Responses to Anonymous Referee #2

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

The study reports real-time characterization and source apportionment of atmospheric PM1 in Seoul, South Korea during winter 2015. Secondary species, i.e. sulfate, nitrate, ammonium, SV-OOA, and LV-OOA, is found to contribute significantly to the ambient PM1. The PM composition is influenced by meteorological conditions, i.e. temperature and relative humidity. Wind speed and direction are particularly important in characterizing regional and local sources of OA. Locally produced OA, i.e. BBOA, COA, and HOA, contributes majorly to the total OA mass, suggesting the importance of air pollution control in Seoul during winter season.

This study is interesting and important as it is one of the first studies in the region to intensively characterize ambient PM in real-time. The results will be useful for developing or improving air pollution control and policy.

Overall the manuscript falls within the scope of Atmospheric Chemistry and Physics journal. The manuscript is well written and only some revisions are needed. Some statements in the manuscript need to be clarified and/or discussed further. After the following comments are addressed, I recommend the manuscript to be accepted for publication.

Specific comments

Experimental methods:

- PAHs measurements are not described in the methods section, yet the data shows up in discussion (e.g. Table 2). Information about PAH measurements should be added either in the methods, or in the SI and refer to it in the main text.

Thanks, as suggested by the reviewer, relevant discussions on PAH estimation has been added at Page 6, line 4-5 as follows: “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)”

- For the backtrajectory analysis, what is the air mass estimated to arrive at the location?

For a given trajectory, air mass arrived at the location represents one that passed over different area along the trajectory.

For clarifications, the following sentence has been added at Page 23 line4-6: “In addition, since air masses in each cluster were expected to have passed over regions indicated by the corresponding trajectories, investigating the composition and masses of aerosol in each cluster can shed lights on how various upwind areas influence air quality at the measurement site”

Relating to the comments on Pg 23 Ln 4-7 below, a short description of areas covered by the air mass trajectories is also provided.

Discussion:

- Pg 10 Ln 9: is the severe haze event related to the high loading periods (Fig. 2)?

Yes, the severe haze event occurred during one of the high loading periods (as marked on Fig.2). For clarification, we have revised the text to read, "Although severe haze with high PM_1 concentration close to 90 µg m^{-3} was observed several times".

- Pg 12 Ln 9-12: it could be helpful to add solar radiation data to Fig. 6 to give an idea of when photochemistry possibly occurs at the location. If solar radiation is not available, temperature could give some insights too.

The closest available solar radiation measurement data are from 20km away from the measurement site, however solar radiation is likely to be fairly homogeneous over a large area. Therefore, as suggested by reviewer, diurnal pattern of solar radiation has been added to Fig. 6 and data source has been discussed at figure caption as "The solar radiation measurement site is located at 20km away from the measurement site."

- Pg 21 Ln 6-13: what does the lack of striking diurnal profile of LV-OOA suggest? I think the lack of diurnal variation is related to the regional source of LV-OOA, which has been observed in other areas, e.g. Budisulistiorini et al. (2015, ACP) in USA, Mohr et al. (2012, ACP) in Spain.

Yes, we also think that the relatively constant diurnal pattern of LV-OOA suggests regional source of this aerosol component. For the clarification, this sentence has been revised as follow and suggested referenced has been added: "however, LV-OOA concentration was relatively constant throughout the day, suggesting regional sources of this aerosol component (Fig. 10j). Similar observations were also reported in other areas such as North America (e.g., Budisulistiorini et al., 2015;Sun et al., 2011b;Woody et al., 2016;Zhou et al., 2016b;Zhang et al., 2005b), Europe (e.g., Mohr et al., 2012;Young et al., 2015), and Asia (e.g., Huang et al., 2010;Jiang et al., 2015;Wang et al., 2016)"

- Pg 23 Ln 4-7: it would be good to add short descriptions of what areas/regions are represented by each cluster. The readers may not be familiar with geography in the study location.

The regions were briefly mentioned in the text. In response to the reviewer comments, we have added more details about the area and the text now reads: "All three Clusters appeared to originate from Russia, however there were some differences among clusters. For example, cluster 1 passed over Mongolia and North Korea whereas Clusters 2 and 3 passed over China. Furthermore Cluster 3 was composed of the longest trajectories."

Figure 14:

- What does the percentage correspond to? If it is related to the average mass concentration of PM1, the percentages for Cluster 1-3 are incorrect.

The percentage values on the bars are the average fractional contributions of each species to the average PM_1 (= NR-PM_1 + BC) mass concentration in different clusters. We suspect that the reviewer's comment was related to the total values in Clusters 1-3 not being 100%. That was because of rounding of the values.

- The figure shows that air mass arrive at 191 m agl at the location, whereas on the figure title, it is 500 m (assuming also agl). Is this a typo or they are different terms? The two elevations are different, so please clarify and/or add description about the backtrajectory analysis.

Thanks, it was typo. The arriving height was not 500 m, but the half of the mixing height calculated by the HYSPLIT program. We have revised the sentence and the text now read: "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"

Technical comments

Pg 3 Ln 28: add reference, such as Hennigan et al. (2009, ACP)

The reference has been added.

Pg 8 Ln 30: insert "respectively" after (SV-OOA).

The sentence has been corrected.

Pg 10 Ln 7: define NIER

The full name of NIER, National Institute of Environmental Research has been added.

Pg 11 Ln 16: do you mean Fig. 4b?

Thanks, it has been corrected to Fig. 4b.

Pg 12 Ln 4-6: insert "Fig. S10" in the sentence.

Done as suggested

Pg 13 Ln 29: it's supposed to be Fig. S4b

Thanks, it has been corrected to "Fig. S4"

Pg 19 Ln 5: *f44* of BBOA is higher than 0.01, I think it's around 0.05. Please check again.

Thanks, it has been corrected to 0.05.

Pg 19 Ln 26-27: BBOA enhances around 9:00 to be more exact.

Thanks, it has been corrected to 9:00.

Pg 20 Ln 21-23: I think NO3 and SO4 instead of NO2 and SO2, respectively are better tracers of oxidized species for comparison with SV-OOA and LV-OOA.

We agree that NO3 and SO4 are better tracers for SV-OOA and LV-OOA and this point was discussed at the line 24 in the submitted version. For clarification, we removed the lists of gaseous species and added the following sentence to discuss the good correlation of NO3 and SO4 with SV-OOA and LV-OOA: "Comparisons between the time series of SV-OOA and LV-OOA with gaseous species, aerosol species, and meteorological parameters further confirmed their secondary nature. As shown in Table 2, SV-OOA and LV-OOA strongly correlated with nitrate (r = 0.87 and 0.63, respectively) and sulfate (r = 0.71 and 0.80, respectively), whereas the correlations between POA factors and the inorganic aerosol species were low (r = 0.09 – 0.41)."

Pg 21 Ln 31: it should be "(Figs. 12a,b)"

Thanks, it has been corrected to Figs. 12a,b

Pg 22 Ln 19: space between O3 and concentration.
Done.

Pg 22 Ln 21-22: what do you refer by "another possible reason", is it a reason of O3 decreases? If it is so, the sentence needs to be clarified.
We indeed refer it as "another possible reason for O3 decrease". For clarification, the sentence has been changed to "another possible reason for O_3 decrease was reduced photochemical reactions due to inhibition of light by high concentration of PM {He, 2014 #416}"

Pg 23 Ln 13: delete "compositions". The aerosol compositions are similar. The difference is only concentrations of species at each cluster.
In fact, aerosol compositions are indeed fairly different among clusters. As shown in Figure 2 and –Figure 12, the average fractions of nitrate (27 vs. 20 %), SV-OOA (8 vs 3 %), HOA (7 vs 4%) and COA (8 vs 7%) decreased from high to low loading periods whereas the fractions of BBOA (9 vs 12 %), LV-OOA (10 vs 20 %), and sulfate (10 vs 12%) all enhanced. For clarifications, specific fractions are now mentioned in the text and the paragraph reads: "Aerosol composition was somewhat different between the high loading and the low loading periods. Since strong wind could inhibit the accumulation of local primary and secondary species while bring in pollutants from upwind sources, the mass fractions of species influenced more strongly by local sources, such as nitrate (27 vs 20 %), SV-OOA (8 vs 3 %), HOA (7 vs 4 %), and COA (8 vs 7 %) were lower during low loading periods compared to more polluted periods, whereas those of regional sources such as sulfate (10 vs 12 %), LV-OOA (10 vs 20 %), BBOA (9 vs 12 %) were enhanced (Fig. 12)."

Table 2:
- Correct m/z values for these compounds: C4H9+ (57), C5H11+ (71), CH2SO2+ (78), CH3SO2+ (79).
- Bold r-value for C2H4O2+ versus BBOA.

Thanks, it has been corrected.

Table 3:
- Add "trace gas concentration" on the table caption.

Done as suggested.

- Subscript the trace gases: e.g. NO2. Also check for this kind of typo elsewhere in the manuscript.

Thanks, it has been corrected in the Table and also check throughout the paper.

Figure 1c: it is not obvious where the city center is located. Add marker for the city center location.
Thanks, we intended to mention that the sampling site is located at the north east of the center of Seoul as shown at the Figure. For the clarification, the Figure caption has been revised to "Center of Seoul" instead of city center.

Figure 2: personally, I think this figure is too crowded. The sub-figures are small and some have many lines (e.g. Fig. 2f, g). Although the lines are colored differently, they are still difficult to differentiate. On, Figure 2i particularly, the line representing average number concentration is almost similar to the gradient color of legend.

As the reviewer suggested, Figure 2 has been modified. Hope that this figure looks better for the interpretation.

Figure 3: be consistent with species on the legend and the caption: e.g. NO3 or NO3-. Also be consistent in the rest of manuscript.
Thanks, it has been revised.

Figure S4b: y-axis is supposed to be dSO4/dLogDva.
Thanks, it has been revised.

References

Budisulistiorini, S. H., Li, X., Bairai, S. T., Renfro, J., Liu, Y., Liu, Y. J., McKinney, K. A., Martin, S. T., McNeill, V. F., Pye, H. O. T., Nenes, A., Neff, M. E., Stone, E. A., Mueller, S., Knote, C., Shaw, S. L., Zhang, Z., Gold, A., and Surratt, J. D.: Examining the effects of anthropogenic emissions on isoprene-derived secondary organic aerosol formation during the 2013 Southern Oxidant and Aerosol Study (SOAS) at the Look Rock, Tennessee ground site, Atmos. Chem. Phys., 15, 8871-8888, doi:10.5194/acp-15-8871-2015, 2015.

Hennigan, C. J., Bergin, M. H., Russell, A. G., Nenes, A., and Weber, R. J.: Gas/particle partitioning of water-soluble organic aerosol in Atlanta, Atmos. Chem. Phys., 9, 3613–3628, doi:10.5194/acp-9-3613-2009, 2009.

Mohr, C., DeCarlo, P. F., Heringa, M. F., Chirico, R., Slowik, J. G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Peñuelas, J., Jiménez, J. L., Crippa, M., Zimmermann, R., Baltensperger, U., and Prévôt, A. S. H.: Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data, Atmos. Chem. Phys., 12, 1649-1665, doi:10.5194/acp-12-1649-2012, 2012.

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.

Sun, Y. L., Zhang, Q., Schwab, J. J., Demerjian, K. L., Chen, W. N., Bae, M. S., Hung, H. M., Hogrefe, O., Frank, B., Rattigan, O. V., and Lin, Y. C.: Characterization of the sources and processes of organic and inorganic aerosols in New York city with a high-resolution time-of-flight aerosol mass apectrometer, Atmospheric Chemistry and Physics, 11, 1581-1602, 10.5194/acp-11-1581-2011, 2011b.

Woody, M. C., Baker, K. R., Hayes, P. L., Jimenez, J. L., Koo, B., and Pye, H. O. T.: Understanding sources of organic aerosol during CalNex-2010 using the CMAQ-VBS, Atmospheric Chemistry and Physics, 16, 4081-4100, 10.5194/acp-16-4081-2016, 2016.

Zhou, S., Collier, S., Xu, J. Z., Mei, F., Wang, J., Lee, Y. N., Sedlacek, A. J., Springston, S. R., Sun, Y. L., and Zhang, Q.: Influences of upwind emission sources and atmospheric processing on aerosol chemistry and properties at a rural location in the Northeastern US, Journal of Geophysical Research-Atmospheres, 121, 6049-6065, 10.1002/2015jd024568, 2016b.

Zhang, Q., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L.: Time- and size-resolved chemical composition of submicron particles in Pittsburgh: Implications for aerosol sources and processes, Journal of Geophysical Research-Atmospheres, 110, 10.1029/2004jd004649, 2005b.

Young, D. E., Allan, J. D., Williams, P. I., Green, D. C., Flynn, M. J., Harrison, R. M., Yin, J., Gallagher, M. W., and Coe, H.: Investigating the annual behaviour of submicron secondary inorganic and organic aerosols in London, Atmospheric Chemistry and Physics, 15, 6351-6366, 10.5194/acp-15-6351-2015, 2015.

Huang, X. F., He, L. Y., Hu, M., Canagaratna, M. R., Sun, Y., Zhang, Q., Zhu, T., Xue, L., Zeng, L. W., Liu, X. G., Zhang, Y. H., Jayne, J. T., Ng, N. L., and Worsnop, D. R.: Highly time-resolved chemical characterization of atmospheric submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass Spectrometer, Atmospheric Chemistry and Physics, 10, 8933-8945, 10.5194/acp-10-8933-2010, 2010.

Jiang, Q., Sun, Y. L., Wang, Z., and Yin, Y.: Aerosol composition and sources during the Chinese Spring Festival: fireworks, secondary aerosol, and holiday effects, Atmospheric Chemistry and Physics, 15, 6023-6034, 10.5194/acp-15-6023-2015, 2015.

Wang, J., Ge, X., Chen, Y., Shen, Y., Zhang, Q., Sun, Y., Xu, J., Ge, S., Yu, H., and Chen, M.: Highly time-resolved urban aerosol characteristics during springtime in Yangtze River Delta, China: insights from soot particle aerosol mass spectrometry, Atmos. Chem. Phys., 16, 9109-9127, 10.5194/acp-16-9109-2016, 2016.