Author response to anonymous referee #1

We thank reviewer #1 for his/her thorough and careful review of our manuscript. Below we reply to the reviewer's comments point by point. We list the original comments in black, our replies in blue, major changes or additions to the manuscript in red.

Major comments

1) The introduction should be expanded, particularly the major conclusions and findings in previous work from the same group, and then the unique of this study can be highlighted.

We have expanded the introduction as detailed below.

**Page 19408, line 10,** *addition to main manuscript***:**

We have previously deployed a high resolution aerosol mass spectrometer (HR-ToF-AMS) at the supersite of the Hong Kong University of Science and Technology (HKUST) to determine typical variations in submicron species concentrations, overall composition, size distributions, PMF-resolved organic factors and degree of oxygenation. The supersite measurements provided valuable insights into characteristics of mainly of secondary components of submicron particulate matter, with dominance of sulfate and oxygenated organic aerosol species observed [Lee et al., 2013; Li et al., 2013, 2015]. Subsequent work was conducted at a downtown location in Hong Kong, next to the roadside to assess important primary aerosol sources in the inner-city to identify contributions of long-range transport to roadside pollution, and to establish characteristic concentration trends at different temporal scales. Cooking aerosol was identified as the dominant component in submicron non-refractory organics, followed by traffic-related emissions [Lee et al., 2015]. However, the campaign was conducted in spring and summer of 2013, when PM levels are typically lower than in fall and winter.

The current work focuses on the characterization of roadside aerosol during the fall and winter seasons, when the influence of transported air mass is greatest and PM pollution in Hong Kong is generally more severe. Episodic haze events were found to be mainly driven by secondary aerosol rather than primary emissions, while day-to-day high PM concentrations were often driven by cooking aerosol. Furthermore, the sources of organic components were investigated in detail, especially SV-OOA. Traffic emissions, local cooking emissions and transported aerosol are all associated with the source of SV-OOA at Mong Kong.

2) Concerning ACSM calibration, did the authors use ammonium sulfate to calibrate RIE(SO4)? Or did the authors have a period of simultaneous measurements between HR-ToF-AMS and ACSM for the intercomparison? The interpretation of ACSM nitrate and sulfate needs to be cautious, particularly sulfate.

We have conducted RIE (SO4) calibrations and obtained an average value of 1.19, rounded up to 1.2, which is exactly the default RIE value. This has been mentioned in the supplementary information of our original submission "The ammonium RIE of 4.57 and sulfate RIE of 1.2 were chosen based on the average from five IE 17 calibrations".
We also did a brief comparison of HR-ToF-AMS and ACSM measurements between August 25 and August 29, with all measured NR-PM1 species correlating well with a Pearson's R values of more than 0.9. The average ratio of SO4, NO3, NH4 and organics of ACSM to that of AMS are 0.90, 0.88, 0.78 and 1.01 respectively as shown in the graphs below. The mass of chloride measured by AMS is much lower than that of ACSM. However, as the concentration of chloride during the sampling period was very low in general, we did not discuss chloride trends in this paper.
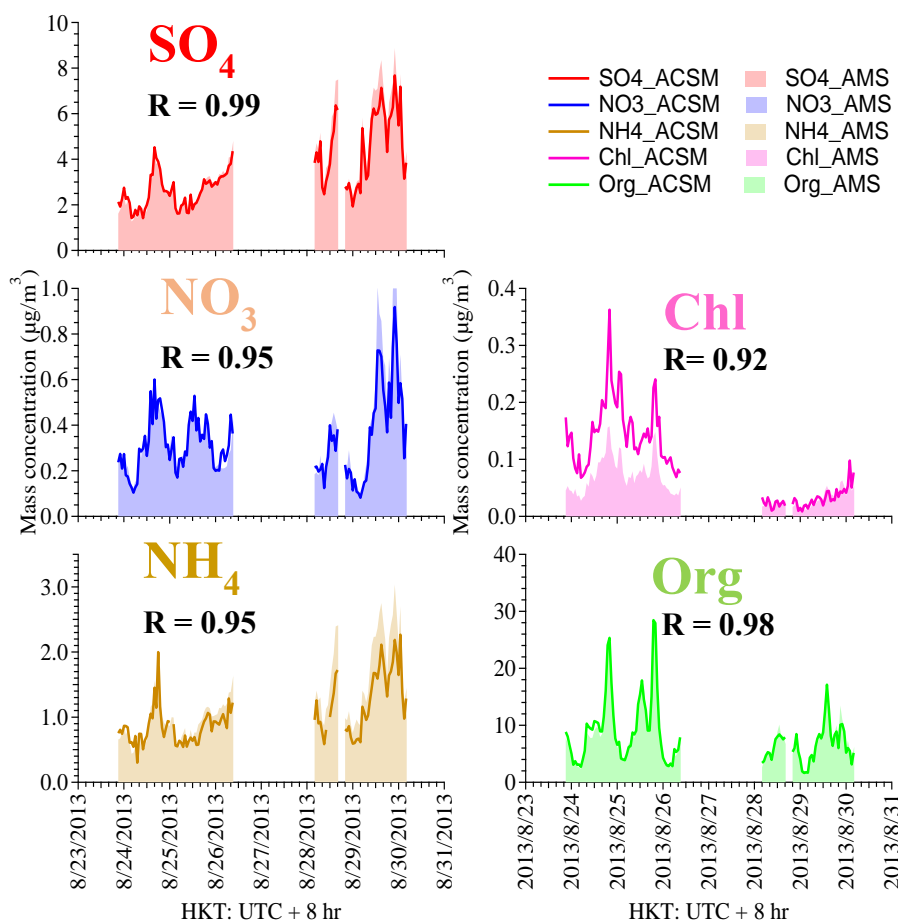
3) Fig. 9 and Fig. 10 were repeated. It's not necessary to use two figures (hourly and daily) to demonstrate the variations and roles of COA. In fact, the time series of Fig. 1b clearly shows that high NR-PM1 peaks were corresponding to high COA. In addition, when plotting Fig. 9, please be careful with the number of points for each bin. Interpreting the data above 60 ug m-3 in Fig. 9 should be careful because Fig. 1b shows a few points above 60 ug m-3.

We agree that the time series of Fig. 1b illustrates the good correlation between NR-PM1 peaks and COA peaks quite well. However, we believe that showing the relationship between the two just from the time series data of Figure 1b does not capture the whole picture. We essentially aim at using Figures 9 and 10 to examine how the PM components are responsible for episodes in different time scales, i.e., hourly basis as shown in Figure 9 and daily basis as shown in Figure 10. In fact, we found that COA showed a significant short-time impact on high hourly PM1 but less impact in daily concentrations in long-time haze events.

The number of individual data for the last two points with the average NR-PM1 concentration exceeding 60 ug/m^3 is 38 and 11, respectively. They cover in total 26 days. We therefore consider them representative.

Minor comments

1. Fig. 1b appears to be stacked plot, please describe it in the caption.

We agree and will modify the manuscript accordingly.

2. Table 4. "four chosen periods" should be "five chosen periods".

This will be changed in the revised manuscript.

3. Page 19411 Line 22-27: this paragraph can be revised and moved to "experimental" section, e.g., after line 3 in page 19409.

This has been changed in the revised manuscript.

4. Page 19413 Line 11-15: It's not consistent with Figure 5 that shows diesel-fueled vehicles are the major means during this time range.

It is correct that the number of diesel-fueled vehicles was slightly higher than that of LPG-fueled ones (mainly taxis) between 10 pm and 4am. However, as discussed in the manuscript, leakage of LPG especially during refueling, is another notable source of emissions apart from the usual on-

road driving emissions. The intense butane concentrations observed in the nighttime are likely related to leakage, as refueling of LPG-fueled taxis often takes place in the late night and early morning hours. To clarify this point, we will add the following to the concerned paragraph.

Page 19413, line 14, *addition to main manuscript***:**

Furthermore, fuel leakage during refueling of LPG vehicles may contribute more than diesel-fueled vehicular emissions to butane even though the number of diesel fueled vehicles is slightly higher than LPG ones at that time

5. Page 19415 Line 1-10: Interpreting the f44 in the spectra need to be very careful. ACSM has large uncertainties in determining f44, and often presents significantly higher value than HR-ToF-AMS (see Crenn et al., AMTD, 8, 9239-7302, 2015 and Frohlich et al. AMT, 8, 2555-2576, 2015). Therefore, higher f44 in SV-OOA than "standard SV-OOA" might be simply due to the ACSM uncertainties.

We compared our resolved mass spectra of SV-OOA with standard ones obtained by Q-AMS rather than HR-ToF–AMS, as both Q-AMS and ACSM share the same quadrupole MS technique.

We acknowledge that uncertainties of ACSM and Q-AMS in the f44 determination can play a role and have added this to the concerned paragraph.

Page 19415, line 10, *addition to main manuscript***:**

Compared to HR-ToF-AMS measurements, ACSM resolved organic spectra have been observed to show higher f44 in other studies (Crenn et al, 2015, Frohlich et al., 2015) due to inherent instrumental uncertainties in the determination of f44. This might have caused the elevated f44 observed in our SV-OOA spectrum.