Anonymous Reviewer #1:

We would like to thank the reviewer for the valuable and constructive comments, which helps us to improve the manuscript. Listed below are our responses to the comments point-by-point, as well as the corresponding changes made to the revised manuscript. The reviewer's comments are marked in black and our answers are marked in blue, in which the revision in the manuscript is further formatted as '*Italics*'.

1. Summary

This paper discusses investigations of the SP2 instrument for measuring black carbon, and its comparison with a differential mobility analyzer (particle sizer) with volatility applied. The technical work appears to be competent. The interpretation, however, appears somewhat rambling. Additionally, these authors published an earlier paper (Atmos Meas Techniques) that investigated many similar issues including measurements from a site near Beijing (Xianghe). I think this paper could be published, but the presentation needs to be sharpened quite a bit. The authors need to explain exactly what questions are being asked, how they are relevant to atmospheric chemistry and radiative transfer, how the findings in this paper provide new information, and summarize recommendations. Especially compared with their earlier paper, the authors could state what questions had been left open, and what the present paper now addresses.

Response: We would like to thank the reviewer for the constructive comments. We have tried to sharpen the presentation of the manuscript and organize it in a way so that scientific questions are clearly stated. Please find more details below.

(1) We thank the reviewer for raising the question on differences between the current study and our previous publication at AMT (Zhang et al. 2016, AMT). Although based on the same data sets (obtained at Xianghe site near Beijing) as in the current

manuscript, the previous paper "Measuring the morphology and density of internally mixed black carbon with SP2 and VTDMA" by Zhang et al. (AMT, 2016) specifically focused on:

- demonstrating an technical approach to determine the effective density of black carbon (BC) cores for internally-mixed BC particles (hereafter named as 'thick-coated BC particles' following the reviewers' suggestion) by a combined system of Single Particle Soot Photometer (SP2) and Volatility Tandem Differential Mobility Analyzer (VTDMA).

The effective density of BC cores for the thick-coated (internally-mixed) BC particles obtained in the previous work is then used in this work to calculate optical size of thick-coated (internally-mixed) BC particles. In the current study, we

- presented the general feature of the mixing state of BC-containing particles: number/mass fraction, diurnal variation, turnover rate, and coated thickness at a polluted sub-urban area of Beijing, China, in summer time;
- validated the sizing ability of SP2 (from ~200 nm to 400 nm) for both BC-containing particles and scattering particles and further explored the possibility of using LEO-fit method and Mie calculation to extend its sizing ability towards larger size range (i.e., from ~400 nm up to ~550 nm, usually the SP2 scattering signal becomes saturated for the particles with diameter larger than ~400 nm); Note that, previously the LEO-fit method is usually used for the BC-containing particles to retrieve the original size of the particles and coated thickness (e.g., Gao et al., 2007; Schwarz et al., 2008; Laborde et al., 2013).
- compared the size distribution of the refractory BC from SP2 with that of non-volatile cores from VTDMA at 300 ℃, revealing a large fraction of low volatile non-BC components at Xianghe even at this high temperature.

The above major results in this work have been left open in the AMT paper, and we believe that the two papers are readily distinguishable and very much hope that the

reviewer is convinced by our clarification.

(2) To sharpen the manuscript and present the scientific questions more clearly, we intensively revised the manuscript according to both reviewers' comments and suggestions (see attached the tracking change version of the revised manuscript), especially modified the abstract, the last paragraph of the Introduction section, the Methods section with a schematic plot of the data processing flow, and reorganized the Results and Discussion section 3.1, as made the following changes:

- The abstract now read as "*Black carbon (BC) particles in the atmosphere play an important role in regulating the earth's climate and their climate effects strongly depend on the mixing state. During the CAREBeijing 2013 campaign, we measured the size-resolved mixing state of BC-containing particles at 200, 250, 300 and 350 nm at a regional site (Xianghe, ~60 km to the south of Beijing) in the North China Plain (NCP) from 8 to 27 July 2013, by combining a single particle soot photometer (SP2) and a volatility tandem differential mobility analyzer (VTDMA). The SP2 measurements showed a clear two-mode distribution of the 'lagtime', based on which the BC-containing particles were classified into the 'thin-coated (or bare)' and 'thick-coated' BC particles. With similar method proposed in our previous study (Zhang et al., 2016), we found that the thin-coated (or bare) BC particles at 200-350 nm exhibited an irregular shape with fractal dimension of ~2.23 and very low effective density of about 0.25-0.45 g cm^{-3}. The average refractive index (RI) of the non-BC particles was retrieved to be ~1.42 by matching their optical size and mobility size pre-selected by the VTDMA. Here, only the unsaturated scattering signals of SP2 (i.e., particles with smaller than ~400 nm) were used in the retrieval and the obtained RI was found to be almost size independent, which was then applied as the RI of the non-BC coating materials of the BC-containing particles. For the thick-coated BC particles, the effective density (1.2 g cm^{-3}) and refractive indices (1.67-0.56i) of the BC core were adopted from Zhang et al. (2016). Based on the above parameters and the BC mass and particle*

scattering cross-section measured by the SP2, the optical sizes of the thick-coated BC-containing particles as well as the large non-BC particles with saturated scattering signal were derived with the leading-edge-only (LEO)-fit method and Mie calculation. We show a very good agreement between the optical particle diameter and the mobility diameter for the large non-BC particles (diameter > ~400 nm), as well as for the thick-coated BC particles. With our approach, the upper sizing detection limit of the SP2 can be extended by from ~400 nm to ~550 nm.

During the measurement campaign, we found that the non-BC particles contributed ~85-90% of the total aerosol numbers in the size range of 200 nm to 350 nm. The rest 10-15% of the particles contained BC, which were portioned out into thick-coated and thin-coated (or bare) BC particles by ~45% and ~55%, respectively. The number fraction of thick-coated BC in total BC-containing particles (F_{In-BC}) shows pronounced diurnal cycles with a peak around noontime and an apparent turnover rate up to 6-9% h^{-1}. Such diurnal cycles are similar to the finding of Cheng et al. (2012), suggesting strong photochemical aging of BC particles and a competing effect of emissions and aging processes. The average coating thickness (CT) of the thick-coated BC particles (200-350 nm) was in the range of 74-108 nm, which increased with increasing particle size. Compared with an average CT of ~30 to ~60 nm in previous measurements in other less polluted locations, e.g., Long Island (NY, USA) in summer time (Sedlacek et al., 2012), Mexico City in spring time (Subramanian et al., 2010), Fukue Island (Japan) in spring time (Shiraiwa et al., 2008) and Weybourne (UK) in summer time (Liu et al., 2013), the observed thick-coated BC particles at Xianghe were more-aged, indicating fast secondary processes and strong light absorbing capacity of BC particles under the polluted conditions in the NCP. Moreover, our measurements revealed a large fraction of low-volatile non-BC components at 300 °C. Above the incandescence detection limit of SP2 (BC particle diameter larger than ~70 nm), the refractory BC particles accounted for only 16-35% of residual

particles measured by VTDMA at 300 ℃ in number. In such case, the assumption that the refractory residual materials at 300 ℃ are BC as in previous VTDMA studies (Philippin et al., 2004; Wehner et al., 2009; and Cheng et al., 2009) may cause significant overestimation of the number of internally mixed BC particles and on the other hand it may lead to an underestimation of the light absorption enhancement factors in NCP due to the remaining low-volatile non-BC materials coating on the BC particles after heated by the thermodenuder."

- At the end of the Introduction section, we added "*In this study, we measured BC mixing state combining SP2 and VTDMA techniques at a polluted sub-urban area of Beijing, China, in summer time. We first validated the sizing ability of SP2 (from ~200 nm to ~400 nm) for both BC-containing particles and non-BC particles and further explored the possibility of using LEO-fit method and Mie calculations to extend its sizing ability towards larger size range (i.e., from ~400 nm up to ~550 nm). Based on the particle size information, we then presented the general feature of the mixing state of BC-containing particles: number/mass fraction, diurnal variation, turnover rate, and coating thickness. At last, we compared the size distribution of BC cores from SP2 with that of non-volatile cores from VTDMA to explore the existence of the low volatile non-BC components at 300 ℃.* "
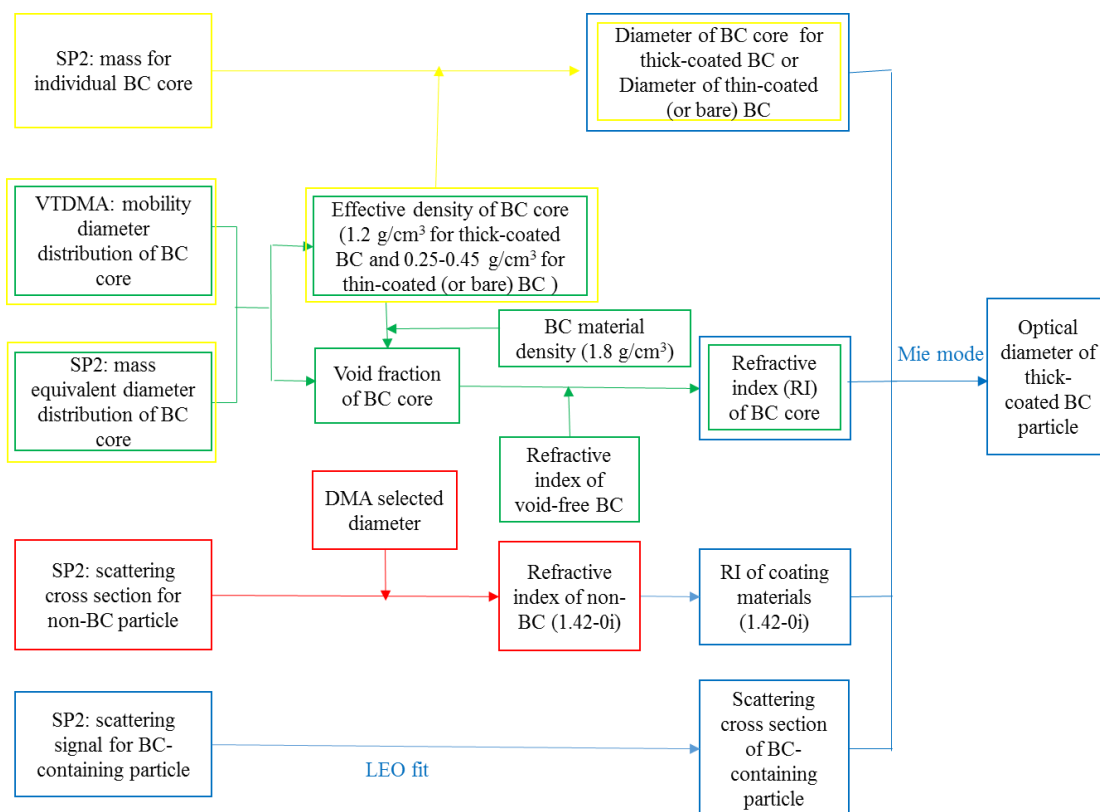
- We add the following plot in Methods section:

SP2: mass for individual BC core

Diameter of BC core for thick-coated BC or Diameter of thin-coated (or bare) BC

VTDMA: mobility diameter distribution of BC core

Effective density of BC core (1.2 g/cm^3 for thick-coated BC and 0.25-0.45 g/cm^3 for thin-coated (or bare) BC)

BC material density (1.8 g/cm^3)

Refractive index (RI) of BC core

Mie mode

Optical diameter of thick-coated BC particle

SP2: mass equivalent diameter distribution of BC core

Void fraction of BC core

Refractive index of void-free BC

DMA selected diameter

SP2: scattering cross section for non-BC particle

Refractive index of non-BC (1.42-0i)

RI of coating materials (1.42-0i)

SP2: scattering signal for BC-containing particle

LEO fit

Scattering cross section of BC-containing particle

Fig. R1 (Figure. 2 in the revised manuscript). The data processing and analysing procedure in this study.

- We reorganized section 3.1 into *3.1 Morphology and effective density of the thin-coated (or bare) BC particles and 3.2 Particle optical size derived from SP2 (3.2.1 Non-BC particles with diameter larger than ~400 nm, 3.2.2 Thick-coated BC-containing particles).*

2. Specific comments

(1) The writing here is not bad. But it needs to be reviewed by a native English speaker. Examples in the introduction: "results in largest uncertainties" (line 15), "while the rest components" (line 22), "The causes are due to that" (line 25). I won't list all of them. In particular "inversed" cannot be used as a verb as it is here, but instead one should say "determined by inversion."

Response: Many thanks. The revised manuscript has been modified to avoid grammatical errors.

(2) I also note that I wrote this review before looking at the comments of the other reviewer because they should be independent. There could be some disagreement or repetition.

Response: Thanks.

(3) Introduction lines 8-10. Most of these citations discussed absorption, not even climate and certainly not BC as second most important warming agent. They do not support the statement.

Response: Thanks. We have added some other citations, which support the statement of "climate effect" (*Gustafsson et al., 2016; Chung and Seinfeld, 2005; Ramanathan and Carmichael, 2008*) and "BC as second most important warming agent" (*Gustafsson et al., 2016; Ramanathan and Carmichael, 2008*).

(4) mixing state: authors should state what "mixing state" means. On page 3 line 19, they say that it was quantified. The reader should know what to look for. Especially the use of "externally-mixed BC particles" throughout the manuscript is awkward. There could be externally-mixed aerosol populations in which bare BC exists, but when counting the bare BC particles only, one wouldn't say these individual particles are "externally mixed."

Response: Thanks for the comments. We have stated/defined the "mixing state" in revised manuscript according to the literature (Oshima et al., 2009): "*BC mixing state (namely, the degree to which BC particles are coated with other aerosol components, Oshima et al., 2009)*"

We agree with the reviewer that "externally mixed" used on aerosol populations seems more appropriate, therefore we changed the terminology of "externally-mixed BC particles (BC externally mixed with other aerosol components)" and "internally-mixed BC particles (BC internally mixed with other aerosol components)" in the manuscript into "*thin-coated (or bare) BC particles*" and "*thick-coated BC*

particles", respectively.

(5) equations 3 & 4. These are not real equations. They are dependences, I think. In that case it should just be stated: the determination of the optical diameter depends on the scattering cross-section and the assumed refractive index, rather than writing as a false equation. Just before this equation, authors state that equations 3 and 4 list input parameters for the Mie model. But scattering cross section is an output from a Mie model, not an input.

Response: We thanks the reviewer to point this out. Following the suggestion, the statement have changed as "*To determine the optical diameter of the non-BC particle ($D_{opt,non-BC}$), its scattering cross section (C_s) and the refractive index (RI_{non-BC}) of non-BC components should be known, as in the relationship (3) (Bohren and Huffman, 1983):……*

For the thick-coated BC particle, its optical size is determined from its scattering cross section (C_s), the diameter of the BC core (D_c) and the refractive index of BC core (RI_c) and coating material (RI_{cm}), as in the relationship (4):……"

We apologize for the inappropriate statement "Equations 3 and 4 list input parameters for the Mie model", and have changed it into "*The optical sizes of the thick-coated BC-containing particles and the non-BC particles, especially those with saturated scattering signal in SP2, are retrieved using LEO-fit method (Gao et al. 2007) and Mie calculation.*"

(6) Fig 3 compares the mobility-determined size with the size determined from the SP2 scattering cross section. But the refractive index used in this determination was already fit, that is, we should expect the two to agree because one was derived from another. I don't think this justifies anything.

Response: We thank the reviewer to raise this concern. However, there might be some misunderstanding on the data processing and analysing procedure. Here, we make a

chart (Figure R1) to illustrate the whole data processing and analyzing procedure. For non-BC particles within the SP2 detection limit (200-400 nm), we matched the optical diameter to the electro-mobility diameter by tuning its refractive index (RI), marked red in Figure R1. The retrieved RI for non-BC particles (~1.42 - 0i) seems to be independent on particle size (Figure 3 in the manuscript). Thus, when retrieving the optical size for the non-BC particles larger than 400 nm (saturated scattering signals in SP2 measurement) using LEO-fit and Mie calculation, we applied the same RI (~1.42 - 0i). On the other hand, for the thick-coated (internally-mixed) BC particles, when retrieving its optical diameter with the LEO-fit method (Gao et al. 2007) and Mie calculation, we did not adjust RI for either BC core or coated materials. In data process, RI for coated material was assumed to be same as the RI of non-BC particles (fixed with ~1.42 - 0i). RI for BC core was estimated as the volume weighted average of the RI of void and BC as Eq. (6) in the manuscript, marked green in Figure R1. The diameter of BC core was calculated from the BC mass given by SP2 and the predefined BC effective density (1.2 g/cm^3), marked yellow in Figure R1. The volume of void in the BC core and the BC core effective density (1.2 g/cm^3) are determined by the method introduced in Zhang et al. (AMT, 2016). After getting the diameter and RI of BC core, RI of coated material and the scattering signal of BC-containing particles from SP2, we used LEO-fit combined with Mie model to retrieve the optical diameter of the thick-coated particles BC (marked blue in Fig. R1).

To make this point clear, we have added Fig. R1 into the Sect. 2.2 in the revised manuscript: "*The data processing and analysing procedure in this study are illustrated in Fig. 2.....*"

For non-BC particles, we separated them into two groups according to the upper sizing limit of SP2 (~400 nm): one group is particles at sizes within detection limit (200-400 nm), which were used to retrieve refractive index of non-BC particles (RInon-BC); another group is particles at sizes larger than detection limit, which were used to evaluate the validation of LEO method and explore the possibility of using LEO-fit method to extend the sizing ability of SP2 towards larger size range. LEO-fit

method is commonly used in BC containing particles (thick-coated) to retrieve their optical size and coated thickness. However, we also used the LEO-fit method to non-BC particles (purely scattering particles). In SP2 studies, the peak height of scattering signal is calibrated using size-resolved PSL sphere (purely scattering particles) to obtain particle scattering cross-section, and then to determine their optical size by Mie calculation. However, for scattering particle larger than 400 nm, the scattering signal in SP2 would have been saturated, so the peak height of scattering signal cannot be obtained. This means that the optical size of scattering particles larger than 400 nm cannot be derived by PSL calibration and Mie calculation. That is why the 400 nm was usually taken as the upper sizing limit of SP2. In this study, we used the LEO-fit method to retrieve the peak height of scattering signal for non-BC particles larger than 400 nm (most of doubly or triply charged particles in our study), and then obtained their scattering cross-section using PSL calculation curve, and finally retrieves their optical size by Mie calculation. It is noted that to retrieve optical size by Mie calculation, we need not only the scattering cross-section of non-BC particles but also their reflective index (RI_{non-BC}). In this work, the RI_{non-BC} was retrieved by matching the optical diameter of non-BC particle at 200-400 nm to their electro-mobility diameter (marked red in Fig. S2), which seems to be independent of size anyway (Figure 2 in the manuscript). Therefore, if the LEO-fit method is valid, the optical size of non-BC particles larger than 400 nm should match their mobility diameter.

For thick-coated (internally-mixed) BC particles, we did not tune *RI* to force the agreement (see Fig. R1). The good agreement between the optical diameter derived from the LEO-fit method and the particle mobility diameter selected by the DMA implies that the assumption on a spherical particle in Mie model calculation can be used for thick-coated BC particles in our case. It also revealed that the LEO-fit method was applicable for ambient thick-coated BC particles observed at our site.

To make this point clear, we have mark the signal saturation point (~400 nm) as blue dashed line in Fig. 3 (Fig R2) and revised the discussion on Fig. 3 (Figure 5 in the revised manuscript), as:

"*3.2.1 Non-BC particles with diameter larger than ~400 nm*

The LEO-fit method introduced by Gao et al. (2007) has been commonly used to retrieve the optical size of the thick-coated BC containing particles. In this study, we adopted the LEO-fit method not only to retrieve the peak height of scattering signal for larger non-BC particles with saturated scattering signals (particle size larger than the upper sizing limit of SP2 (~400 nm), most of double or triple charged particles in our study), but also to re-determine the scattering peak heights of non-BC particles (<400 nm) with full Gaussian distribution of the scattering signals detected by SP2. The scattering cross-section can be then obtained according to the calibration of PSL particles with well-defined sizes. Afterwards, the Mie calculation is used to translate the scattering cross-section into optical particle size, where the refractive index of 1.42 is applied for all the non-BC particles (RI_{nonBC} = 1.42 - 0i).

Although we use the DMA1 of the VTDMA selected particles with diameter ranging from 200 to 400 nm, due to multiple charge, particles larger than 400 nm could also been selected with certain probabilities. We thus calculated the size of the large particles with double or triple charges whose mobility size is 200, 250, 300 or 350 nm. In Figure 5A, we show the comparison of the mobility particle size as if they have only single charge and their optical size derived from LEO-fit and Mie calculation for non-BC particles. The agreement of particles with mobility diameter smaller than ~400 nm is of no surprise, because the RI_{non-BC} was retrieved by matching the optical diameter of non-BC particle at 200-400 nm to their electro-mobility diameter (marked red in Fig. 2). However, since the retrieved RI_{non-BC} shows independency on particle diameter, we applied the same refractive index for the particle larger than 400 nm. It can be found that the optical size from 400 nm to ~550 nm particles also agreed very well with their mobility size as if they have only single charge. This demonstrates that the optical diameters of the large particles (from ~400 nm up to ~550 nm) can be still retrieved from the LEO-fit method (Gao, et al., 2007).

3.2.2 Thick-coated BC-containing particles

Similar to the non-BC particles, the optical particle diameter of thick-coating BC particles derived from the LEO-fit method and Mie calculation (Fig. 2) showed an

excellent agreement with their mobility diameter selected by the DMA1 for both single particles and multiple charged large particles, with a difference of ~1% (Fig. 5B). the size of singly charged aerosol particles (200-350 nm, black circles in Fig. 5B) were within the detection limit of SP2 scattering signal, but some double or triple charged aerosol particles (red circles in Fig. 5B) was larger than the upper sizing limit of SP2 scattering signal (~400 nm). Figure 5B shows that the optical size of double charged or triple charged thick-coating BC particles agreed well with their theoretically calculated mobility diameter (323, 414, 438 and 508 nm). The agreement implies that the assumption on a spherical particle in Mie model calculation can be used for thick-coated BC particles in our case. It also revealed that the LEO-fit method was applicable for ambient thick-coated BC particles under polluted conditions in China."
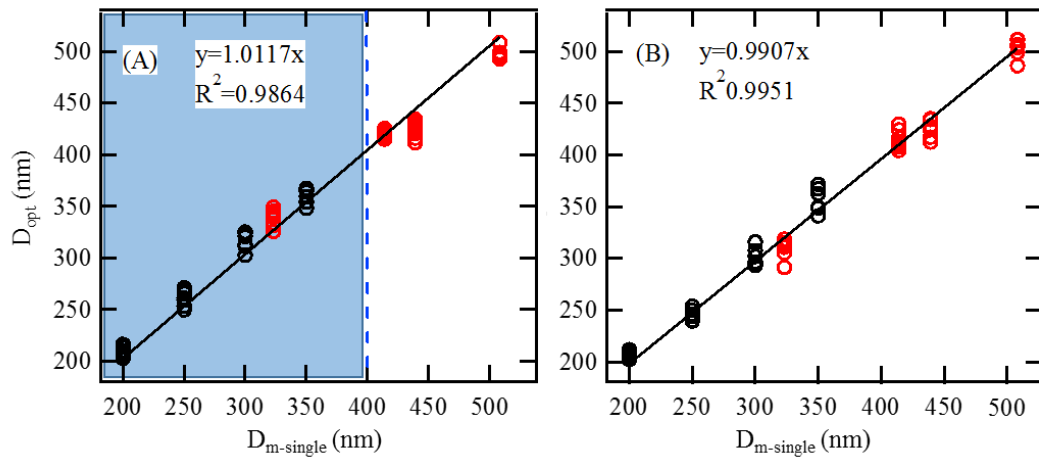

Fig. R2. (Figure 5 in the revised manuscript) Comparison of the mobility particle size as if they have only single charge ($D_{m\text{-}single}$) and their optical size (D_{opt}) derived from LEO-fit and Mie calculation for non-BC particles (A) and thick-coated BC particles (B). The black circles and red circles represent the single charged particle samples and double/triple charged particle samples, respectively. The blue dash line represents the upper limit of SP2 scattering detection (~400 nm). The light blue shade area represents the sizes range within SP2 sizing detection limit for purely scattering particles (i.e., non-BC particles).

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(7) Line 15 in section 3 says, 'The agreement revealed that the LEO method was applicable for ambient aerosol particles." The authors haven't discussed the LEO method enough to enable the reader to evaluate this statement. If the LEO method were valid, what would we see? If it were invalid, what would we see or not see?

Response: Thanks for the comments. We have added more discussion about LEO-fit method in the revised manuscript. Please see details in our response to the Specific Comment (6). For the non-BC particles with diameter larger than 400 nm and the thick-coated BC particles, if the LEO-fit method were valid, we would see an agreement between the retrieved optical diameter and the pre-defined mobility diameter when accounted for the multiple charge effects. If it were invalid, that is the optical diameter does not fit the mobility diameter, it may imply that (1) the refractive index of the large non-BC particles is different from the smaller non-BC particles, (2) the morphology of the large non-BC particles is significantly deviated from 'sphere', or (3) the 'spherical core-shell assumption' in Mie calculation is not applicable for the thick-coated BC particles at our site.

(8) Line 15 also: "It also validated the assumption of a spherical BC particle in the Mie model calculations." I don't think it does anything of the kind. It says that the BC in the particle perhaps doesn't change the scattering very much whether it is spherical or non-spherical.

Response: Thanks. We agree with the reviewer that the particle shape matters more to the side scattering, but not the total scattering. We have changed "It also validated the assumption of a spherical BC particle in the Mie model calculations." into "*The agreement implies that the assumption on a spherical particle in Mie model calculation can be used for the thick-coated BC particles in our case.*"

(9) Figure 4 does not show the fractal dimension of the particles, but they are inferred from the data shown in figure 4.

Response: Thanks. We have changed "Figure 4 shows the fractal dimension and effective density of externally-mixed BC particles observed during the campaign period." into "*Figure 4 shows the mass and effective density of the thin-coated (or bare) BC particles at different mobility diameter observed during the campaign period.*"

(10) Section 3.1.2. I agree that the density of a bare BC particle (aggregate) should be much less than the density of the void-free particle. I don't think anyone has ever suggested that it would have the density of the void-free particle. I am wondering what is the relevance of the density given here (0.25-0.45 g/cm3). Where would it be used.

Response: Thanks to the reviewer to point this out. The effective density (0.25-0.45 g/cm^3) is used to calculate the volume of the thin-coated (or bare) BC particles when BC mass is measured by SP2, and then to calculate their sizes which are shown in Fig.5 (red lines) and Fig. 7 (red lines) in the manuscript. In previous studies (Metcalf et al., 2012; Schwarz et al., 2008; Subramanian et al., 2010), to so such calculation the BC effective density has been assumed to the same as material density of carbon (1.8 g cm^{-3}), which is much larger than the effective density that we derived.

To make this point clear, we have made following changes in the manuscript: "*The effective density of the thin-coated (or bare) BC particles at 200-350 nm observed at the Xianghe Site was found to be very small, ranging from 0.25 to 0.45 g cm^{-3}. It is significantly lower than the material density of carbon, ~1.8 g cm^{-3} (Metcalf et al., 2012; Schwarz et al., 2008; Subramanian et al., 2010). The large discrepancy between the effective density and material density of BC indicated that the thin-coated (or bare) BC particles measured at Xianghe was not a void-free structure. In order to compare with the sizing of VTMDA, we calculated the equivalent mobility diameter of the thin-coated (or bare) BC particles by dividing the rBC mass detected by SP2 by the effective density, which is significantly larger than the one calculated with the*

material density of carbon (see Figure S3)."

(11) Section 3.1.3. I am confused by this section. 1. Some of this information, I think should be in the Methods section. 2. Again there is an assessment of the LEO-fit without too much explanation. It would be useful to see what reader should expect compared to what is found. 3. ": : :which demonstrated the potential of optical sizing instruments: : : in determining the bipolar charge distribution of aerosol particles." Since the theoretical distribution seems to work fine, why do we need the optical sizing instrument? It seems like the authors are just looking for reasons to use the SP2 that are not needed.

Response: Thanks for the comments. In the Section 3.1.3 The size distribution of charged particles (in the manuscript), we aimed to further demonstrate that the size and number of larger particle (most of double or triple charged particles in our study) derived from theoretically calculation can be comparable to those obtained from the SP2 LEO-fit. This further support that the SP2 LEO-fit for larger particles (~400-550 nm) is valid.

To make this point clear, the Section 3.1.3 in the manuscript (The size distribution of charged particles) have been revised and moved into Section 3.2.2 in the revised manuscript, as:

"*To further explore the possibility of using LEO-fit method to extend sizing ability of SP2 towards larger size range, we discussed the number size distribution of charged particles (i.e., single, double or triple charged aerosol particles), which is calculated as the sum of optical particle number size distribution of non-BC particles and thick-coating BC particles, plus the mobility particle number size distribution of thin-coating (or bare) BC particle (Fig. S4A). Most of theoretically calculated diameter of double charged or triple charged aerosols agrees well with the measured diameter (323, 414, 438 and 508 nm) except for the double charged particles with a diameter of 601 nm. The result revealed that the LEO fit used in our study was valid*

for particles with diameters lower than ~550 nm. According to the particle number size distribution, we further calculated the ratio of number concentration between single and double charged particles ($R_{single\text{-}double}$), which was then compared with the theoretical calculated $R_{single\text{-}double}$ values derived from bipolar charged distribution (Wiedensohler, 1988; Wiedensohler et al., 2012) (Fig. S4B). The measured $R_{single\text{-}double}$ showed a good agreement with the theoretically calculated $R_{single\text{-}double}$ with a slope of 0.91 and a R^2 of 0.93, which supported that the LEO-fit for larger particles with saturated scattering signals was valid."

(12) Section 3.2. (comparison VTDMA/SP2) The finding of refractory, non-BC material inthe small particles could be important. But the SP2 has difficulty measuring the small particles (or maybe just different efficiency) because of low scattering. How was this accounted?

Response: Thanks to the reviewer to raise this concern. There might be some misunderstanding. Here, we only compare the number size distribution of refractory residues measured by the DMA2 of VTDMA and the refractory BC detected by SP2 above its incandescence detection limit (>70 nm refractory BC). It is noted that the SP2 data used in Fig. 7 (Figure 9 in the revised manuscript) is based on only incandescence measurements. The number size distribution of refractory BC from SP2 measurement (red lines in Fig. 7 in the manuscript) is corrected for SP2 incandesce particle-counting efficiency according to calibration curve (Fig. R3 in response).

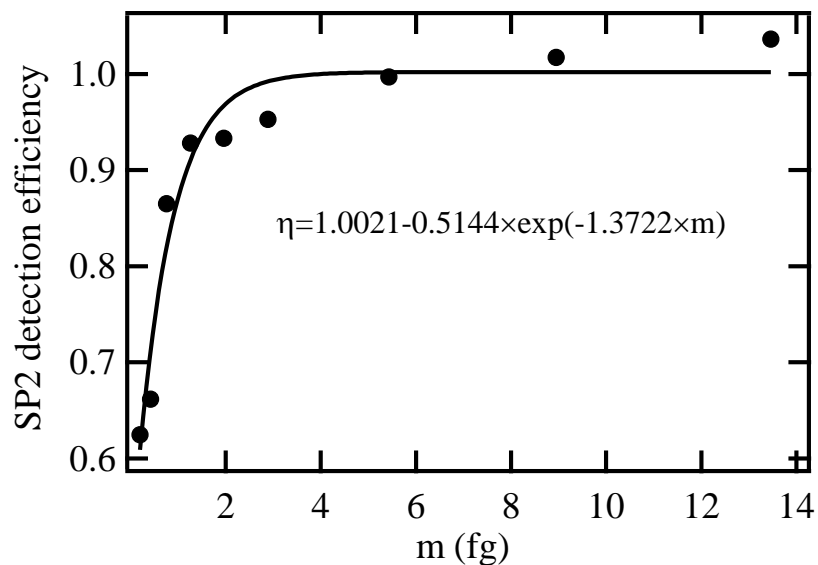$$\eta=1.0021-0.5144\times exp(-1.3722\times m)$$

Fig. R3 (Figure S9 in the revised manuscript). The SP2 detection efficiencies (η) in each BC mass-bin.

To make this point clear, we have mark the lower detection limit of SP2 (~70 nm) as green dashed line in Figure 7 (Fig R4 in response, Figure 9 in the revised manuscript) and the Sect. 3.2 have been rewritten as: "*3.4 Low volatile non-BC materials at 300 °C*

If and how the mixing state will influence the light absorption of BC is still under debate (Cappa et al., 2012; Jacobson, 2001; Liu et al., 2015; Liu et al., 2017). As aforementioned, it is on one hand due to the complex morphology and inhomogeneity of ambient BC-containing particles. On the other hand, the literature reported light absorption enhancement factor is usually measured as the difference in the aerosol light absorption with and without removing the coating materials by heating the aerosol sampling line with thermodenuder at about 200 to 400 °C (Cappa et al., 2012; Liu et al., 2015; Liu et al., 2017). The intrinsic assumption of the method is that all non-BC materials can be removed completed by the thermodenuder at this temperature range and the remaining component is BC. To valid this assumption, we compare the size distribution of BC cores from SP2 with that of non-volatile cores

from VTDMA at 300 ℃ (Figure 9). Here the SP2 detected BC is regarded as a reference of pure refractive BC mass, since SP2 detects it according to the incandescence light emitted at rBC vaporization temperature of ~4000 K. Note that the SP2 detection efficiency (Fig. S9) and experimentally determined effective density (1.2 g cm^{-3} for BC cores of thick-coated BC particles and 0.25-0.45 g cm^{-3} for thin-coated (or bare) BC particles) have been considered in the calculation of the rBC size distribution.

As shown in Fig. 9, the mobility size distribution of rBC measured by SP2 was significantly different with that of the refractory residues at 300 ℃ measured by VTDMA. The SP2 measured one showed two peaks, one at around the prescribed size (200, 250, 300 and 350 nm selected by DMA1) and another one at a smaller size, representing the thin-coating BC (or bare BC) and thick-coating BC particles, respectively. For VTDMA measurement, the number size distribution of low-volatile residues at 300 ℃ also showed a peak at the prescribed size, and other two peaks at smaller sizes, which is similar to the previous measurements in Beijing (Cheng et al., 2012). Above the detection limit of SP2 incandescence (BC particles with diameter larger than ~70 nm), the number concentration of BC particles measured by SP2 accounted for only 16-35% of the residual particles measured by VTDMA. This large discrepancy indicated a large fraction of non-BC low-volatile components, which could not completely evaporate at 300℃ exists in the aerosol particles at Xianghe site, such as low volatile organic matter, refractory mineral dust, trace metals and sea salt (Cheng et al., 2009; Ehn et al., 2014; Kalberer et al., 2004). Furthermore, we found that most of the non-shrinking refractory residues (~78-91%) (the 1st peak of the DMA2) are detected as rBC particles by SP2, while about 80-90% of the residues larger than ~70 nm in the other two peaks (the 2nd and 3rd peaks) of the DMA2 (shrank particles) are not rBC, as they did not show incandescence signal in SP2. In previous studies involving thermodenuder technique, it has often been assumed that the refractory residual materials at 250 to 400 ℃ were BC components (Philippin et al., 2004; Wehner et al., 2009; and Cheng et al., 2009; Cappa et al., 2012; Liu et al.,

2015; Liu et al., 2017). But, this assumption may not hold at the sub-urban site of Beijing (Xianghe), and may cause a significant overestimation of the number of internally mixed BC particles. On the other hand, this assumption may leads to the underestimation of the light absorption enhancement factors in the North China Plain due to the remaining low-volatile non-BC materials coating on the BC particles after heated by the thermodenuder."
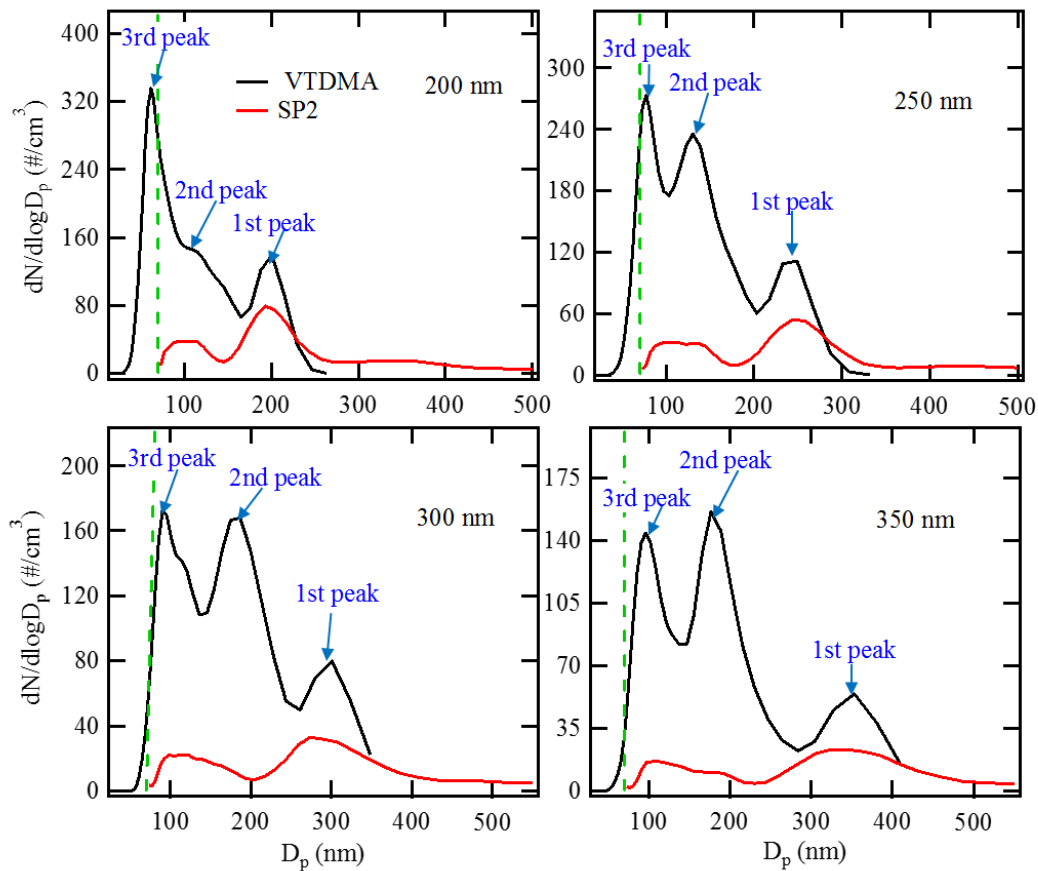

Fig. R4. (Figure 9 in the revised manuscript) Mobility size distributions of the refractory BC measured by SP2 (red lines) and that of the residual materials from VTDMA measurement at 300 ℃ (black lines). The green dash line represents the lower limit of SP2 incandescence detection (~70 nm). The initial particle sizes selected by DMA1 were 200, 250, 300 and 350 nm.

(13) Section 3.3. This discussion equates aging with coated thickness to find that BC containing particles are "more aged" and then associates these with long-range transport. Typically studies refer to some kind of photochemical age (independent measure of aging). It hasn't really been proven here that these particles have been in the atmosphere longer since emission than particles without thick coated. Authors should be clear that their statements are inferences and not proven.

Response: Thanks for the comment. We agree with the reviewer that our statements are not proven. BC is primary aerosol component emitted during combustion process (biomass and fossil fuel burning). BC aging degree (i.e., coated thickness and D_p/D_c ratio) in the atmosphere strongly depends on secondary aerosol production (Metcalf et al., 2013) by photochemical age. The more secondary aerosols (i.e., non-BC species such as SOA, sulfate and nitrate) formed in the atmosphere the higher BC aging degree and thicker coated. BC aging process may occur locally or during regional transport. In this study, we cannot distinguish between them.

We have changed "long range transport" into "secondary process" and in manuscript it reads as "*The smaller contribution of BC as a primary component, revealed that the aerosol particles sampled at our site were likely undergone a strong secondary aging process.*"

(14) thick coated: Authors should be clear about what they mean by "thick coated." I could not find a definition. They identify coateds as thick and then say this will lead to lensing and enhanced absorption. But a x3 increase in mass due to coated is only 40% increase in radius and 30-40% increase in absorption. Claims about strong enhancement, would be better if some physical basis were discussed. I also think the number of uncoated particles (what's called 'externally mixed' in the paper) should be shown here. Fig 9 suggests that the bare particles would actually be the largest fraction.

Response: Thanks for the suggestion. In this work, "thick coated" represents the coated amount of BC-containing particles with lagtime no less than 1.6 μs in SP2 measurement. For our SP2 measurement, we found a bimodal distribution of lagtime (Fig. R5). Here we adopted a classification method based on a threshold lagtime/coated thickness) which is commonly used previous studies (e.g., Schwarz et al., 2006; Sedlacek et al., 2012; Subramanian et al., 2010). In literature, the two peaks were always observed and considered as the thick-coated BC and thin-coated BC, respectively.

According to reviewer's suggestion, we have clarified about "thick coated" BC in the revised manuscript as "*As shown in Fig. S1, the lagtime (Δt), namely the time difference between the occurrences of the peak of the incandescence and scattering signals measured by SP2 (Moteki and Kondo, 2007; Schwarz et al., 2006; Sedlacek et al., 2012; Subramanian et al., 2010), exhibits a clear two-mode distribution. Accordingly, we further classified the BC-containing particles into thin-coated (or bare) BC (Δt <1.6 μs) and thick-coated BC (Δt >=1.6 μs).*"
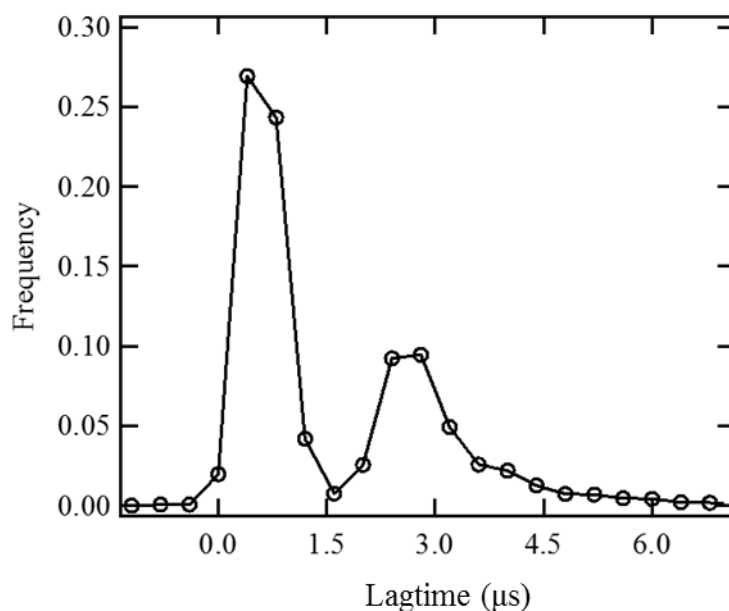

Fig. R5. (Figure S1 in the revised manuscript) The distribution of lagtime in our SP2 measurement.

According to the second suggestion, we have added some discussion on

absorption enhancement caused by thick-coated BC particles in the revised manuscript: "*Aged BC particles with thick coating under the polluted conditions in China may imply that the light absorption of the thick-coated BC particles observed in the North China Plain could be significantly enhanced by lensing effects (Fuller et al., 1999; Lack and Cappa, 2010; Liu et al., 2015; Moffet et al., 2009). Although, the actual magnitude of absorption enhancements of BC aerosol in the atmosphere remains unresolved due to complex morphology and inhomogeneity of ambient BC-containing particles (Adachi et al., 2013; Cappa et al., 2012; Fuller et al., 1999; He et al., 2015; Liu et al., 2015; Liu et al., 2017), the good agreement between LEO-fit retrieved optical particle diameter and mobility one for the thick-coated BC particles (Fig. 5B) suggested that the spherical core-shell assumption may be applicable in our case. Based on this assumption in the Mie simulation, the light absorption enhancement caused by the thick coatings on BC particles was ~1.8-2.1 during our campaign period.* "*

According to the last suggestion, the number fraction of thin-coated (or bare) BC particles (what's called 'externally mixed' in the paper) is also shown in Fig. 10 in the manuscript (Fig. R6 in response; Figure. S4 in the revised manuscript). Correspondingly, we also revised the discussion on Figure 10 (Figure S5 in the revised manuscript): "*Although 60-70% of the BC-containing particles at 200-350 nm have a D_p/D_c ratio in the range of 1-2 (Fig. S5), which is consistent with large fraction (49-62%) of thin-coated (or bare) BC particles, on average, the D_p/D_c ratio of all BC-containing particles at 200-350 nm can still reach to 1.5-2.4. This indicates that the thick-coated BC particles, distinguished as the second mode according to the 'lagtime' (Fig.S1), were quite aged with significantly amount of coating materials on them.*"
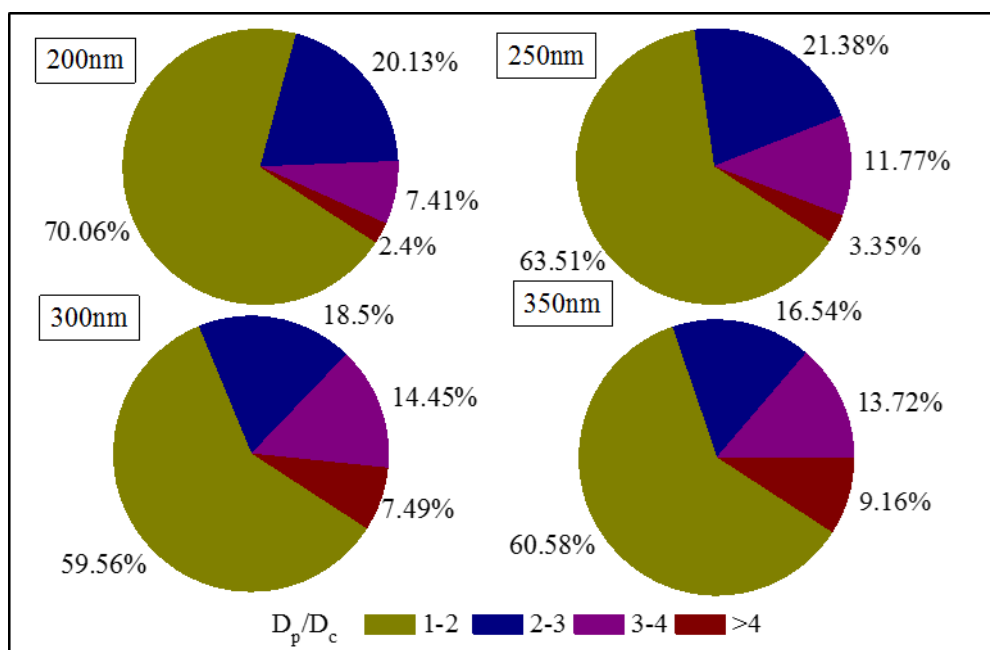
Fig. R6. (Figure S5 in the revised manuscript) The number fraction of different D_p/D_c ratio for BC-containing particles with DMA1 selecting size (200-350 nm).

(15) Figure 10 & figure 11. I don't understand a discrepancy here. Figure 10 shows (as stated in the text) that the mode of Dp/Dc (particle to core) ratio is 2-3 and many of the ratios are greater than 3. But I don't see particles with such large Dp/Dc in figure 11. Dp/Dc = 3 means that coated thickness is 2x core radius. Fig 11 shows that hardly any coated thicknesses are 2x greater than the core radius. Anyway I think that fig 11 would be easier to understand if radius were on the x axis.

Response: Thanks for the comment. As shown in Fig. 11 in the manuscript (Fig. R7 in response), for BC particles selected by DMA1, the coated thickness (CT) was around theoretic value (i.e., $CT=(D_{selected}-D_{core})/2$; dashed lines in Fig. R7). In this study, the particle size (D_p) selected by DMA1 was 200-350 nm with single charge (white dashed lines in Fig. R7), accompanied with double and triple charged particles (black and gray dashed lines in Fig. R7) at 300-600 nm. Figure 11 in the manuscript shows that core diameters of thick-coated particles (D_c) have a distribution with mode at 80-160 nm, meaning that mode of D_p/D_c ratio is 2-3 and many of the ratios are greater than 3. Therefore, the results shown in Fig. 11 (in the manuscript) consistent

with that in Fig. 10 (in the manuscript). Because the size of BC cores was characterized by diameter (D_c) in above discussion, we still use core diameter in Fig. 11 rather than core radius to keep consistent throughout the manuscript. We have moved Fig. 11 to Supplement (Fig. S7).
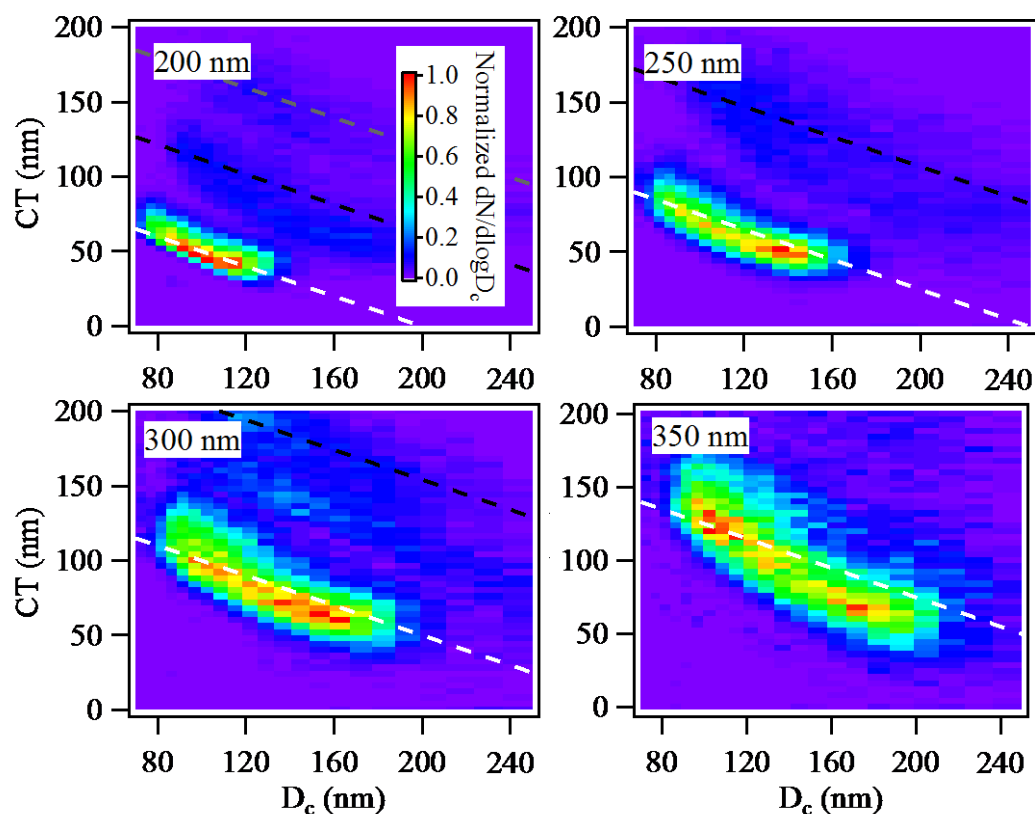

Fig. R7 (Figure S8 in the revised manuscript). Coated thickness (*CT*) of thick-coated BC particles at 200-350 nm (selected by DMA1) as a function of rBC core size (*Dc*). The dash line is $CT=(D_{selected}-D_c)/2$, i.e., the theoretic *CT*; the white, black and grey colors reflect the single, double and triple charged particles.

(16) Figure 11 description, "The CT of internally-mixed BC particles decreased as Dc increased: : : " Isn't this required from the way the figure was made? The DMA selects one diameter. That diameter is just the core + coated.

Response: Thanks for the comment. Fig. 11 (Fig. R7 in response) provides additional information on the relation between coated thicknesses and core size. For a certain

core, the coated thickness in Fig. 11 shows a wide range rather than a value of $CT=(D_{selected}-D_{core})/2$, because the size of the particles selected by DMA still exhibited a distribution. Moreover, Figure 11 also shows the modes of coated thickness for the doubly and triply charged particles (black and gray dashed lines in Fig. R7).

Since Fig. 11 gives some information of the influence of DMA transfer function and multiple charged particles on the distribution of *CT*, we decided to move it to Supplement (Fig. S8). We have modified Fig. 11. The discussion on Fig. 11 have been revised in the manuscript "*Figure S8 shows the coating thickness of thick-coated BC particles as a function of rBC core size (D_c). For a certain core size, the coating thickness in Fig. S7 shows a wide distribution rather than a value of $CT=(D_{selected}-D_c)/2$ (here, $D_{selected}$ is the mobility diameter selected by DMA1), because the size of the particles selected by DMA1 still exhibited a distribution determined by the DMA transfer function. For the single charged particles (white dashed lines in Fig. S8) at 200-350 nm, the D_c and CT showed a wide mode, which were in the range of ~80-200 nm and ~50-150 nm, respective. Moreover, Figure S8 also shows the modes of coating thickness from the double and triple charged particles (black and gray dashed lines in Fig. S8). The wide range of D_c and CT revealed that the thick-coated BC particles at our site consisted of a mixture of BC from various sources of the North China Plain. The more coatings on BC the longer and/or faster secondary process the BC-containing particles underwent.*"

(17) The final statement, about "importance of BC aerosols in polluted regions in China to climate change", is not supported. Perhaps it is true, but although the absorption may be a little stronger (per mass) than in developed countries, it does not make the aerosols hugely more important. Furthermore, no inferences about climate change were calculated, such as radiative forcing.

Response: Thanks to the reviewer to point this out. We have deleted "Our results suggested the importance of BC aerosols in polluted regions (e.g., Jing-Jin-Ji region) in China to climate change", and added "*Aged BC particles with thick coating under the polluted conditions in China may imply that the light absorption of the*

thick-coated BC particles observed in the North China Plain could be significantly enhanced by lensing effects (Fuller et al., 1999; Lack and Cappa 2010; Liu et al. 2015; Moffet et al., 2009)." in revised manuscript.

References:

Adachi, K., and Buseck, P. R.: Changes of ns-soot mixing states and shapes in an urban area during CalNex, *J. Geophys. Res.: Atmos.*, 118, 3723-3730, 2013.

Bohren, C. F., and Huffman, D. R.: Frontmatter, in: Absorption and Scattering of Light by Small Particles, *John Wiley*, New York, NY, 1983.

Cappa, C. D., Onasch, T. B., Massoli, P., Worsnop, D. R., Bates, T. S., Cross, E. S., Davidovits, P., Hakala, J., Hayden, K. L., Jobson, B. T., Kolesar, K. R., Lack, D. A., Lerner, B. M., Li, S. M., Mellon, D., Nuaaman, I., Olfert, J. S., Petaja, T., Quinn, P. K., Song, C., Subramanian, R., Williams, E. J., and Zaveri, R. A.: Radiative Absorption Enhancements Due to the Mixing State of Atmospheric Black Carbon, *Science*, 337 (6098), 1078-1081, 2012.

Cheng, Y., Berghof, M., Garland, R. M., Wiedensohler, A., Wehner, B., Müller, T., Su, H., Zhang, Y., Achert, P., Nowak, A., Pöschl, U., Zhu, T., Hu, M., and Zeng, L.: Influence of soot mixing state on aerosol light absorption and single scattering albedo during air mass aging at a polluted regional site in northeastern China, *J. Geophys. Res.*, 114, (D10), D010883, 2009.

Cheng, Y. F., Eichler, H., Wiedensohler, A., Heintzenberg, J., Zhang, Y. H., Hu, M., Herrmann, H., Zeng, L. M., Liu, S., Gnauk, T., Brüggemann, E., and He, L. Y.: Mixing state of elemental carbon and non-light-absorbing aerosol components derived from in situ particle optical properties at Xinken in Pearl River Delta of China, *J. Geophys. Res.-Atmos.*, 111, 2006.

Cheng, Y. F., Su, H., Rose, D., Gunthe, S. S., Berghof, M., Wehner, B., Achtert, P., Nowak, A., Takegawa, N., Kondo, Y., Shiraiwa, M., Gong, Y. G., Shao, M., Hu, M., Zhu, T., Zhang, Y. H., Carmichael, G. R., Wiedensohler, A., Andreae, M. O., and Pöschl, U.: Size-resolved measurement of the mixing state of soot in the megacity Beijing, China: diurnal cycle, aging and parameterization, *Atmos. Chem. Phys.*, 12, 4477-4491, 2012.

Chung, S. H., and Seinfeld, J. H.: Climate response of direct radiative forcing of anthropogenic black carbon, *J. Geophys. Res.-Atmos.*, 110, 2005.

Ehn, M., Thornton, J. A., Kleist, E., Sipila, M., Junninen, H., Pullinen, I., Springer, M., Rubach, F., Tillmann, R., Lee, B., Lopez-Hilfiker, F., Andres, S., Acir, I.-H., Rissanen, M., Jokinen, T., Schobesberger, S., Kangasluoma, J., Kontkanen, J., Nieminen, T., Kurten, T., Nielsen, L. B., Jorgensen, S., Kjaergaard, H. G., Canagaratna, M., Maso, M. D., Berndt, T., Petaja, T., Wahner, A., Kerminen, V.-M., Kulmala, M., Worsnop, D. R., Wildt, J., and Mentel, T. F.: A large source of low-volatility secondary organic aerosol, *Nature*, 506, 476-479, 2014.

Fuller, K. A., Malm, W. C., and Kreidenweis, S. M.: Effects of mixing on extinction by carbonaceous particles, *J. Geophys. Res.-Atmos.*, 104, 15941-15954, 1999.

Gao, R. S., Schwarz, J. P., Kelly, K. K., Fahey, D. W., Watts, L. A., Thompson, T. L., Spackman, J. R., Slowik, J. G., Cross, E. S., Han, J. H., Davidovits, P., Onasch, T. B., and Worsnop, D. R.: A Novel Method for Estimating Light-Scattering Properties of Soot Aerosols Using a Modified Single-Particle Soot Photometer, *Aerosol Sci. Technol.*, 41, 125-135, 2007.

Gustafsson, Ö., and Ramanathan, V.: Convergence on climate warming by black carbon aerosols, *Proc. Natl. Acad. Sci*. USA., 113, 4243-4245, 2016.

He, C., Liou, K. N., Takano, Y., Zhang, R., Levy Zamora, M., Yang, P., Li, Q., and Leung, L. R.: Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison, *Atmos. Chem. Phys.*, 15, 11967-11980, 2015.

Kalberer, M., Paulsen, D., Sax, M., Steinbacher, M., Dommen, J., Prevot, A. S. H., Fisseha, R., Weingartner, E., Frankevich, V., Zenobi, R., and Baltensperger, U.: Identification of Polymers as Major Components of Atmospheric Organic Aerosols, *Science*, 303, 1659-1662, 2004.

Laborde, M., Crippa, M., Tritscher, T., Jurányi, Z., Decarlo, P. F., Temime-Roussel, B., Marchand, N., Eckhardt, S., Stohl, A., Baltensperger, U., Prévôt, A. S. H., Weingartner, E., and Gysel, M.: Black carbon physical properties and mixing state in the European megacity Paris, *Atmos. Chem. Phys.*, 13, 5831-5856, 2013.

Lack, D. A. and Cappa, C. D.: Impact of brown and clear carbon on light absorption

enhancement, single scatter albedo and absorption wavelength dependence of black carbon, *Atmos. Chem. Phys.*, 10, 4207–4220, 2010.

Liu, D., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, D. V., Reddington, C. L., Kong, S., Williams, P. I., Ting, Y.-C., Haslett, S., Taylor, J. W., Flynn, M. J., Morgan, W. T., McFiggans, G., Coe, H., and Allan, J. D.: Black-carbon absorption enhancement in the atmosphere determined by particle mixing state, *Nature Geosci*, 10, 184-188, 2017.

Liu, S., Aiken, A. C., Gorkowski, K., Dubey, M. K., Cappa, C. D., Williams, L. R., Herndon, S. C., Massoli, P., Fortner, E. C., Chhabra, P. S., Brooks, W. A., Onasch, T. B., Jayne, J. T., Worsnop, D. R., China, S., Sharma, N., Mazzoleni, C., Xu, L., Ng, N. L., Liu, D., Allan, J. D., Lee, J. D., Fleming, Z. L., Mohr, C., Zotter, P., Szidat, S., and Prévôt, A. S. H.: Enhanced light absorption by mixed source black and brown carbon particles in UK winter, *Nat Commun.*, 6, 8435, 2015.

Metcalf, A. R., Craven, J. S., Ensberg, J. J., Brioude, J., Angevine, W., Sorooshian, A., Duong, H. T., Jonsson, H. H., Flagan, R. C., and Seinfeld, J. H.: Black carbon aerosol over the Los Angeles Basin during CalNex, *J. Geophys. Res.-Atmos.*, 117, 2012.

Metcalf, A. R., Loza, C. L., Coggon, M. M., Craven, J. S., Jonsson, H. H., Flagan, R. C., and Seinfeld, J. H.: Secondary Organic Aerosol Coated Formation and Evaporation: Chamber Studies Using Black Carbon Seed Aerosol and the Single-Particle Soot Photometer, *Aerosol Sci. Technol.,* 47, 326-347, 2013.

Moffet, R. C. and Prather, K. A.: In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates, *P. Natl. Acad. Sci. USA*, 106, 11872–11877, 2009.

Oshima, N., Koike, M., Zhang, Y., Kondo, Y., Moteki, N., Takegawa, N., and Miyazaki, Y.: Aging of black carbon in outflow from anthropogenic sources using a mixing state resolved model: Model development and evaluation, *J. Geophys. Res.: Atmos.*, 114, 2009.

Philippin, S., Wiedensohler , A., and Stratmann, F.: Measurements of non-volatile fractions of pollution aerosols with an eight-tube volatility tandem differential mobility analyzer (VTDMA-8), *Aerosol Sci.*, 35, 185-203, 2004.

Ramanathan, V., and Carmichael, G.: Global and regional climate changes due to

black carbon, *Nature Geosci,* 1, 221-227, 2008.

Schwarz, J. P., Gao, R. S., Spackman, J. R., Watts, L. A., Thomson, D. S., Fahey, D. W., Ryerson, T. B., Peischl, J., Holloway, J. S., Trainer, M., Frost, G. J., Baynard, T., Lack, D. A., de Gouw, J. A., Warneke, C., and Del Negro, L. A.: Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions, *Geophys. Res. Lett.*, 35, 2008.

Schwarz, J. P., Gao, R. S., Fahey, D. W., Thomson, D. S., Watts, L. A., Wilson, J. C., Reeves, J. M., Darbeheshti, M., Baumgardner, D. G., Kok, G. L., Chung, S. H., Schulz, M., Hendricks, J., Lauer, A., Kärcher, B., Slowik, J. G., Rosenlof, K. H., Thompson, T. L., Langford, A. O., Loewenstein, M., and Aikin, K. C.: Single-Particle Measurements of Midlatitude Black Carbon and Light-Scattering Aerosols from the Boundary Layer to the Lower Stratosphere, *J. Geophys. Res.: Atmos*., 111 (D16), D16207, 2006.

Schwarz, J. P., Spackman, J. R., Fahey, D. W., Gao, R. S., Lohmann, U., Stier, P., Watts, L. A., Thomson, D. S., Lack, D. A., Pfister, L., Mahoney, M. J., Baumgardner, D., Wilson, J. C., and Reeves, J. M.: Coateds and their enhancement of black carbon light absorption in the tropical atmosphere, *J. Geophys. Res.: Atmos*., 113, 2008.

Sedlacek, A. J., Lewis, E. R., Kleinman, L., Xu, J., and Zhang, Q.: Determination of and evidence for non-core-shell structure of particles containing black carbon using the Single-Particle Soot Photometer (SP2), *Geophys. Res. Lett.*., 39 (L06802), L050905, 2012.

Subramanian, R., Kok, G. L., Baumgardner, D., Clarke, A., Shinozuka, Y., Campos, T. L., Heizer, C. G., Stephens, B. B., de Foy, B., Voss, P. B., and Zaveri, R. A.: Black carbon over Mexico: the effect of atmospheric transport on mixing state, mass absorption cross-section, and BC/CO ratios, *Atmos. Chem. Phys*., 10, 219-237, 2010.

Wehner, B., Berghof, M., Cheng, Y. F., Achtert, P., Birmili, W., Nowak, A., Wiedensohler, A., Garland, R. M., Pöschl, U., Hu, M., and Zhu, T.: Mixing state of nonvolatile aerosol particle fractions and comparison with light absorption in the polluted Beijing region, *J. Geophys. Res*., 114 (D17): 2009.

Wiedensohle, A., and Fissan, H. J.: Bipolar Charge Distributions of Aerosol Particles in High-Purity Argon and Nitrogen, *Aerosol Sci. Technol.*, 358-364, 1991.

Zhang, Y., Zhang, Q., Cheng, Y., Su, H., Kecorius, S., Wang, Z., Wu, Z., Hu, M., Zhu, T., Wiedensohler, A., and He, K.: Measuring the morphology and density of internally mixed black carbon with SP2 and VTDMA: new insight into the absorption enhancement of black carbon in the atmosphere, Atmos. Meas. Tech., 9, 1833-1843, 2016b.