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.

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 below), which is one of the keys to improve our understanding of coating formation on BC particles.

Response: We accepted the suggestion and carefully rechecked the classification process. We identified BC-containing particles sources not only based on their mass spectral patterns, but also compared the time series of particle types with different gas or particle tracers. We compared the NOx concentration with the traffic emitted BC-containing particles numbers and water-soluble K+ in ambient aerosol measured by MARGA with BBBC particles number. More detailed discussions can be seen in the response to comment 6 and revised manuscript.

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.

Response: In this experiment, there was no significant residential burning near the sampling site. However, around Shanghai, there are rural areas where people burn the crop residues as daily household fuel. We constantly observed high number fraction (15-20%) biomass burning particles in Shanghai especially during and right after harvest seasons (Yang et al., 2009;Fu et al., 2015). The biomass burning BC-containing particles observed in this experiment were all deeply aged and their relative coating thickness didn't showed obvious temporal variation.

To interpret the temporal variation of relative coating thickness of traffic emitted BC-containing particles during the heavy pollutant episode, we added more data to support our points. More discussions can be seen in the response to comment 11 and revised manuscript.

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.

Response: We shortened the abstract by removing some detailed experimental data, including the PM concentration, BC size distribution and number fractions of different particle types. The observations of the fast growth rate of BC particles and the resulting thick coating were highlighted in the revised 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.

Response: We added the following paragraph in the introduction, citing the suggested works.

"Recently, a soot particle aerosol mass spectrometer (SP-AMS) was developed to characterize rBC and non-refractory particulate matter simultaneously (Cross et al., 2010;Onasch et al., 2012;Corbin et al., 2014). SP-AMS was previously used to quantify rBC mass concentration, mixing state and chemical composition in urban environment and biomass burning influenced air (Lee et al., 2015a;Lee et al., 2015b;Willis et al., 2015)."

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/m3? 2) Page 35389, line 5-6: It would be useful to determine the d50 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/cm3 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.

Response:

1) When using the 250 nm Aquadag to do calibration, the number concentration can be reached around 2×10^5 particle cm^{-3} (~540 μg m^{-3}) with 100% SP2 detection efficiency. We believe that this assumption is still valid when PM concentration is high.

2) Done. We modified Figure S3 to show d50.

3) Zhang et al. (2015) recently found the ambient BC core had an average shape factor of ~1.2 and an average density of ~1.2 g cm^{-3}, indicating that ambient BC cores have a near-spherical shape with an internal void of ~ 30 %. With the aging process, the effective density of BC core increased and BC cores transformed to a more regular and compact shape during aging (Zhang et al., 2015). In this study, we focused on the aged BC-containing particles. We assumed a density of 1.8 g cm^{-3} was more accurate. This value was also recommended in many previous studies(Moteki et al., 2010;Moteki and Kondo, 2010;McMeeking et al., 2011).

If a smaller density was used, the bimodal distribution would remain the same. The core size distribution curves like in Fig. 2 would shift to larger size direction. The coating thickness could be reduced because of the increased core diameter. However, these changes would not affect our major conclusions since the thickness comparison are done in a relative sense.

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 PM2.5 cut-off cyclone was used here.

Response:

1) The cut size of SPAMS was 200- 2000 nm. We described the sampling range of SPAMS in the last paragraph of Section 3.3.

2) We used a $PM_{2.5}$ cyclone on the main inlet. We added this information to the experimental section.

5. Page 35392, line 15-17: CO should associate with any combustion source including fossil fuel combustion and biomass burning.

Response: Agree. We deleted this sentence.

6. Page 35393-35395, Section 3.3: The results of BC particles classification require 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, NOx 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-NOx and BCOC-SOx). It would be useful to summarize their correlations in a table.

Response:

We accepted the suggestion and carefully rechecked the classification process.

For BBBC particles, we combined ART-2a and ion-marker methods to validate the classification. We have done lab study and field measurement on the chemistry of biomass burning (mostly crop straw burning in China) BC-containing particles (Huo et al., 2015). Briefly, except for black carbon fragment ions (C_n^+ and C_n^-) in both positive and negative ion mass spectra, +39 (K^+), -26 (CN^-), -42 (CNO^-) were used as the most important tracers for BBBC particles here. Given the extremely high detection sensitivity of potassium (due to the high ionization cross-section of potassium at 266 nm) in SPAMS, we defined that +39 (K^+) as the marker must have the peak area more than 1000. Meanwhile, the peak area of +56 (CaO^+/Fe^+) and -76 (SiO_3^-) must be less than 50 because paucity of Si, Ca and Fe is the major characterization of biomass burning particles differing from coal burning particles. Because the K-rich nature of biomass material, +113 (K_2Cl^+) or +213 ($K_3SO_4^+$) were constantly observed in the mass spectra of biomass burning particles by ATOFMS, then here were alternatively selected as markers for BBBC particles instead of +39 (K^+). Lastly, -71 ($C_3H_3O_2^-$), as a significant fragment of levoglucosan, was a marker to confirm our classification. We have applied these ion markers when regrouping the ART-2a results.

We reclassified the NaKBC particles by running ART-2a. We found that more than 88% particles in this group could be re-assigned to either BBBC or KBC patterns. The rest could be assigned to pure BC, BCOC-NOx or BCOC-SOx. Therefore, we eliminated the NaKBC group, and added these particles into the other BC groups according to the mass patterns.

Water-soluble K^+ in ambient aerosol measured by an online analyzer for Monitoring Aerosols and Gases (MARGA, ADI 2080, Netherlands) was used to compare with the SPAMS biomass burning particles. K^+ mass concentration by MARGA agreed well with BBBC particles number (R^2=0.64), which confirmed our classification. We added the operation of MARGA in the experimental. We added the following figure as Fig. S6.

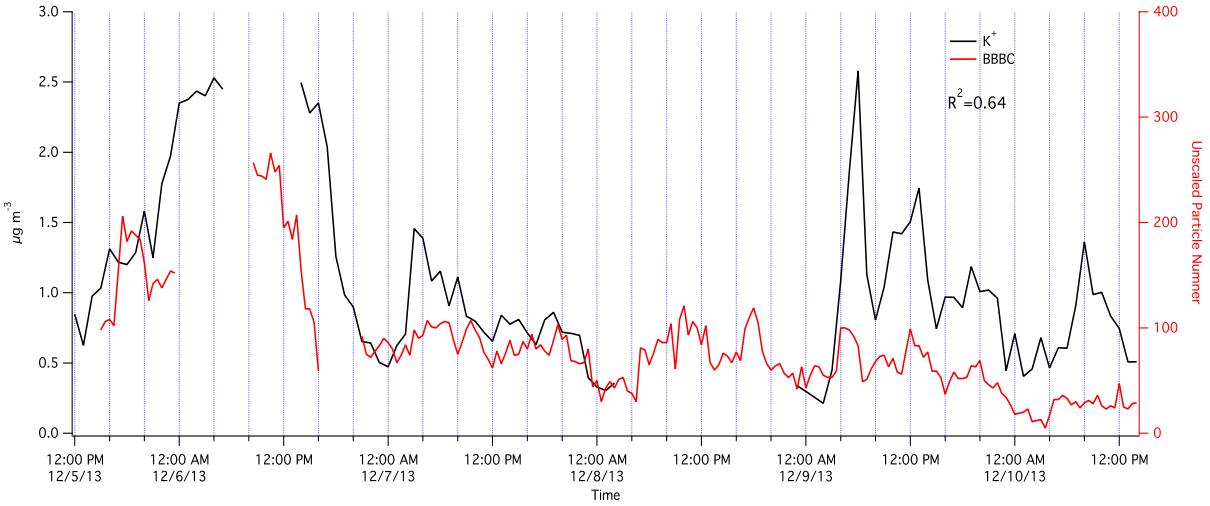

Figure S6. Temporal variations of K+ mass concentration in particles (measured with MARGA) and biomass burning BC-containing particles (measured with SPAMS).

The newly obtained KBC group exhibited pronounced diurnal variation, with two major peaks during early morning (4:00- 7:00 LT) and night hours (20:00- 22:00 LT). In the manuscript, we assigned the KBC to the diesel truck emission. Shanghai municipal government regulates that the heavily loaded diesel trucks cannot go into downtown area from 7:00 - 20:00. The diurnal variation of KBC consisted with the traffic flow of diesel trucks and confirmed our assignment. We added this part of discussion in the revised manuscript. We added the following figure as Fig. S7.

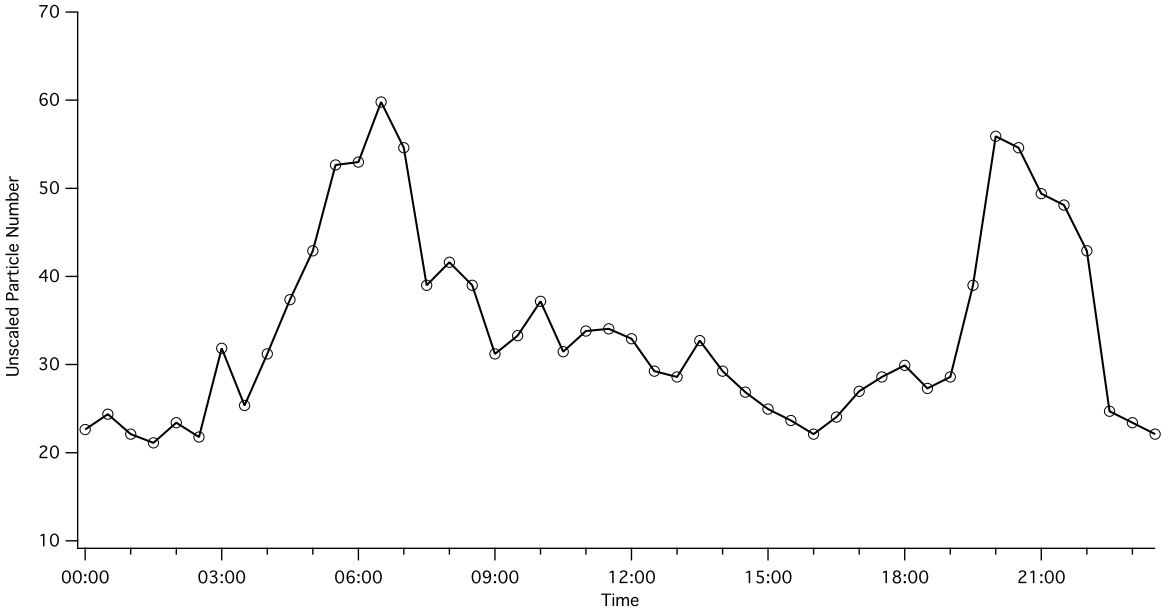

Figure S7. Diurnal variation of KBC particles measured with SPAMS.

NOx can be used as a tracer of local traffic emission in urban. Likely, the mass concentration of NOx should positively correlate to the particles emitted from vehicles (i.e. KBC, BCOC-NOx and BCOC-SOx). In this study, the NOx concentration agreed well with the sum of KBC, BCOC-NOx and BCOC-SOx particles numbers (R^2=0.65). We added this part of discussion in the revised manuscript. We added the following figure as Fig. S8.

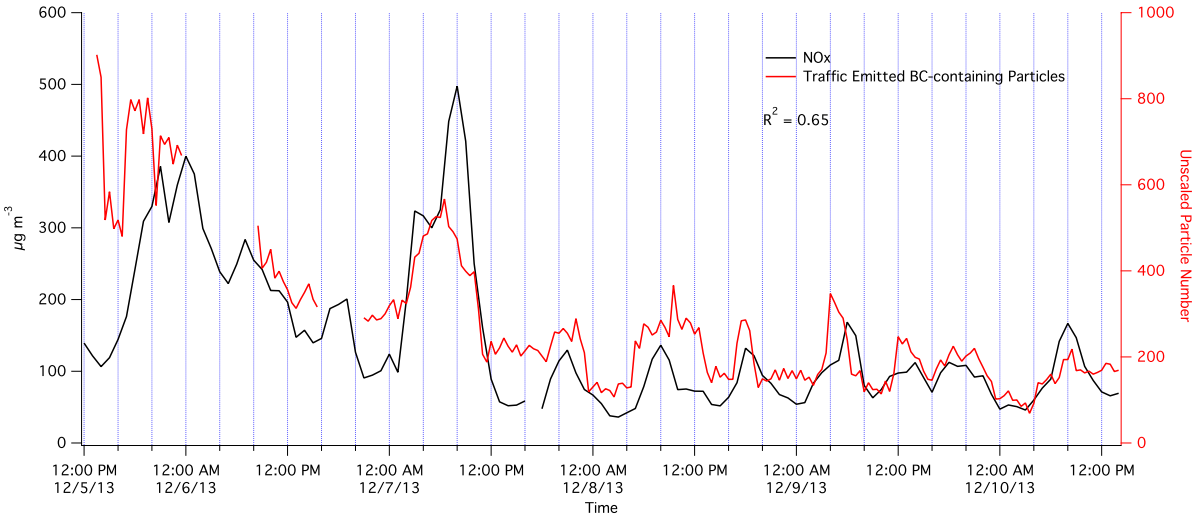

Figure S8. Temporal variation of NOx mass concentration and traffic-emitted BC-containing particles measured with SPAMS.

Moreover, to show more details of the mass spectra patterns, we moved Fig. 3 to supplementary as Fig. S5 and enlarged each mass spectrum.

7. Fig.3 and Section 3.3: 1) The criteria for identifying whether BC particles are internally mixed with organic carbon is unclear. If C3H+ 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+ and detectable CN- signal) except that NaKBC has a stronger Na+ 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.

Response:

1) The OC-containing particles were defined when area ratio of C_3H^+/C_3^+ larger than 0.2. Besides, OC-containing particles should also have some other OC ions, such as +43 (CH_3CO^+), +50 ($C_4H_2^+$), +51 ($C_4H_3^+$), +61 ($CH_3C(OH)=OH^+$) or +62 ($(CH_3)_2NHOH^+$).

2) See the response to comment 6.

3) Relative area

4) The number size distribution shown in Fig. 2 did support this argument.

8. Fig. 5 and Page 35396, line 9-10: The direct comparison of Dp (by SP2) and Dva (by SPAMS) is inappropriate. As I point out above, Dp is calculated by assuming certain particle density and spherical shape whereas Dva 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.

Response: Similar to the TSI ATOFMS, the size distribution obtained by SPAMS usually showed a single mode pattern due to the size-dependent sampling efficiency (Liu et al., 1995;Noble and Prather, 1996). As done in many previous ATOFMS and SPAMS studies

(Liu, 2003;Spencer et al., 2006;Huang et al., 2013), we used the number fraction of BC-containing particles in total sampling particles to illustrate the size distribution here. Therefore, the total value of colored area (i.e. classified particle number fraction) in Fig. 5b was smaller than one. We stated this in the first paragraph of section 3.4.2.

Although we did not carry the size-resolved calibration of sampling efficiency on SPAMS, we believe that the bimodal distribution of BC particles in Fig. 5b was real just like the results in the previous ATOFMS studies (Healy et al., 2013;Healy et al., 2012) and our SP2 measurements. Here, we did not try to make direct quantitative comparison between the data from two instruments. Instead, we used the similar bimodal distribution as an evidence to interpret the size resolved mixing states of BC particles from different sources.

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 (see comment #6).

Response: See the response to 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.

Response:

Since the two modes are overlapped, it is possible that some of the particles in the droplet mode are from the tail of condensation mode. Here, we used the minimum value between the two peaks in Fig. 5(a) (black line) and its corresponded Dp (320 nm) as the separation of the condensation mode and droplet mode. We added this separation line in Fig. 5(c) to separate the condensation mode particles (left side) and the droplet mode particles (right side).

Recently, Willis et al. (2015) found that about 10% of the fresh vehicle-emitted BC particles contained many organics (HOA-rich). Since there was no coating thickness information except the mass fraction of BC in HOA-rich particle, it was hard to do the direct comparison to our data. In our experiment, most of the small BC core with thick coating particles were observed during the extreme pollution event. As shown in Fig. 6(a), we observed the growth of small BC core particles. We believe that the small BC core with thick coating particles presented in Fig. 5(c) were mostly from the atmospheric ageing processes instead of freshly emission from vehicles.

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 NO2. However, SO2 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.

Response: We accepted the suggestions and added more data (from MARGA) and discussions in the part. Since we did not carry any VOC measurement or particle phase

organic analysis, we paid more attention to the secondary inorganic aerosol formation here. See the following paragraph and figure we added in the manuscript and supplementary.

"Variations of the major chemical species in the vehicle-emitted BC-containing particles (selected by SPAMS) were also analyzed. The relative peak areas of nitrate -63 (NO_3^-) and organic carbon (i.e., +27 ($C_2H_3^+$), +43 (CH_3CO^+)) showed a relatively high level during 16:00 LT on 5 December- 13:00 LT on 6 December (Fig. 6(b)). Guo et al. (2014) observed that gaseous emissions of volatile organic compounds, nitrogen oxides from urban transportation and sulfur dioxide from region industry were responsible for large secondary particle matter formation in Beijing. Fig. S10 shows the mass concentrations of SO_2, NO_2, the mass ratio of NO_2/SO_2, MARGA-measured mass concentrations of particulate sulfate and nitrate, and the mass ratio of NO_3^-/SO_4^{2-} in PM_1 during the whole sampling period. 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. During the heavy air pollution episode (12:00 LT, 5 December 2013 – 13:00 LT, 6 December 2013), both NO_2 and particulate nitrate increased dramatically along with the traffic emitted BC particle growth (as shown Fig. 6), while SO_2 and sulfate remained at a relatively low level. Apparently, the gas to particle conversion of NO_2 to nitrate played a more important role in the particle growth during this pollution episode. In the previous field studies(Huebert et al., 1988;Yao et al., 2002), the high mass ratio of NO_3^-/SO_4^{2-} (>1.0) was regarded as a sign of dominant traffic emission. Wang et al. (2015) found that the high mass ratio of NO_2/SO_2 resulting from traffic emissions was a major reason in triggering the heavy haze in Shanghai. In this work, the evaluation of BC-containing particles also suggested that high concentrations of NO_2 and possibly volatile organics and their transformations play a vital role for particle growth and the increase of PM loading in urban area especially during a heavy pollution episode. Reductions in the emissions of gaseous precursors are critical for remediation of the severe urban haze pollution in China."

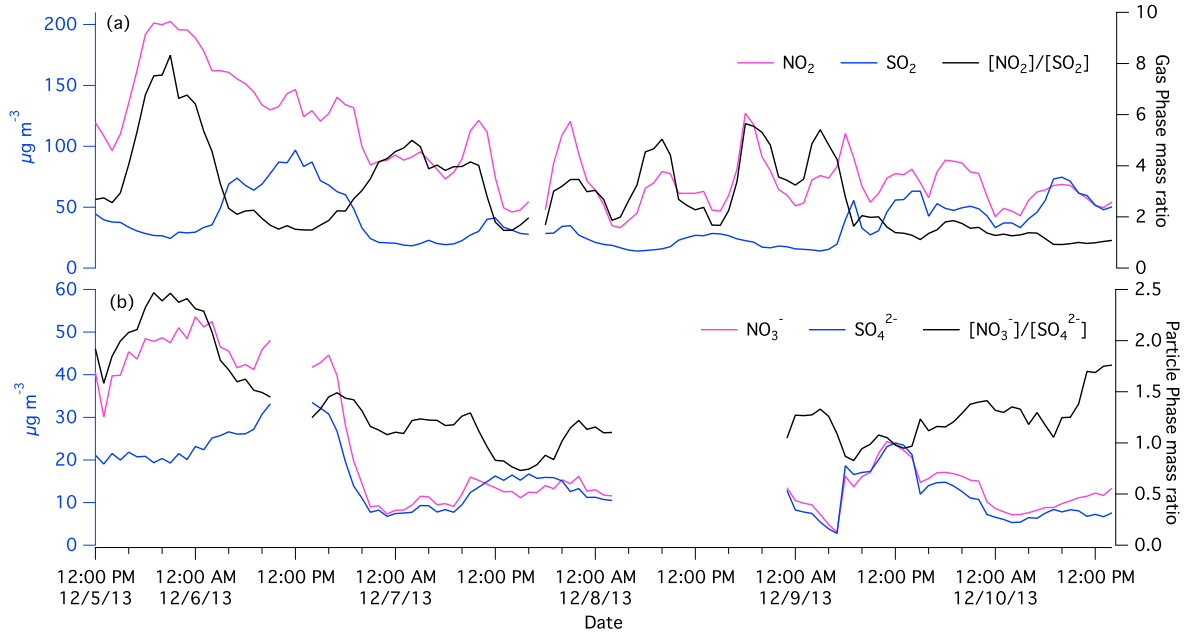

Figure S10. (a) Temporal variations of the NO_2 and SO_2 mass concentration in the atmosphere and mass ration of NO_2/SO_2 with 60 min resolution. (b) Temporal variation of NO_3^- and SO_4^{2-} mass concentration in particles and mass ratio of NO_3^-/SO_4^{2-} with 60 min resolution.

Minor or technical comments:

1. Page 35388, line 24: Please report the effective density used.

Response: We combined the DMA and APM to detect the density of The Aquadag® black carbon particles, as shown in the following figure (Fig. S1 in the revised supplementary). This result agreed with the work of Gysel et al. (2011).

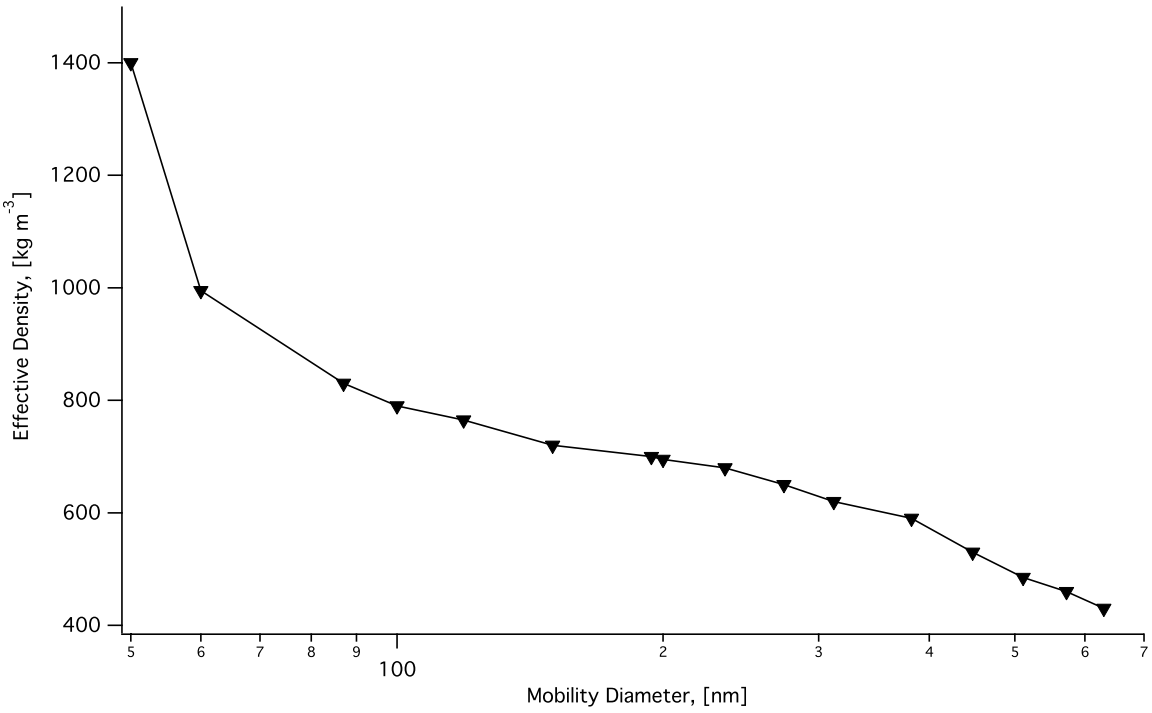

Figure S1. Effective density of Aquadag® black carbon as a function of mobility diameter.

2. Page 35389, line 15: I think it should be laser current instead of laser power.

Response: Done.

3. Page 35390, line 17: Please specify the manufacturer of SPAMS.

Response: The SPAMS in the manuscript is developed by the Hexin Analytical Instrument Co., Ltd., Guangdong, China. We added this information to the experimental section.

4. Page 35391, line 12: Please correct the unit for particle size.

Response: Done.

5. Page 35394, line 1-2: Please clarify if the author means nitrate or nitrite.

Response: Changed to "nitrate".

Again, we thank you very much for your effort and contribution!

Sincerely,

Xin Yang

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