

## ***Interactive comment on “Size distribution and mixing state of black carbon particles during a heavy air pollution episode in Shanghai” by X. Gong et al.***

### **Anonymous Referee #2**

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#### General comments:

This work reports black carbon (BC) particles measurements in Shanghai for 5 days using a Single Particle Aerosol Mass Spectrometer (SPAMS) and a Single Particle Soot Photometer (SP2). Seven particle types are identified based on clustering of SPAMS data, which is used to explain the coating thickness of BC particles observed by SP2 measurement. Overall, I don't recommend this manuscript to be published in Atmospheric Chemistry and Physics unless a revised version has a significant improvement in terms of scientific quality. In particular, this work should include a more comprehensive analysis for identifying sources of different particle types (see specific comments

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below), which is one of the keys to improve our understanding of coating formation on BC particles. The changes in relative coating thickness during the biomass burning influenced period is interesting but further discussion is required to make this work scientifically sound.

#### Specific comments:

1. Abstract: The abstract should explicitly highlight the scientific significance of this work, which is unclear to me in the current version. Most of the detail can be removed to shorten the abstract.
2. Page 35387, first paragraph: Aerodyne soot particle aerosol mass spectrometer with single particle measurement capability (LS-SP-AMS) has been recently used to quantify BC mixing state and chemical composition by mass in urban environment and biomass burning influenced air mass (Willis et al., 2015, Lee et al., 2015ab). Please briefly mention the technique here.
3. Section 2.1: 1) Page 35388, line 15-16: Is this assumption still valid when PM concentration is high during episode, say  $> 100$  microgram/m<sup>3</sup>? 2) Page 35389, line 5-6: It would be useful to determine the d<sub>50</sub> of BC particles from Fig. S2. 3) Page 35389, line 20: The ambient BC particles is likely highly porous/fractal structure. If a density of 1.8 g/cm<sup>3</sup> is used, the mass equivalent diameter likely represents the lower limit of BC core size. Can the author comments on how this uncertainty may affect their conclusion.
4. Sampling and SPAMS cut size: 1) Page 35391, line 12: What is the particle diameter cut-size measured by the SPAMS? 2) Page 35391, Section 2.3: Please specify that a PM<sub>2.5</sub> cut-off cyclone was used here.
5. Page 35392, line 15-17: CO should associate with any combustion source including fossil fuel combustion and biomass burning.
6. Page 35393-35395, Section 3.3: The results of BC particles classification require

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further justification. Similar to the TSI ATOFMS technique, mass spectra of different clusters looks similar to each other in general (i.e. most particles consist of potassium, nitrate and sulfate fragments), and hence it is not straightforward to identify their sources based on their mass spectral characteristics alone. A general approach is to compare the time series of particle types with different gas or particle tracers or source apportionment analysis (e.g. positive matrix factorization), which is currently missing in the manuscript. For example, NO<sub>x</sub> can be used as a tracer of local traffic emission in urban that likely correlates to the particle types emitted from vehicles (i.e. BCOC-NO<sub>x</sub> and BCOC-SO<sub>x</sub>). It would be useful to summarize their correlations in a table.

7. Fig.3 and Section 3.3: 1) The criteria for identifying whether BC particles are internally mixed with organic carbon is unclear. If C<sub>3</sub>H<sup>+</sup> fragment is used as a tracer of OC, KBC particles also has this fragment. Please clarify. 2) NaKBC and BBBC particles mass spectra are almost the same (i.e. strong K<sup>+</sup> and detectable CN<sup>-</sup> signal) except that NaKBC has a stronger Na<sup>+</sup> signal. Similar to the above comment, please compare time series of the two particle types and re-evaluate the potential sources of NaKBC particles. It is recommended to do the similar comparison for each particle types as well. 3) What is the unit of y-axis in Fig. 3? 4) Page 35395, line 5-6: SP2 data should be able to support this argument.

8. Fig. 5 and Page 35396, line 9-10: The direct comparison of D<sub>p</sub> (by SP2) and D<sub>va</sub> (by SPAMS) is inappropriate. As I point out above, D<sub>p</sub> is calculated by assuming certain particle density and spherical shape whereas D<sub>va</sub> actually depends on particle morphology. More importantly, the reviewer expects that the total value of colored area (i.e. classified particle number fraction) in Fig. 5b should be equal to one. Therefore, I don't think the green line in Fig. 5b can represent the boundary between condensation and droplet mode particles. This also make the arguments of size dependent chemical composition questionable.

9. Page 35396, line 12-25: The statements in this paragraph are too general. It is necessary to perform a more detail analysis for identifying different particle sources

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(see comment #6).

10. Page 35367, line 8-20: Is it possible that the small BC core particles in droplet mode represents the tail of condensation mode particles (I am assuming that the authors use a green line in Fig. 5a to separate the two particle modes)? Furthermore, the authors should note that Willis et al. (2015) observed both thinly and thickly coated BC particles from fresh vehicle emissions using Aerodyne soot particle aerosol mass spectrometer (SP-AMS). They are not necessary to be aged particles.

11. Page 35398, line 13-Page 35399, line 5: Again, the statements in this paragraph are too general and further investigation is required. The authors only focus on the potential impacts of NO<sub>2</sub>. However, SO<sub>2</sub> is increasing during that period and hence it would be interesting to plot the sulfate fragments measured by the SPAMS. It is also recommended to discuss the potential impacts from biomass burning plume during that period, which can be a major sources of VOCs for secondary organic coating formation. The authors may need to change their conclusion/recommendation on air quality control after considering all these factors.

Minor or technical comments:

1. Page 35388, line 24: Please report the effective density used.
2. Page 35389, line 15: I think it should be laser current instead of laser power.
3. Page 35390, line 17: Please specify the manufacturer of SPAMS.
4. Page 35391, line 12: Please correct the unit for particle size.
5. Page 35394, line 1-2: Please clarify if the author means nitrate or nitrite.

References:

Lee, A.K.Y., Willis M.D., Healy, R.M., Wang, J.M., Jeong, C.-H., Wenger, J.C., Evans, G.J., and Abbatt, J.P.D. (2015a), Single particle characterization of biomass burning organic aerosol (BBOA): Evidence for non-uniform mixing of high molecular weight

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organics and potassium, Atmospheric Chemistry and Physics Discussion, 15, 32157-32183.

Lee, A.K.Y., Willis, M.D., Healy, R.M., Onasch, T. and Abbatt, J.P.D. (2015b), Mixing state of carbonaceous aerosol in an urban environment: Single particle characterization using a soot particle aerosol mass spectrometer (SP-AMS), Atmospheric Chemistry and Physics, 15, 1823-1841.

Willis M.D., Healy, R.M., Riemer, N., West, M., Wang, J., Jeong, C.-H., Wenger, J.C., Evans, G.J., Abbatt, J.P.D.\* and Lee, A.K.Y. (2015), Quantification of black carbon mixing state from traffic: implications for aerosol optical properties, Atmospheric Chemistry and Physics Discussion, 15, 33555-33582.

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 35383, 2015.

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