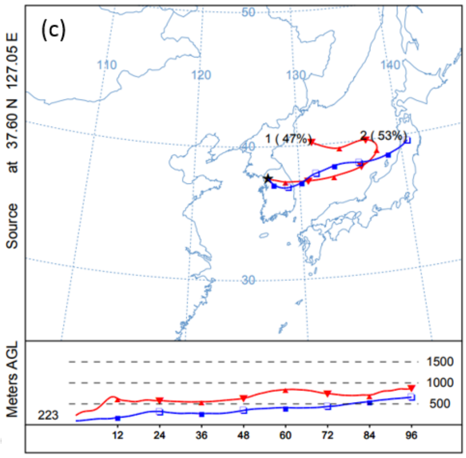
It is true that SO2 oxidation by O3 and NO2 are both pH-dependent and the reaction rates increase with pH. We did not measure/or calculated the pH of aerosol in this study, however, NH3 concentration in SMA is likely high since aerosol particles in this region are bulk neutralized (see Fig. S10), suggesting that oxidation by both O3 and NO2 could occur actively. In addition, a recent study reported that NO2 could contribute to sulfate formation in aerosol water under polluted conditions. Thus, by considering elevated PM concentration during nighttime, NO2 reaction is also possible.
For clarification, sentence below is added;

"Possible oxidants during night are NO_2 and O_3 since particles appeared to be bulk neutralized (Fig. S10), where SO_2 oxidation by both oxidants could occur actively (Seinfeld and Pandis, 2006). Furthermore NO_2 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. Mainlaind 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 NO2 and O3 is a measure of the production of NO3 and in turn, as a proxy for the formation of N2O5 and consequently HNO3. 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_2 and O_3 ($[NO_2][O_3]$) during winter was ~ a factor of 2 lower during night (Figs. 4a,b), indicating that nighttime nitrate formation is more significant in spring. $[NO_2][O_3]$ is a proxy for nighttime formation rate of particulate nitrate, since the reaction between NO_2 and O_3 produces N_2O_5 and nitrate radical ($\cdot NO_3$), which can react heterogeneously to form HNO_3 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.

Responses to Anonymous Referee #2

General comments

This study analyzes the emission sources and SOA formation processes in spring in the Seoul Metropolitan Area (SMA). The field measurement was well planned and a good data set is provided. The authors analyzed the data carefully and showed some interesting findings. However, the paper is too long, and does not reasonably focus on what they really want to know. Therefore, it is probably OK to be published in this journal after appropriate revision according to the following comments.

Major comments

(Overall) The length of the manuscript is better to shorten into half or two-thirds. The information and analyses should be more focused what the authors really want to know. Please clearly state the major findings in the main body and abstract. Less-important information should be shortened or moved to the supporting information. If I understand correctly the motivation of the authors, I recommend changing the constitution of the manuscript as follows. (1) The information written in Section 3.1 and 3.2 seems not critical. So the volume of these sections can be shortened greatly or may be merged into other sections. (2) It may be good to restructure the result section into three: (1) haze event, (2) high organic event, and (3) high sulfate event. Then authors can state the pollution mechanism and sources for each event.

In response to this comment, we have shorten the paper and cut the length by ~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.

(Overall) The authors indicated the importance of SOA on PM mass. It is good if the authors can show where (which region) the precursors (e.g. VOCs and NOx) came from, and the relative importance of ASOA and BSOA. In addition, please clarify the area or area size (and time scale) of “local”, “regional”, and “long-range transport”.

Good suggestions. First, the source regions of precursors appeared to be diverse, including both local emissions and regional transport. We mentioned the approximate locations of precursors in section 2.1 and show them on Fig. 1. Also for the importance of ASOA and BSOA, we think that ASOA might be more important since we did measurement at urban area however, as mentioned at section 2.1., when plum is dominant from East, perhaps BSOA is more important but not clear since no measurement has been done. Also VOC measurement (Fig. 2) doesn’t clearly provide the source whether it is from the anthropogenic or biogenic. Without further supporting measurement/or information, it is too speculative to mention the importance of ASOA and BSOA. Relevant descriptions are below;

“Briefly, KIST is located ~ 400 m from a busy highway and is surrounded by a residential area and a commercial area, thus the air quality at this site tends to be influenced by abundant anthropogenic and primary sources. During spring, KIST, SMA in general, is influenced by highly consistent winds from west and south west (Fig. 1 c, d), where a number of cities and large-scale industrial facilities are located (Fig.

1a) and are significant sources of NO_x and SO_x (Kim et al., 2017). However, sometimes, dominant wind was blown from north and east, where emissions from agricultural and biogenic sources are generally more intense (Fig. S1). “

Finally for the clarification of area size (and time scale) of “local”, “regional”, and “long-range transport”, since we did not model this observation, we does not have exact scale for the above information. However, in this manuscript, when we are mentioning local, the scale is inside of city of Seoul, regional is from outside of Seoul. And long range transport is mainly discussing about the transportation of pollutants from the outside of Korea. For the clarification, relevant discussions are added at section 2.1 as below.

“In this manuscript, pollutants from inside and outside of the SMA are treated as “local” and “regional” scale pollutants, respectively. Air pollution episodes associated with transport from outside of Korea is considered as “long-range transport”.”

(Overall) Recently, in many places especially in cold season, biomass burning is a large source of ambient PM. It would be good to state about how large of the biomass burning in this field campaign.

This campaign happened during spring/or and late spring, which is a warm season. Thus, it is hard to see the impact of biomass burning observing only 4 organic factors; SV-OOA, LV-OOA, HOA, and COA.

(p4,L1-15) The motivation of choosing “spring” as the study season should be clearly stated. It may be good to show average PM levels in four seasons in SMA. Are winter and spring the worst?

This study was performed during spring to investigate the photochemical formation of secondary species (e.g., O3, SOA etc), not because air quality was worse during spring. In this regards, the motivation was discussed in the introduction. In response to the reviewer’s comment, we now briefly overview the seasonal PM concentration and then discuss why this study was done during spring although spring air quality is not the worst. Now the relevant section reads;

“In addition to various emission sources, previous studies have shown that the concentration and the composition of ambient aerosol in SMA are influenced by atmospheric processes and meteorological conditions as well (Heo et al., 2009;Kim et al., 2017). According to measurements by the Seoul Research Institute of Public Health and Environment, $PM_{2.5}$ concentrations in SMA during past 9 years was generally higher during winter (DJF, average ± $1\sigma = 30 \pm 16\ \mu g/m^3$) and spring (MAM; $29 \pm 14 \mu g/m^3$) than in summer (JJA; $23 \pm 13\ \mu g/m^3$) and fall (SON; $23 \pm 14\ \mu g/m^3$). Previous studies have shown that elevated anthropogenic emissions (e.g., from heating) coupled with a lower planetary boundary layer (PBL) height and stagnant meteorological conditions are mainly responsible for poor air quality in Seoul during winter, although long-range transport of pollutants from upwind areas may have some influences as well (Kim et al., 2014;Kim et al., 2017). The severe air quality problem during spring in SMA is frequently driven by long range transport of wind-blown dust (yellow dust) and smokes from fires from the west and northwest (Kim et al., 2010). In addition, compared to winter, photochemical formation of secondary aerosol is expected to be more intense due to increased solar radiation and higher temperature during spring and affects air quality in SMA more actively. However, so far there is little information available on the formation, properties and transport of atmospheric aerosol during spring in SMA, although a fundamental understanding of aerosol chemistry and dynamics is necessary for predicting how changes in atmospheric composition influence air quality in this region."

(p7, L15-17) I am bit curious about the validation method of AMS quantification. Did the authors validated AMS and SMPS quantification accuracy independently? I think the parallel measurement can give us only supporting information.

Thanks, we did the parallel measurement for the AMS and SMPS. For the further validation, we compare our PM1+BC to PM2.5 concentration measured at the closest sampling site. It showed a good correlation (r=0.76) with a slop of 0.67, i.e., on average 67% of the PM2.5 mass is associated with submicron particles. To supporting the measurement, we add this analysis at the supporting material as Fig. S4 with relevant discussions as following;

"In addition, total PM_1 mass (= NR-PM_1 measured by AMS + BC) correlates well with $PM_{2.5}$ mass measured using beta attenuation mass monitor (Thermo, FH62C14) at the Gireum site (~5 km to the west of the KIST site), showing the 67 % of $PM_{2.5}$ (Fig. S4)."

(p11, L2) "haze periods, high organic/or sulfate period". . . These three periods should be shown in Fig.2. Fig.2 is too small and busy. The reader cannot pick-up the information correctly. Fig.11 and Fig.S20 is good. So it may be good to move Fig.S20 to the main body of the manuscript.

Thanks, we mainly discussed two specific events; haze period and high organic period. High sulfate period occurred together with haze period. Fig. 2 is busy but this is the overview of the observation thus, we are trying to keep this figure here by enhancing the quality and showing the event period.

(p17, L31) "The diurnal pattern of COA displayed a large evening peak at ~ 19:00, i.e., dinner time, and a small lunch time peak at ~ 12:00." I cannot see these peaks in Fig.8f. To me, it is highest at around 22:00 and decrease by 18:00.

Yes, as reviewer mentioned, 19:00 does not show the peak, rather enhancement starts from that time when the dinner time start. For the clarification, now the sentence reads;

"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 ~ 12:00 corresponding to lunchtime emissions."

(p20, L20-21) "17:09 to 17:15" It is good if the authors can explain why the SOA increased in the evening (not afternoon). Is the SOA formed in the afternoon then transported? Or formed in the evening?

Thanks. Initial enhancement was driven by the transport which happen together with the enhancement of other species, then remain high due to intensive photochemical formation under stagnant conditions. We make this clear in the paragraph and now reads;

"PM_1 concentration jumped from 11 to 55 µg m^{-3} between 17:00 to 17:45 on May 20, during which concentrations of all PM_1 species (except for COA), SO_2, NO_2, and biogenic and anthropogenic VOCs (e.g., isoprene and toluene) increased sharply (Fig. 2h). As shown in Fig. S20, the onset of this pollution episode was associated with a change of wind direction from southeast to northwest, indicating that it was mainly caused by transport of polluted air masses. Wind speed was low and wind direction alternated between north and east during the next three days, and the concentrations of most air pollutants rose and fell in correlation with the wind shifts. However, LV-OOA remained elevated after the initial sharp rise from 5.6 to 16 µg m^{-3} and increased to a maximum concentration of ~ 25 µg m^{-3} on May 23."

(p23, L11) Please clearly state why you can conclude "spring plumes were long range transported".

The evidences for the "spring plumes were long range transported" are discussed at the third paragraph of the section 3.4, saying that

"On May 24, there was a short clean period (7:30 to 11:30; Period S1) when average PM_1 concentration was only 9 µg m^{-3} due to precipitation. After the precipitation, PM concentration started to increase substantially, accompanied with a change of aerosol composition. During both Period S1 and S2 (May 24, 11:30 – May 26, 18:00), the predominant wind direction was southwest (Fig. 11b). Analyses of the MODIS images (Fig. S21), backtrajectories, (Fig. S22) and meteorological conditions (Discover AQ report, Davis Peterson, NRL) all indicated direct transports of air masses from southwest, where large SO_2 emission sources are located. The change of PM_1 composition during Period S2 reflected the influence from such regional transport processes. For example, the mass fractions of species associated with regional sources, such as sulfate (28 vs 20% during entire period) and LV-OOA (18 vs 15%), increased (Fig. 11l, Table S2), whereas the fractions of local pollutants such as SV-OOA (5 vs 12%), HOA (5 vs 10%), COA (5 vs 7%) and BC (4 vs 7%) decreased compared to averaged PM_1 composition during entire period. In addition, the mass fraction of nitrate, one of the local secondary species, also enhanced (20 vs 17%), and this was mainly due to the gas-particle partitioning of HNO_3 and nighttime heterogeneous reactions in the nitrate formation facilitated by high RH (78%) and low temperature (18 °C) (Table S2). A good correlation (r^2=0.48) between nitrate and RH corroborates the role of aqueous processes (Fig. S23). "

To prevent readers to confuse by this summary paragraph here, the sentence relevant in the summary paragraph has been removed.

(Fig.1) The scales in Fig.1a&b cannot be read. Larger scale map (about 50-100 km size) is better in Fig.1a. Fig.1c,d,e are too small to read. “industrial facilities are located (west and south) and agricultural and biogenic areas (east and south)” cannot be understood from this map.

New figures including larger scale map with lager figure of c,d and e. Including the point sources (e.g., indusrial facilities and biogenic areas) hasn’t described in the figure since that makes figure more crowded. Instead, we describe those information in the text.

(Fig.3) Two of the right bottom figures (Org-EC and HOA-LV-OOA) should be shown in the same style of other figures.
Thank you for the suggestions. However, two of the right bottom figures show the different values than the rest of other figures, e.g. Concentration vs fractions. So we want to keep this format. However for the purpose of clarification, we mention this in the figure caption as follows;

“Two of the figures at the bottom from the right show the fraction of PM_1 mass and organic mass respectively.”

(Fig.8 e,f,g,h) The figure is too busy. BC, NO3, Ozone, etc should be moved (or omitted) to other figures.
Thanks for the suggestions, however, those are useful information to understand the diurnal patterns of each organic factors. Thus we want to keep them on the figure. We will try to enhance the quality of figure.

Minor comments
(Abstract, p2, L1-2) It is better to insert a simple explanation in the begging of the abstract about why the authors selected “spring” as the measurement season.

Thanks for the suggestion. We have already discussed why we did this during spring in the introduction as follow. Since the abstract is already long enough, we are thinking that it isn’t necessary to add this in the abstract again.

“The severe air quality problem during spring in SMA is frequently driven by long range transport of wind-blown dust (yellow dust) and smokes from fires from the west and northwest (Kim et al., 2010). In addition, compared to winter, photochemical formation of secondary aerosol is expected to be more intense due to increased solar radiation and higher temperature during spring and affects air quality in SMA more actively. However, so far there is little information available on the formation, properties and transport of atmospheric aerosol during spring in SMA, although a fundamental understanding of aerosol chemistry and dynamics is necessary for predicting how changes in atmospheric composition influence air quality in this region.”

(p3, L17-19) Doesn’t Korea have environmental quality standard of PM2.5? If they have, the authors should show these values as well here.

The annual air quality standard of PM2.5 in Korea is 25µg m^{-3} and this information is now provided in the manuscript.

(p3, L20-21) The approximate area size (x km2) of SMA should be described.

The approximate size of Seoul is 605.21 Km^2 (approximately 15 km in radius) and this has been added.

(p7, L1, and others) I think the URL information should be moved to the "References" section.
Thanks, it has been corrected.

(p7, L25) "Fig.6a and b" seems not correct figure number.
Thanks, it is the correct figure number but for the clarification. "Evolution pattern" changed to "Diurnal pattern".

(p13, L3-4) ″~20.0 ppb″ should be ″~20 ppb″. ″~41.7 ppb″ should be ″~42 ppb″.
Thanks, it has been corrected.

(p13, L22) ". . .number concentration (Fig. 3),". Fig.8 should be cited here.

Thanks, both figure 3 and 8 are related with this sentence, therefore figure 8 is added as suggested.

(p21, L29) "Fig.12" should be "Fig.11".

Thanks, it has been corrected.

(Table 1) The "0" at the column of "minimum conc" should be shown as "ND". The "-" at the column of "Fraction of total PM" should be shown as "100".

Thanks, it has been corrected.