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.

Response to Reviewer 1

1. *The manuscript presents a short collection of data from two single-particle instruments measured during a pollution episode in Shanghai, China. A single particle soot photometer (SP2) measured refractory black carbon (rBC) number and mass distributions as well as optical size. A single particle aerosol mass spectrometer (SPAMS) measured mass spectra of individual particles in parallel. Together the instruments pro- vided information on the abundance and composition of BC-containing particles. To my knowledge this is the first direct comparison and analysis of single-particle data from the two approaches, and while the manuscript does not exploit the advantages of the two techniques as much as might be possible, it still represents a valuable contribution and presents information from a unique and important region (Chinese megacities). I recommend it for publication in ACP once the following comments have been addressed:*

General comments

2. *There are a few places where I think more could be done with the co-located single particle instruments. For example, was there much consistency in results between SPAMS "mixed" particle clusters and SP-derived coating thickness?*

Response: We discussed the correlation between the particle mixing state revealed by SPAMS and the SP2-derived coating thickness in Section 3.4.2. In the revised manuscript, we have added a more detailed discussion in this section, including some new data from online ion chromatograph measurements (MARGA) and the observation of gas to particle conversion to support the consistency between the two instruments. Please also see our response to the Question 11 of Reviewer 2.

3. *In addition, more details should be provided on the comparisons between the two instruments:*

+ were the number concentrations calculated for the same diameter range for both instruments?

Response: The particles number of particles detected by SPAMS (or ATOFMS) is not quantitative (labeled as unscaled particle number in the manuscript) because of the unknown sampling efficiency (including total transmission efficiency and size-dependent sampling efficiency). The strength of this instrument is to characterize the chemical composition at a single particle level and determine the number fractions of different particle types. Besides, SPAMS detects the vacuum aerodynamic diameter in the size range of 200 - 2000 nm while SP2 detects the optical diameter of BC-containing particles in the size range of ~170 to 800 nm. Without the information of each particle's morphology and density, we couldn't convert the two different diameters precisely. It is hard to do any further quantitative comparison between the two instruments beyond the time-resolved correlation as presented in Fig. S4 and Fig. S5. In this work, we paid more attention to the mixing state and its evolution of BC-containing particles revealed by the two instruments rather than the mass or number concentrations of BC particles.

4. *+ was the reduced detection efficiency (about half) for the SPAMS accounted for in the comparison?*

Response: In this experiment, we obtained 385 683 particles with mass spectra, accounting for 56% of the total sized particles. We had no chemical information of the rest 44% particles and they were not included in the comparison.

5. *+ what is considered "internally mixed" for both instruments? Were all rBC particles measured by the SP2 included, or only those with a coating thickness above some value. If so, why?*

Response: For SPAMS, we could tell BC particles are internally mixed or pure through the mass spectral patterns. Internally mixed BC particles present not only signals of carbon cluster ions (C^-_n and C^+_n) in both positive and negative ion mass spectra, but also signal of secondary species like sulfate or nitrate.

For SP2, the internally mixed BC particles are those with a certain coating thicknesses. The internally mixed BC particles have both scattering (related to optical diameter) and incandescence (related to BC core diameter) signals. In the SP2 measurement, particles with optical diameter above ~170 nm and BC core diameter above 70nm can be observed. This is due to the scattering and incandescence detector sensitivity. Therefore, we miss some internally mixed BC particles with small core and thin coating thickness.

6. *Related point: I am not aware of any studies that directly compare single-particle MS results to a co-located SP2, so think the scatter plot in the supplementary material is worth including in the main paper. If there are some recent papers comparing SP2 to single-particle MS suggest including a short comparison of the results here to those.*

Response: We moved Fig. S4 from supplementary material to the main text as Fig. 3. We haven't found any published data comparing SP2 to single particles MS.

7. *35384 – 6: The term "refractory black carbon (rBC)" should be used when presenting any results specifically from the SP2. Recommend changing text here and throughout the manuscript. The distinction will also help distinguish results from the mass spectrometer versus SP2.*

Response: We accepted this suggestion. We used refractory black carbon (rBC) when presenting results from the SP2 and used black carbon (BC) when presenting results from the SPAMS throughout this manuscript. Both terms were explained in the introduction.

8. *35384 – 9: Add "in diameter" after 60-400 nm (assuming results are reported as diameters).*

Response: Done

9. *35387 – 28: It might be beneficial to add a small note to clarify that the "SPAMS" here is NOT the same as the SP-AMS (Soot Particle-AMS), which shares the laser system of the SP2 and therefore might lead to a little confusion.*

Response: Reviewer 2 also suggested that some related SP-AMS works should be cited. We added the following paragraph in the introduction to cite 6 SP-AMS works and also clarified the SPAMS was different from SP-AMS in the fourth paragraph of introduction.

"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., 2015b;Lee et al., 2015a;Willis et al., 2015)."

10. *35389 – 14/15: Change "laser power" to "laser current". Note that a constant laser current does not necessarily mean a constant cavity laser power. Ideally PSL could be used to verify the laser power before and after the study period, however considering the data are reported for a five-day period I do not think this is especially important in this case.*

Response: We changed "laser power" to "laser current" here. We used the 220 nm PSL to detect the laser power before and after the sampling period. The nearest equivalent scattering signal verified the constant cavity laser power.

11. *36390 – 7: suggest changing/adding ". . .was calculated as (Dp-Dc)/2, which assumes a concentric core-shell morphology."*

Response: Done

12. *35390 – 17: Is the SPAMS a laboratory-built instrument or the commercial instrument available from Livermore Instruments (SPAMS 3.0). Please specify here in addition to citing the reference.*

Response: The SPAMS is a commercial instrument developed by Hexin Analytical Instrument Co., Ltd., Guangdong, China. We added this information in the manuscript when first introducing SPAMS.

13. *35391 – 8: Are the final number concentrations corrected for this factor when comparing to SP2? Please clarify.*

Response: Please see the responses to comments 2 & 3.

14. *35391 – 28: Please state if there was a sizecut (cyclone, impactor) used on the main inlet, or if the instruments sampled all transmitted particles (with their response limited by upper size limit of the instruments).*

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

15. *35392 – 1: Suggest renaming this section because it focuses more on the trace gas measurements then meteorology. Also, according to the caption for Figure 1 data were taken from the Hongkou Station. This information should be provided in the main text as well.*

Response: We renamed the section title as: Overview of the meteorology and air quality.

We also added the information of Hongkou Station in Section 3.1.

16. 35392 – 25: The data shown in Figure 2 are study averages? Please state.

Response: This distribution was over the entire study period. We stated this in the first sentence of section 3.2.

17. 33393 – 8: All comparisons are to SP2-measured values, correct? Should be stated if so.

Response: Yes. We added the following sentence here to clarify this point: "All of the values quoted above were based on SP2 measurement." We also added similar statements in several other parts of the manuscript for clarity.

18. 35396 – 5: Should be stressed here that the results discussed here are from the LEO- fitting of SP2 data and not BC cores but the total, mixed diameter particle.

Response: Yes, the results here are the entire diameter (D_p) of BC-containing particles. We changed "D_p" to "the entire diameter (D_p)" here.

19. 35396 – 12: The paragraphs in this section move back and forth between SP2 and SPAMS data, so it would be a little more clear to state when SPAMS data is being discussed and when we are back to SP2 data.

Response: The logic flow of Section 3.4.2 was: Firstly, we showed the similarity (two-mode distribution) between SP2 and SPAMS measured size distributions. Then we used the size-resolved chemical information from SPAMS to support the BC core vs. ACT data from SP2. The comparison of the two measurements back and forth are inevitable here. Separating the two set of data in discussion would cut the logic flow.

20. 35397 – 4: The condensation and droplet modes are defined in terms of vacuum aerodynamic diameter in the manuscript, but I did not see what value was used to distinguish the modes for the SP2. I assume to make the two plots shown in Figure 5c the data was split for some mixed particle diameter. It would be useful to show that as a line on both figures, because by definition you cannot have any particles above it for the condensation mode or below for the droplet mode. It would be good to shade this region of Figure 5c either black or weight since it is an invalid data region for both plots. Finally, it is not stated if this analysis was performed for all particles measured during the study or only during specific time periods.

Response: Thank you for this suggestion. We used the minimum value between the two peaks in Fig. 5(a) (black line) and its corresponding 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).

This analysis was performed for all the particles during the study. We clarified this point in the first sentence of section 3.4.2.

21. 35397 – 26: This is a nice example of using both instruments to better understand what is happening, though there should be some care in treating the correlation as direct evidence (how well do the large, thickly coated particles correlate with other BC particle categories

identified by the SPAMS?). I do think the figure (S5) should be moved out of supplementary material into the main text. Also, the text discusses the number fraction of BC from biomass burning, but not mass fraction (which could be estimated from the SP2 data). A 20% contribution (or higher, for mass) from biomass burning seems high for an urban region, but can the authors discuss more. Do they expect significant residential burning near their site?

Response: Those larger core (80–130 nm) with thicker coating (120–300 nm) particles showed a poor correlation (R^2<0.24) with SPAMS detected traffic emitted BC-containing particles.

We moved Figure S4 from supplementary material to the main text. Since the information in Figure S5 and S4 were similar, we decided to keep Figure S5 in the supplementary section.

There is no significant residential burning near the sampling site. However, around Shanghai, there are some 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).

22. *35397 – 13: Do either of these studies report PM? Values in Shanghai were probably much higher than in the other locations mentioned, so you might have much more rapid mixing with BC acquiring thicker coatings faster.*

Response: The studies we cited here did not report PM mass concentrations. Laborde et al. (2013) reported that number concentration was around 10^3-10^4 particle cm^{-3}. We agreed that the extremely high PM concentration could contribute to the rapid mixing of BC in this study.

23. *35398 – 16: There should be a note here that the choice of BC core diameter (60-80 nm) means that you will only obtain optical sizing for the most thickly coated particles due to the scattering detector sensitivity (minimum optical diameter of about 170 nm). Do the larger BC cores display a similar rapid coating behavior?*

Response: We agree with this comment. The SP2 only obtained optical size more than 170 nm, so the observed RCT of BC-containing particles grew from 2 to 9 in this period, as shown in Fig. 6a. We might miss the initial growth of some small BC particles, but we could still capture most part of the fast growth of traffic emitted BC-containing particles. In response to this suggestion, we added the following note: "Note that we could only obtain optical sizing information from sufficiently coated particles because of the SP2 obtained minimal detectable optical diameter of ~170 nm."

The larger BC cores were composed of both biomass burning BC particles and some traffic emitted BC particles. The biomass burning particles were already deeply aged when reaching the sampling site. The overall growth trend of larger BC core particles was not obvious.

24. *35398 – 26: "incensement"?*

Response: We changed to "a relatively high level".

25. *Figure 4: Please add label for color scale including units (dN/dlogDp)? Caption for (b) states the red lines shows number concentration but axis label gives particle number.*

Response: In the original manuscript, the color scale presented the number concentration with a 10nm resolution. In the revised manuscript, we redrew Fig. 4(a) using dN/dlogDp as the color scale. Since we moved Fig. S4 to the main text, we deleted the red lines in the revised Fig. 4(a & b). We also corrected the figure caption accordingly.

26. Figure 6: Information for color scale should be provided.

Response: Done.

Response to Reviewer 2

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.

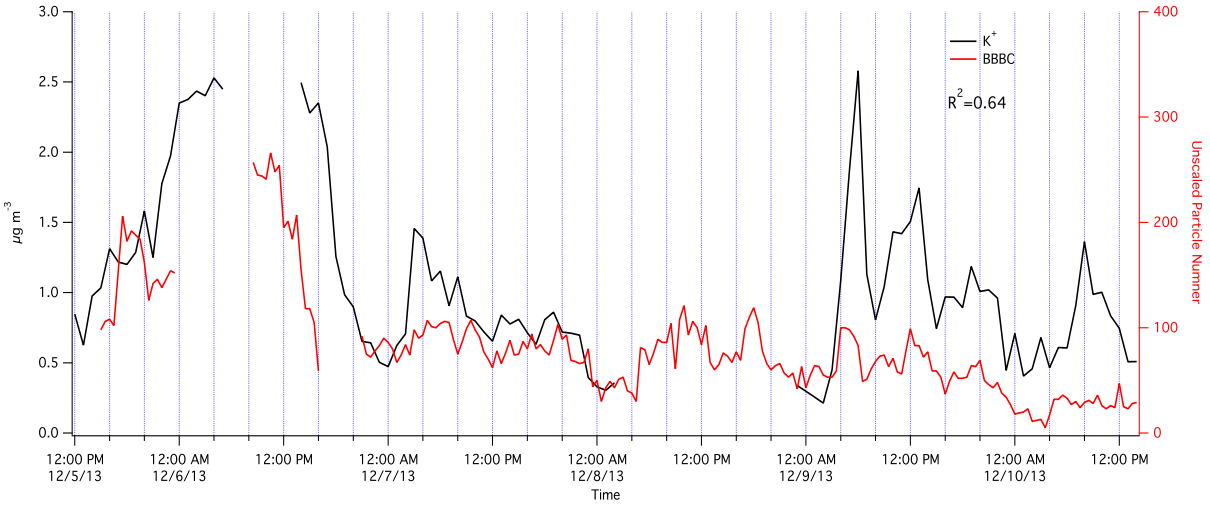

Figure S6. Temporal variations of K+ mass concentration in particles (measured with MARGA) and biomass burning BC-containing particles (measured with SPAMS).

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.

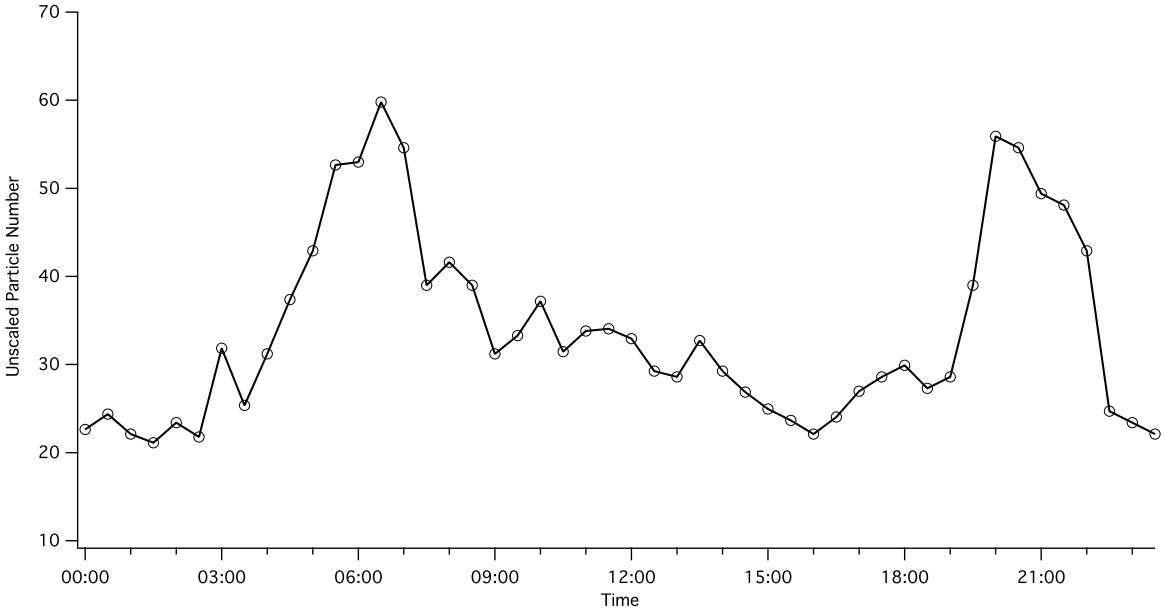

Figure S7. Diurnal variation of KBC particles measured with SPAMS.

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.

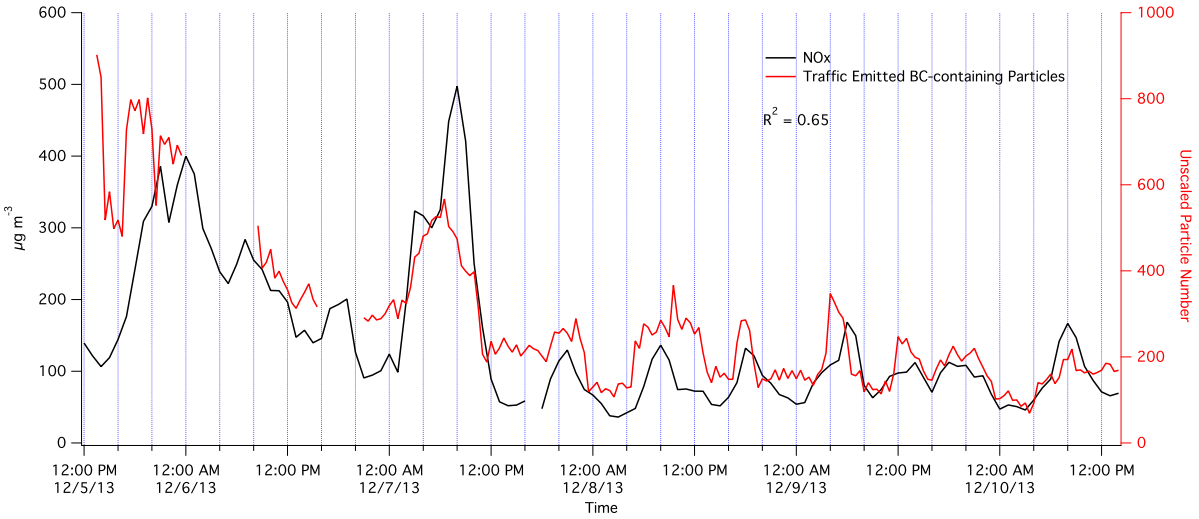

Figure S8. Temporal variation of NOx mass concentration and traffic-emitted BC-containing particles measured with SPAMS.

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."

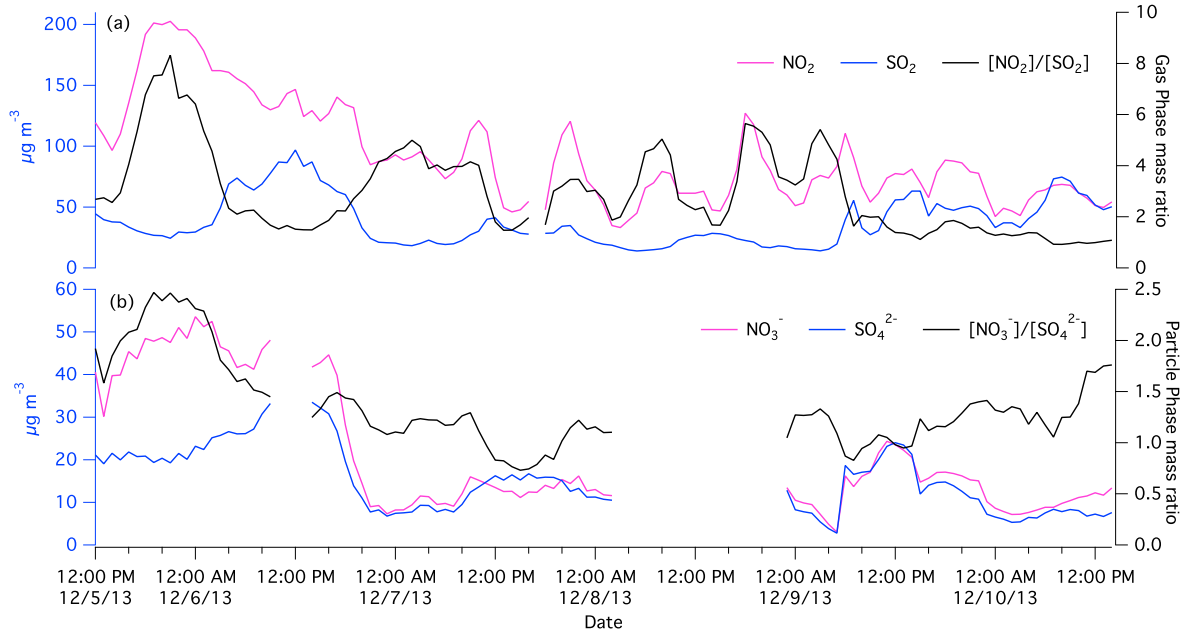

Figure S10. (a) Temporal variations of the NO_2 and SO_2 mass concentration in the atmosphere and mass ration of NO_2/SO_2 with 60 min resolution. (b) Temporal variation of NO_3^- and SO_4^{2-} mass concentration in particles and mass ratio of NO_3^-/SO_4^{2-} with 60 min resolution.

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).

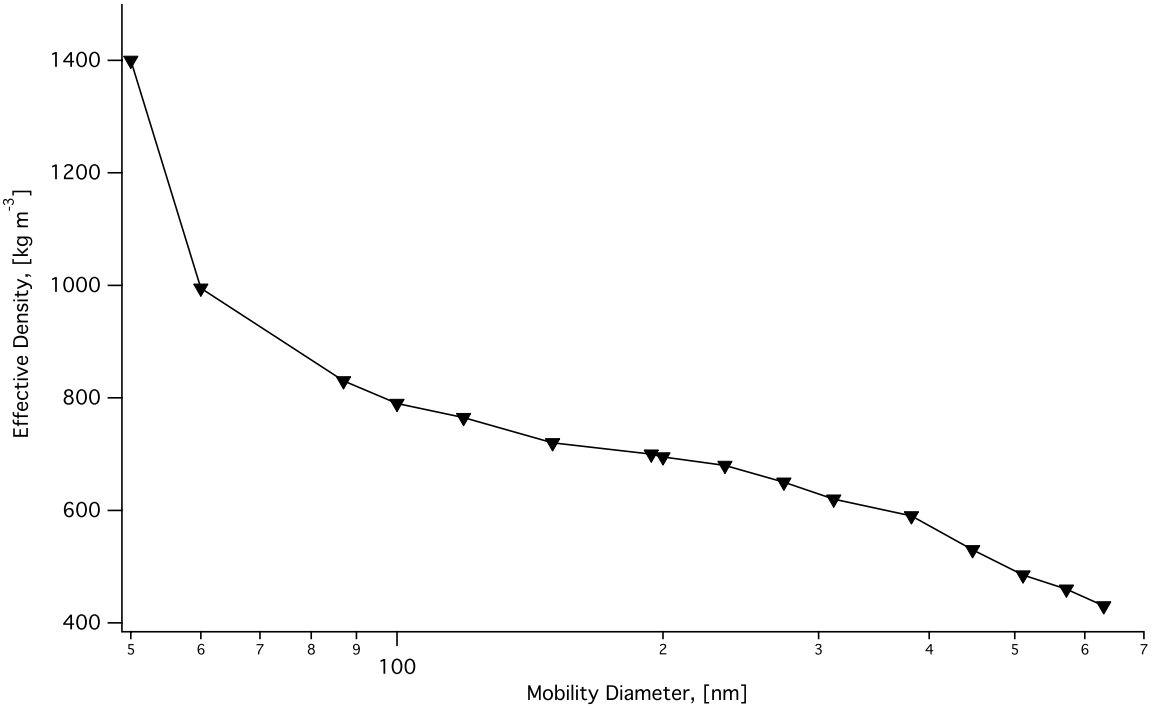

Figure S1. Effective density of Aquadag® black carbon as a function of mobility diameter.

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”.

Response to Reviewer 3

This study presents a short period in-situ measurement of black carbon in Shanghai urban area. Both Single Particle Aerosol Mass Spectrometer (SPAMS) and Particle Soot Photometer (SP2) were employed to get comprehensive information of BC- containing particles, including the number size distribution of BC-containing particles and BC cores, chemical composition of BC-containing particles, and the core-size and coating thickness of BC-containing particles. The combination of SPAMS and SP2 pro- vides a perfect tool to investigate the aging process of BC particles in the atmosphere. This study is probably the first one conducting such a measurement in heavy polluted megacities in China. Although there is not detailed discussion on the aging of BC particles from different sources, it provides some interesting results about the mixing state of BC in heavy polluted atmosphere. I therefore recommend the final publication of this study on ACP.

Specific comments:

1) Abstract: The abstract should be condensed. It is not necessary to list all the numbers like the PM concentrations in the first paragraph and the number fraction of the 7 groups in the second paragraph.

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) Introduction: I suggest the author to better motivate the study. Some similar studies in other big cities are listed in the 5th paragraph of section 1. What information is still missing or not clear? What may this study contribute to our knowledge?

Response: We rewrote the last paragraph of introduction and added the following paragraph to stress the motivation of this study.

"All of the studies mentioned above relied on either an SP2 instrument or a single particle mass spectrometer to characterize BC particles. Combining these two methods would provide the chemical and physical prosperities of individual BC particles simultaneously and greatly enhance our understanding of their sources and evolution processes. Furthermore, most previous SP2 studies focused on the BC particles during relatively clean days. Quantitative analysis on the mixing state of BC particles during heavy pollution episodes is still lacking."

3) Sect. 2.1.3: To derive the optical diameter of a particle, did you assume a core-shell particle structure in the Mie calculation? As mentioned in this paragraph, an effective negative coating thickness will be yielded if small amount of BC stays on the surface of a big particle. I think in atmosphere, the structure of most BC-containing particles is neither ideally core-shell nor BC staying on surface, but something in between (e.g. BC coated with other species but not at the center). Do you have any estimate about the possible influence of this effect on the ACT yielded with your method?

Response: The SP2 data analysis was based on the LEO-fit method. In this method, a concentric core-shell structure of BC-containing particles was assumed. In ambient atmosphere, China et al. (2013) quantified the morphology of BC particles and classified them into four categories: ~ 50 % were embedded (heavily coated), ~34 % were partly coated, ~12 % had inclusions and ~4 % were bare. We believed that during the heavily air pollution period, the heavily coated

BC-containing should account for much more than 50%. The ACT in the manuscript did not present the precise coating thickness of BC-containing in the atmosphere, but displayed the aging degree of BC-containing particles. Moteki et al. (2014) combined the LEO-fit and lag-time methods to identify the morphological types of BC-containing particles. However, this was beyond the scope of this manuscript.

4) Sect. 3.2 para. 2: These comparisons are not necessary. I think it does not make too much sense to compare your 5-day measurement during extreme pollution episode with other results with might be yielded in different seasons, pollution situations, and types of site. Such a comparison may give the audience a wrong impression.

Response: The cited works were all measured by SP2. So far, there are limited SP2 measurement all over the world. We do think that a comparison is justified even though different environments are compared.

5) Sect. 3.3: Some of the groups (e.g. KBC and NaKBC) have similar mass spectra and are attributed to similar sources, and are not really distinguished in the follow sections. Please consider reducing the number of groups. Or keep the classification as it is in this section, and combine some of them in the later discussion (fig. 4 and 5).

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.

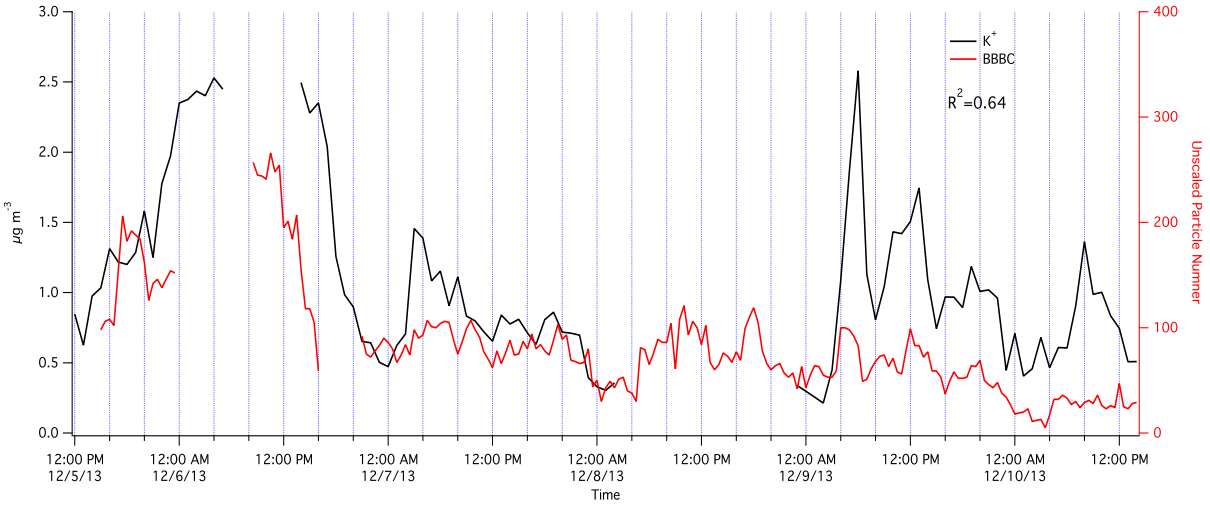

Figure S6. Temporal variations of K+ mass concentration in particles (measured with MARGA) and biomass burning BC-containing particles (measured with SPAMS).

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.

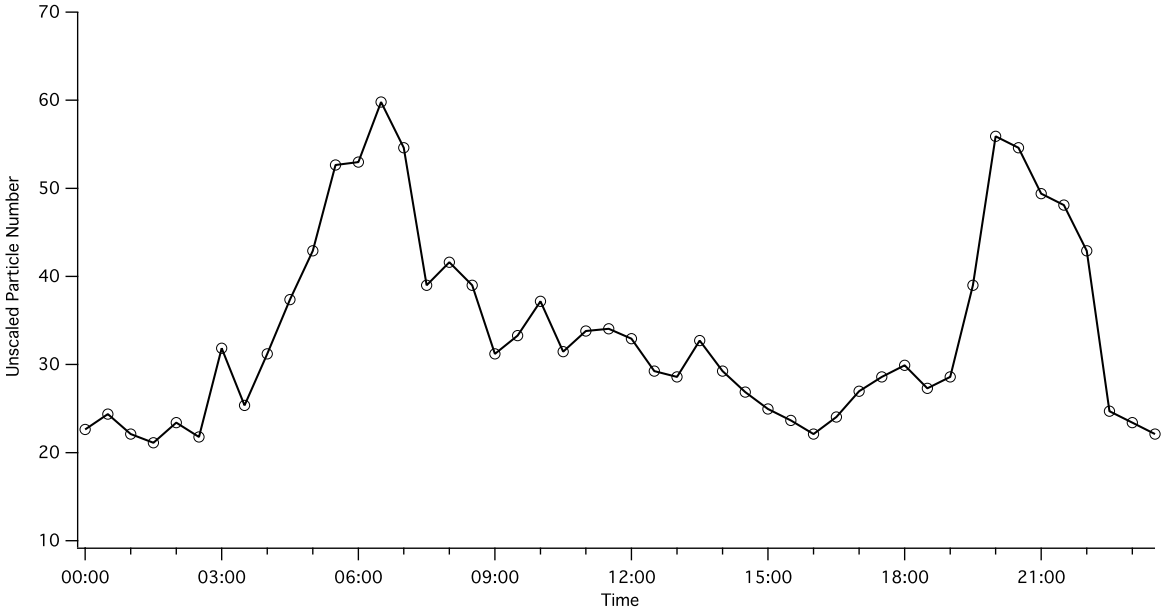

Figure S7. Diurnal variation of KBC particles measured with SPAMS.

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.

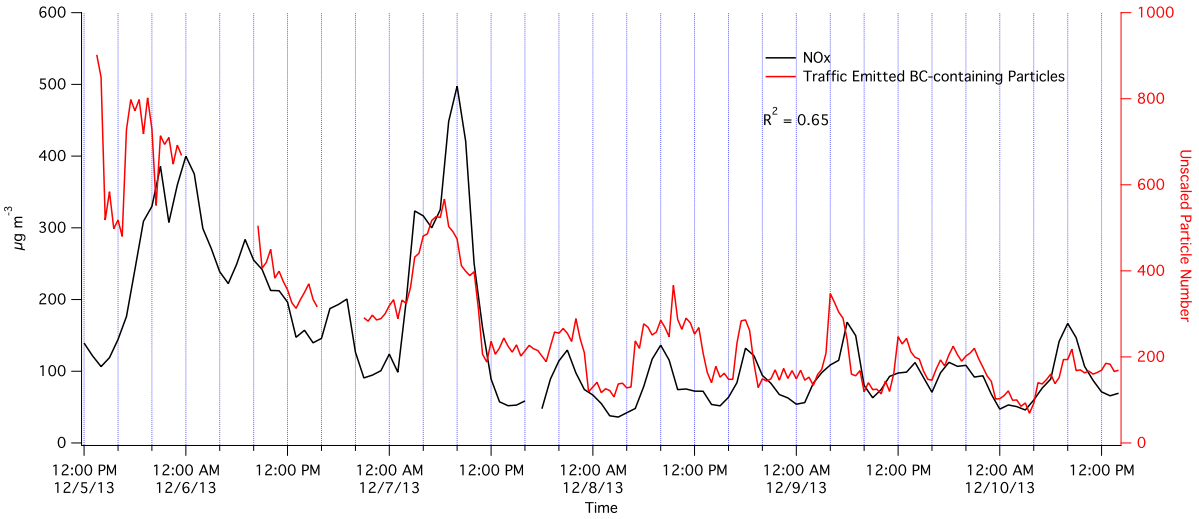

Figure S8. Temporal variation of NOx mass concentration and traffic-emitted BC-containing particles measured with SPAMS.

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

6) Sect. 3.4.2: The author mentioned that BCOC-NOx particles are much older and are supposed to have larger size (P35396L19). But in Fig. 5B we can see the ratio of BCOC-NOx particle number to BCOC-SOx particle number is even higher in condensation mode. Do you have any explanation on this?

Response: In this work, particles detected by SPAMS were mostly aged including the condensation mode particles. The higher BCOC-NOx number fraction in condensation mode is because of the less fractions of KBC and BBBC particles in this mode. In droplet mode, all the KBC and BBBC particles presented strong nitrate signals. To avoid any confusion, we deleted this statement and rephrased the discussion in the revised manuscript.

7) Sect. 3.4.2: Fig. 6 only shows the information of traffic-emitted BC-containing particles. I think it is not enough to support the conclusion of "Reductions in the emissions of gaseous precursors, such as NO2 in the urban area, are critical for remediation of the severe urban haze pollution in China.".

Response: In this work, traffic-emitted BC-containing particle was taken as an example to see the ageing process of primary particles. To make our conclusion more convincing, in the revised manuscript, we included the MARGA data on PM_1 to illustrate the important contribution of nitrate to the PM concentration. Please also 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."

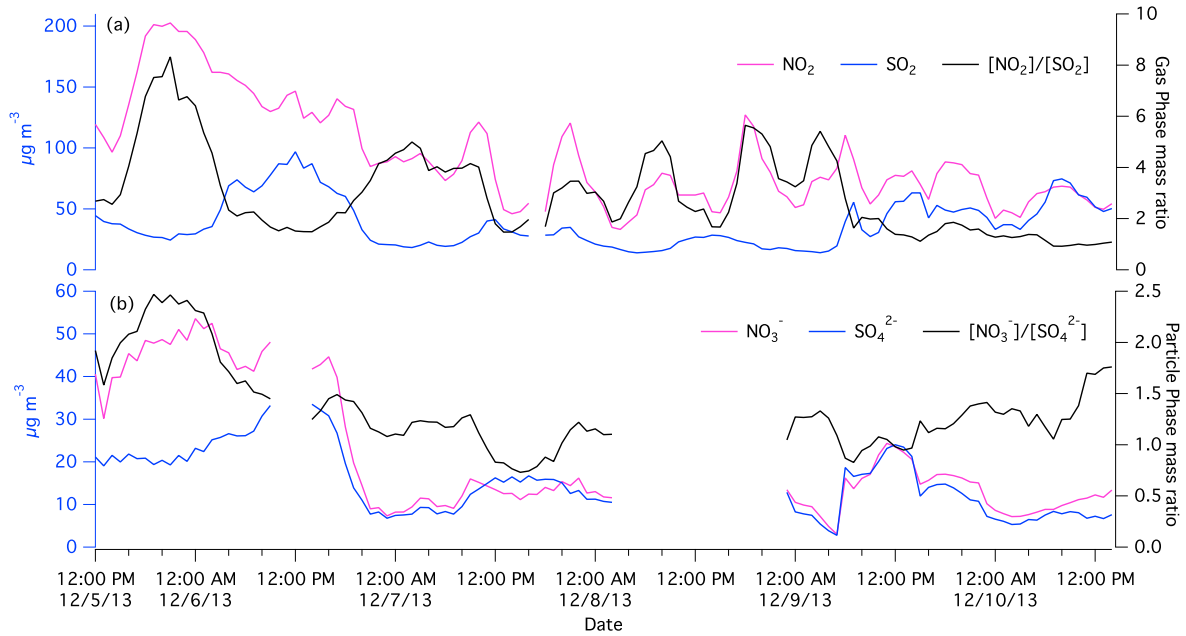

Figure S10. (a) Temporal variations of the NO_2 and SO_2 mass concentration in the atmosphere and mass ration of NO_2/SO_2 with 60 min resolution. (b) Temporal variation of NO_3^- and SO_4^{2-} mass concentration in particles and mass ratio of NO_3^-/SO_4^{2-} with 60 min resolution.

8) P35387L4: it is better to use rBC here and after for SP2 measurement

Response: We accepted this suggestion. We used refractory black carbon (rBC) when presenting results from the SP2 and used black carbon (BC) when presenting results from the SPAMS throughout this manuscript. Both terms were explained in the introduction.

9) P35385L26: absorption cross section

Response: Fixed.

10) P35390L17: please give the information of manufactory of SPAMS

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

11) P35391L12: please give the full name of Dva

Response: We added the full name of D_{va} (vacuum aerodynamic diameter) in the revised manuscript.

12) P35396L13: I would not say BBBC particles dominate the droplet mode if they take less than 30% number.

Response: We changed "dominant" to "higher".

13) P35398L26: I did not see any "significant incensement". Maybe use "a relatively high level" instead.

Response: Fixed.

14) Figures: it is better to use the same style of unit as in the text, e.g. ugm-3 instead of ug/m3.

Response: Fixed.

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

Sincerely,

Xin Yang

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