Response to reviewer 2

We sincerely thank the reviewer for the valuable comments and suggestions. Below we list our point-by-point replies to the comments and the descriptions of the changes we made in the revised manuscript.

1) Point 2 in comment #3: Please add the value of d50 to the manuscript.

We redrew the Fig. S3 in the supplementary section. The detection efficiency vs. D_{ME} information was clearly shown in Fig. S3. The detection efficiency of D_{ME} = 45 nm rBC was about 3.7%. The detection efficiency of 50% corresponded to D_{ME} = 75 nm. We added the information to the Fig. S3 caption.

2) Point 3 in comment #3: Please include the response to the manuscript.

Done. We added the response to the manuscript at line 158- 165.

3) Comment #6: Please include the criteria for the identification of BBBC particles in the manuscript/Supplementary Information.

Done. We added the criteria to the supplementary information as the Section 2 and addressed the information at line 305- 306.

4) Comment #8: Further clarification for Figure 5b is required. Only the particle number fraction of pure BC particles is not included in this plot, which means pure BC particles contributed more than 50% of particle number in each size bin. However, Table 1 shows that the pure BC particles only accounted for 0.62% of total particle number measured by the SPAMS. Please explain the inconsistency.

As discussed in the comment 8, we used the number fraction of internally-mixed BC particles in **total** sampling particles to illustrate the size distribution here. The **non-BC-containing** particles contributed more 50% in each size bin.

5) Comment #10: Please add the response in the first paragraph to the manuscript.

Done. We added the response to the manuscript at line 374- 379.

6) Comment #11: Please add SO2 time series to Figure 6a. SO2 and aerosol sulphate

concentrations were increasing on December 6, matching the observed increase of relative coating thickness measured by SP2. Please discuss.

We have added the SO_2 concentration to Fig. 6a. We do believe the gas to particle conversion of SO_2 to sulfate made a contribution to the particle growth. However, the average mass ratios of NO_2/SO_2 in gas phase and NO_3^-/SO_4^{2-} in particles phase were 2.8 and 1.4 respectively. The SO_2 and particulate sulfate had a slight increase and remained at a relatively lower level during the heavy air pollution episode. Apparently, the gas to particle conversion of NO_2 to nitrate played a more important role than SO_2 to sulfate in the particle growth during this pollution episode. Please see the discussion at line 466- 471.

7) Line 299-301: Although water-soluble K+ correlated reasonably well with the BBBC particles number, the BBBC particle number did not match the high concentration of K+ on December 9. Please explain.

The water-soluble K^+ mass presented some prominent peaks on December 9. The BBBC number showed similar variation trend but the absolute value remained at a low level. We believe the discrepancy was due to two different dimensions here. The BBBC particle numbers did not necessarily match the absolute mass concentrations of K^+ all the time if the amount of K^+ in individual particles varied during different events.

8) Line 304-326: Are the diurnal cycles of BCOC-NOx and BCOC-SOx particles similar to that of KBC particles?

The BCOC-NOx and BCOC-SOx were deeply aged and did not show obvious diurnal patterns.

9) Line 340-349: Please specify the two instruments have different cut-off diameter in this paragraph.

Done. We specified at line 351- 352.

10) Figure 3 and S9 are the same. Please check if the plot in Figure 3 is correct.

We correct the mistake and used the right version of Fig. 3.

11) Figure 6: The meaning of relative area (y-axis) is unclear. Is it relative to total signals?

The relative area means the fraction of a certain cation or anion area in the total positive or negative spectrum. It is commonly used in the single particle mass spectrometry analysis (Pratt and Prather, 2009; Zhang et al., 2013; Wang et al., 2014).

Literature:

Pratt, K. A., and Prather, K. A.: Real-Time, Single-Particle Volatility, Size, and Chemical Composition Measurements of Aged Urban Aerosols, Environmental Science & Technology, 43, 8276-8282, 10.1021/es902002t, 2009.

Wang, X., Ye, X., Chen, H., Chen, J., Yang, X., and Gross, D. S.: Online hygroscopicity and chemical measurement of urban aerosol in Shanghai, China, Atmos. Environ., 95, 318-326, http://dx.doi.org/10.1016/j.atmosenv.2014.06.051, 2014.

Zhang, G., Bi, X., Li, L., Chan, L. Y., Li, M., Wang, X., Sheng, G., Fu, J., and Zhou, Z.: Mixing state of individual submicron carbon-containing particles during spring and fall seasons in urban Guangzhou, China: a case study, Atmos. Chem. Phys., 13, 4723-4735, 10.5194/acp-13-4723-2013, 2013.