Responses to Anonymous Referee #1

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

General comments

This manuscript reports results obtained during a field campaign performed at Seoul, South Korea, during 6 weeks in winter 2015/16. The authors deployed an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (hereafter the AMS) and several co-located instruments to measure the particle concentration, chemical composition, and size distribution. The authors performed the usual data treatment, including a source apportionment of organics by positive matrix factorization, a comparison of the different air masses impacting the sampling site with a back trajectory analysis, and a comparison of several selected periods (high vs. low loading). The authors have a very interesting dataset in the hands, and all the information included in the supplementary material shows that the data treatment has been done very carefully. The manuscript is well written, and the content will be of high interest for the readers of Atmospheric Chemistry and Physics. I warmly recommend the publication of this manuscript after the authors address the following comments.

Specific comments

1) Section 2.2 "Measurements": in addition to the five main species, PAHs results are also quickly given later in the Section 3. So I would suggest that the authors mention here that the total PAHs concentrations were also extracted from the organic mass spectra, with eventually some information on the related entries in the fragmentation table.

Following the suggestion by the reviewer, we have added discussions on PAH estimation on Page 7: "Furthermore, the total concentrations of particle-bound polycyclic aromatic hydrocarbons (PAHs) were estimated using the method described in Dzepina et al. (2007). However, instead of apportioning the unit mass resolution (UMR) spectra, PAH-related ions were determined via fitting the high-resolution mass spectra (W-mode) (Xu et al., 2014). In addition, a RIE of 1.35 with respect to nitrate was applied to calculate mass concentrations of PAHs from AMS data (Dzepina et al., 2007)

2) Page 10, lines 19-21: the authors quickly mention that new particle formation events were observed several times during this study. This is an important observation which deserves some additional discussion in the section 3.2 "Size distributions of the main components of PM1" (unless the authors plan to write a separate paper on this topic). In particular, it would be very interesting to use the pToF data of the AMS to identify which chemical species were involved in these new particle growth events.

Thanks for the suggestions. We are indeed interested in understanding what chemical processes were involved in the new particle growth events in Korea and are currently performing detailed analysis using data both from this study and from a spring campaign when new particle events were also observed. We will report our findings in a future publication.

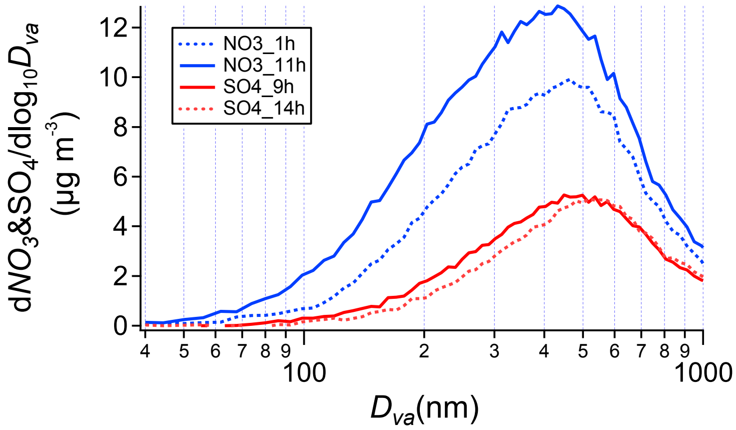
3) Section 3.1.2 "Diurnal patterns of PM1 composition": I think it would be important to include here a discussion on the diurnal pattern of the wind direction. Indeed, according to Figure S11, the wind direction has a clear diurnal pattern, with a wind coming from the south during the day, and from the west during the night. Therefore, diurnal patterns of the different species discussed in this section seem not only driven by the dynamics of the boundary layer height or photochemistry, but also by a shift in the wind direction.

We mentioned the effect of diurnal variation of wind in the text where sulfate diurnal pattern is discussed: "nighttime transport of air mass from industrial facilities located on the west and southwest outskirts of Seoul (Fig. 1b) might be responsible." In response to the review comment, we have now made it clear in the sentence: "for which nighttime transport of air mass from industrial facilities located on the west (Fig. S11) and southwest outskirts of Seoul (Fig. 1b) might be responsible." For other compounds such as nitrate, since they were mainly associated with local sources, we didn't discuss the effect of wind.

4) Page 13, lines 27-30: the authors claim that the size distributions of sulfate stayed fairly constant in concentrations and size distribution. Actually, this is not really the case, given that the concentration dropped quite fast between 9h and 14h (Figure 5b).
I'm also wondering whether there is a discrepancy between the diurnal pattern of sulfate shown in Figure 5b (strong diurnal pattern, varying between 3 and 5 µg/m3) and the hourly averaged size distributions shown in Figure S4b, where the surface areas of the 24 size distributions look very similar (so no diurnal pattern observed here).

The reviewer was right that the average concentration of sulfate did vary somewhat but sulfate size distribution was indeed relatively constant. For the clarifications, the sentence has been revised as follows: "The size distributions of sulfate showed a prevalent droplet accumulation mode (D_{va} = 500 nm) that stayed fairly constant compared to the one of nitrate (Fig. S4b)."

As for reviewer's comment on a discrepancy between the diurnal patterns of sulfate shown in Figure 5b and Figure S4b, the sulfate concentration in the diurnal profile ranges from 2.5 to 3.3 (The figure has been corrected) which is narrower than the one of nitrate (5.9-8.3). As shown in the graph below, the size distribution of sulfate was similar between the times of maximum and minimum concentrations comparing to the one of nitrate, although there are differences at the left hand side of size distribution curves.



Then, they say that the size distributions of nitrate stayed relatively constant throughout the day as well. This is also not the case, given that the size distributions became much broader between 9h and 15h (Figures 5c and S4a).
This part also has been changed to as follows: “the size distributions of nitrate became broader between 9:00 and 15:00 with significant changes in concentrations (Fig. 5c, Fig. S4a).”

5) Page 24, line 17: the authors say that during low loading periods, there was an enhancement of the fractional contribution of BBOA. Actually, according to Figures 12c and 12d, it is mainly LV-OOA which had an important enhancement (from 10 to 20%). In the same time, BBOA increased only from 9 to 12%.
Thanks for the comments. We agree with the reviewer that the highest enhancement occurred with LV-OOA whereas the enhancement of BBOA was smaller. Nevertheless, we consider an enhancement from 9% to 12 % is significant and worthwhile to mention. In response to the reviewer’s comment, we have revised the text to report the actual fractional numbers: “In addition, an enhancement of the fractional contributions of BBOA from the high loading periods (9%) was observed during low loading periods (12%).”

6) Page 24, line 19: Actually, the authors had previously showed the high correlation between PAHs and BBOA, but they did not say anything about the link between PAHs and vehicle emissions. According to Table 2, the correlation between PAHs and HOA is much smaller (r = 0.48, vs. 0.90 between PAHs and BBOA).
We have revised the text to make this point clear: “and that PAHs in PM_1 were mainly from biomass burning (r = 0.90)”

7) Figure 14: clusters 1, 2, and 3 seem to come more or less from the same region in the northwest of the sampling site. It would be interesting to include in the supplementary material four maps (one per cluster) with the details of all the individual back trajectories included in each cluster. These graphs are easily generated by the HYSPLIT software and can show us whether a given cluster is the average of similar back trajectories, or whether back trajectories are completely different but give an average cluster coming from the northwest.

Individual back trajectories in each cluster are now shown in Fig. S12. Also in section 2.4 of the main text, the following sentence has been added: “Individual back trajectories in each cluster are shown in Fig. S12”.

Technical comments
8) Page 3, lines 23-24: “on air pollution, as well”
Thanks, the sentence has been corrected accordingly.

9) Page 4, lines 16-17: “the mass detection of submicron non refractory submicrometer particulate matter (NR-PM1)”
Done as suggested

10) Page 9, line 16: the authors mention that the ending location of the back trajectory analysis was at an elevation of 500 m. This is obviously not the case in Figure 14, where the ending location is at a much lower altitude than 500 m.

Thanks, it is not 500 m but the half of the mixing height. Therefore, the corrected sentence in the text now reads:
"The trajectories were released at half of the mixing height at the KIST (latitude: 37.60N; longitude: 127.05E) and the average arriving height for the back trajectories for this study was approximately 191 m"

11) Page 12, lines 25-26: "could to be due to the dilution effect"
The sentence has been corrected accordingly.

12) Page 13, lines 28-29: "constant in concentrations and size distribution (Fig. S5b)"
Thanks, it has been corrected to "Fig. S4b"

13) Page 16, lines 4-5: the O/C ratios given here do not correspond to those reported in Figure 10(a-e) and Table S1 (neither to the Aiken-ambient method, nor to the improved Canagaratna-ambient method).
Corrected

14) Page 20, line 29: "sulfate in Seoul mainly is mainly associated with regional sources"
The sentence has been corrected accordingly.

15) Page 22, line 19: "Average O3_concentration"
This has been corrected accordingly.

16) Page 23, line 5: "low RH (50 %),_and long distance transport"
This has been corrected accordingly.

17) Table 2: if r = 0.70 is the threshold for boldfaced values, please put the following value in bold:
- $C_2H_4O_2^+$ (60) vs BBOA

On the other hand, the following value must not be boldfaced:
- $C_3H_5O_2^+$ (73) vs SV-OOA
All have been corrected accordingly.

18) The resolution of several figures (2, 3, 7, 9, S2, S3, S7) is quite poor. Please pay attention to this point during the submission of the final version of the manuscript.
We will include high resolution figures in the revised manuscript.

19) Figure 1: the bottom right part of panel (b) is hidden by panel (c). It would be important to shift the different panels, so that we see the entire panel b with the locations of all the point sources of SO_2 (currently, we do not see those located on the southeast of the sampling site). BTW, it would be also interesting to mention in section 2 "Experimental methods" where the authors found all the information about the numerous point sources and their concentrations.
The figure has been amended in the revised manuscript not to hide panel (b). Also this has been addressed in the text and figure legend (concentration). This sentence in the revised manuscript

now reads: “there are a number of industrial facilities that are significant anthropogenic sources for SO_x and the highest emission rate range was measured at 4856-17222 ton/year.” Unfortunately, other point sources are not available.

20) The title of the supplementary material is not the same as the one of the main manuscript.
Thanks, the title of the supplementary materials has been changed according to the main manuscript.

21) Figure S1: in the panels (c) and (d), the labels of the left y-axes as well as those of the two colorscales are missing.
Thanks, the figure has been corrected accordingly in the revised manuscript.

22) Figure S4: the legend of the y-axis of the panel (b) should be “dSO4…” instead of “dNO3…”.
Corrected

References

Dzepina, K., Arey, J., Marr, L. C., Worsnop, D. R., Salcedo, D., Zhang, Q., Onasch, T. B., Molina, L. T., Molina, M. J., and Jimenez, J. L.: Detection of particle-phase polycyclic aromatic hydrocarbons in Mexico City using an aerosol mass spectrometer, Int J Mass Spectrom, 263, 152-170, 2007.

Xu, J., Zhang, Q., Chen, M., Ge, X., Ren, J., and Qin, D.: Chemical composition, sources, and processes of urban aerosols during summertime in northwest China: insights from high-resolution aerosol mass spectrometry, Atmospheric Chemistry and Physics, 14, 12593-12611, 10.5194/acp-14-12593-2014, 2014.