Anonymous Referee #3:

The authors investigated the evolution of BC optical properties, and concluded that under more polluted conditions, the aging process will enhance the coating of BC-containing particles and thus contribute to larger enhancement of BC particle light absorption. They further claim that pollution control strategy will have co-benefit effects on both air quality and climate. The content is suitable for publication within the scope of ACP, while some revisions are required. Please see detailed comments below.

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, and the revision in the manuscript is further formatted as '*Italics*'.

1. Major issues

(1) The manuscript is still in need of a better discussion on uncertainties. Some examples are listed below, while I would suggest the authors do a systematic discussion on all the associated uncertainties, not just here and there.

Response: We thank the reviewer for raising the important issue. Following the reviewer's suggestion, we have systematically discussed all the associated uncertainties. Here, to the uncertainties mentioned by the reviewer is as follows.

(a) Page 4, lines 1-3: The authors mentioned the correction to Aethalometer data in the SI, where there is something confusing to me. First, the authors said they retrieved the correction factor by comparing absorption coefficients measured by AE and MAAP, but since AE was measuring at 660 nm while MAAP at 670 nm,

are the authors just neglecting the difference? Second, the authors used an average value of 2.6 for all their AE measurements while they did determine a pretty wide range of the C, from 1.9 to 4.0, then how did the authors decide the uncertainty of 10% confidently?

Response: We thank the reviewer for raising this question. For the first question, we have discussed the uncertainty from the inconsistence of wavelength for AE and MAAP measurements. Considering that the absorption is inversely proportional to wavelength (*Bond and Bergstrom, 2006*), the difference of wavelength for AE (at 660 nm) and MAAP (at 670 nm) measurements, would lead to an uncertainty of ~1.5% for the corrected absorption coefficients in AE measurement. The related statement has been added in the revise supplement, as "*Noted that the AE and MAAP measurements used to calculate the factor C were at different wavelengths, namely 660 nm and 670 nm, respectively. Considering that the absorption is inversely proportional to wavelength (Bond and Bergstrom, 2006), the difference in wavelength would lead to an uncertainty of ~1.5% for the corrected absorption coefficients in AE measurement.*"

To the second question, we have reestimated the uncertainty related to the factor *C*. The uncertainty in the factor *C* of AE measurements obtained in our study was dominated by the uncertainty in MAAP measurements. In this study, we corrected the MAAP data using the algorithm reported by Hyvärinen et al. (2013). Hyvärinen et al. (2013) compared the results from a PAS against those derived from the MAAP in Beijing, and estimated the uncertainty of ~15% in absorption coefficients derived from MAAP based on the developed algorithm. Therefore, we estimated that the factor *C* derived by comparing AE and MAAP measurement would exhibit an uncertainty of ~15%. The related statement has been revised as "*In this study, the uncertainty in the factor C was dominated by the uncertainty in MAAP measurements. We corrected the MAAP data using the algorithm reported by Hyvärinen et al. (2013). They estimated that the uncertainty in absorption coefficients derived from MAAP based on the developed algorithm was ~15% by comparing the results from a PAS against those derived from the MAAP in Beijing.*

This indicated that the factor C used in our study (~2.6) would exhibit an uncertainty of ~15% from the uncertainty in MAAP measurements."

(b) Page 4, line 26: the authors used 1.50-0i as the RIs value, is there any reason why? Is there some information on, e.g., the chemical compositions of the coating materials, to support that the use of 1.50-0i is reasonable? Otherwise I would suggest the authors consider some sensitivity test on RIs values as well as on RIc.

Response: Thanks for the comment. Following the reviewer's suggestion, we have demonstrated that the use of 1.50-0i for the refractive index of coating materials based on their chemical compositions during the campaign period (Fig. R1 in the response and new Fig. S4 in the revised manuscript). It is known from the literature (Schkolnik et al., 2007; Mallet et al., 2003; Marley et al., 2001) that major inorganic components of ambient aerosol from urban emission (nitrate, sulfate, mineral dust, sea salt and trace metal) have a refractory of (1.5-1.6)-0i and there is a range of (1.4-1.5)-0i for the refractory of organic components. In this study, we used the values of 1.55-0i and 1.45-0i as refractive indexes of inorganic and organic components of coating materials. The components of coating materials was similar to non-refractory compositions in PM_1 particles (Peng et al., 2016). Figure R1 (new Fig. S4 in the revised manuscript) shows that the fraction of inorganic and organic components in coating materials are ~51% and ~49%, respectively. The refractive index of a mixture particle can be calculated as the volume weighted average of the refractive indices of all components (Hänel, et al. 1968; Marley et al., 2001; Bond and Bergstrom, 2006; Schkolnik et al., 2007), as $\widetilde{m} = \sum_i \widetilde{m}_i c_i$, where $\widetilde{m}$ is the refractive index of a mixture particle; $\widetilde{m}_i$ is the refractive index of particle species; c is the volume ratio of particle species. Based on the equation, the refractive index of coating materials of BC-containing particles (RI_s) was ~1.50-0i during the campaign period.

On the other hand, we have considered some sensitivity test on the refractive index of rBC cores (RI_c), see the statement in page 4 line 26-28 in the manuscript and Fig S3 (new Fig S4 in the revised supplement) in the supplement.

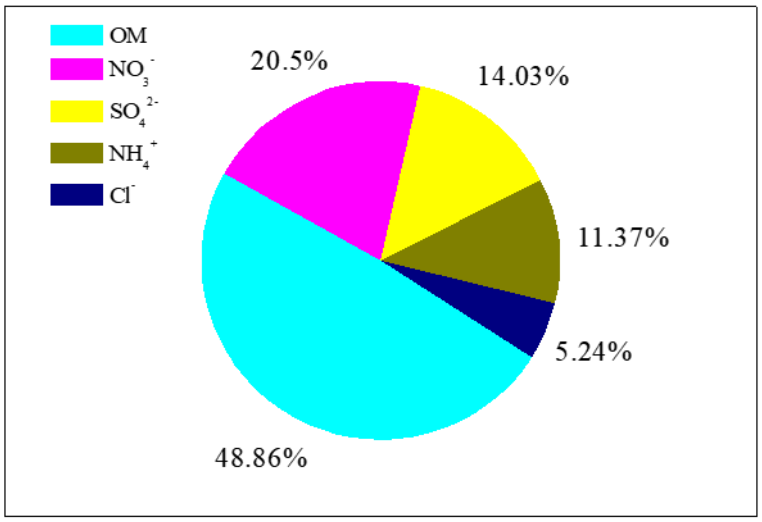

Figure R1 (Fig. S4 in the revised manuscript). Non-refractory compositions of PM_1 particles during the campaign period.

To make this point clear, we have added Fig. S2 and the related discussion in the revised manuscript, as "*The RI_s value used in this study are 1.50-0i based on the chemical compositions of coating materials during the campaign period. The components of coating materials was similar to non-refractory compositions in PM1 particles (Peng et al., 2016). Figure S4 reveals that the fraction of inorganic and organic components in coating materials of BC-containing particles are ~51% and ~49%, respectively. It is known from the literature (Schkolnik et al., 2007; Mallet et al., 2003; Marley et al., 2001) that major inorganic components of ambient aerosol from urban emission (nitrate, sulfate, mineral dust, sea salt and trace metal) have a refractory of (1.5-1.6)-0i and there is a range of (1.4-1.5)-0i for the refractory of organic components. In this study, we used the values of 1.55-0i and 1.45-0i as refractive indexes of inorganic and organic components of coating materials. The refractive index of a mixture particle can be calculated as the volume weighted average of the refractive indices of all components (Hänel, et al. 1968; Marley et al., 2001; Bond and Bergstrom, 2006; Schkolnik et al., 2007), as $\tilde{m} = \sum_i \tilde{m}_i c_i$, where $\tilde{m}$ is the refractive index of a mixture particle; $\tilde{m}_i$ is the refractive index of particle species; c is the volume ratio of particle species. Based*

on the equation, the refractive index of coating materials of BC-containing particles (RI_s) was ~1.50-0i during the campaign period. ”

(c) Page 8, line 3: I am not sure how the authors determine that the Mie calculation has an uncertainty “smaller than 7%”. The authors have shown in Figure S3 and the associated discussions that different RI values could result in 3%-10% difference in Dc. Assume it is on average 5%, then the mass concentration of rBC would be different by 16% (1.05^3, the cubic is converting from diameter to volume), not mentioning the uncertainties on the estimation of e.g., density, mixing, etc. I would suggest the authors do a much more careful job when they are talking about uncertainties.

Response: Thanks for the comments. There might be some misunderstanding on the uncertainty of Mie calculation given here. We would like to kindly clarify that the uncertainty of 3%-10% shown in Fig. S3 from different RI_c values in Mie calculation was for the whole diameter of BC-containing particles (D_p), not for diameter of rBC core (D_c). We did not use Mie calculation to determine the mass concentration of rBC, which was obtained from SP2 measurements. Therefore, the mass concentration of rBC would not be different due to using different RI_c values. We have recalculated the uncertainties on the calculated light absorption. In this study, the MAC for bare BC derived from Mie calculation, using the RI (i.e., 2.26-1.26i) given here, is 3.5-4.4 m^2/g at 880 nm (Fig. R2 in the response). Bond and Bergstrom (2006) suggested a value of 7.5 m^2/g for the MAC of bare BC at 550 nm. Considering that the absorption is inversely proportional to wavelength (Bond and Bergstrom, 2006), the MAC of bare BC at 880 nm is estimated to be ~4.7 m^2/g, which was slightly greater than that (~4.3 m^2/g) obtained from Mie calculation in our study. This indicated the uncertainty of MAC for bare BC from Mie calculation was ~8%. We estimated that the uncertainties of calculated BC light absorption related to MAC of bare rBC from Mie calculation was ~8%.

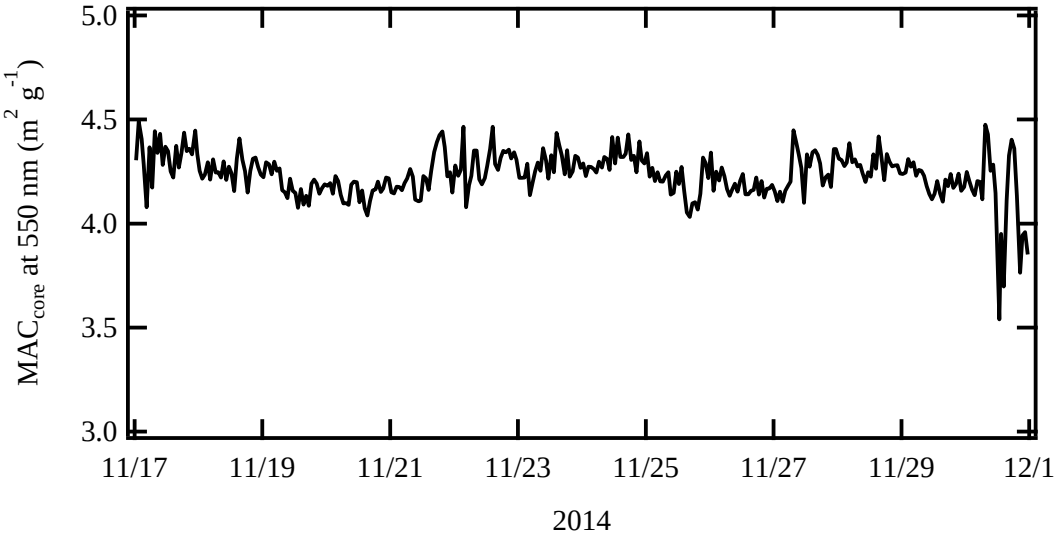

Figure R2 (Fig. S7 in the revised manuscript). The time series of MAC derived from Mie calculation for BC cores (i.e., bare BC) at 880 nm.

Correspondingly, we added the new Fig. S7 and related discussion in the revised supplement, as "*Based on Mie calculation, we obtained the MAC of rBC core (MAC_c) at 880 nm in the range of 3.8-4.5 m^2/g with an average of ~4.3 m^2/g during the campaign period (Fig. S9). Bond and Bergstrom (2006) suggested a value of 7.5 m^2/g for the MAC of bare BC at 550 nm. Considering that the absorption is inversely proportional to wavelength (Bond and Bergstrom, 2006), the MAC of bare rBC at 880 nm is estimated to be ~4.7 m^2/g, which was slightly greater than that (~4.3 m^2/g) obtained from Mie calculation in our study. This indicated the uncertainty of MAC for bare rBC from Mie calculation was ~8%. We estimated that the uncertainties of calculated BC light absorption related to MAC of bare rBC from Mie calculation was ~8%.*"

(2) About the processes contributing to the enhancement of BC light absorption. The authors are trying to add some discussions on the causes of BC coating and thus light absorption enhancement, but these discussions read somewhat weird if there is no sufficient evidence to support. Similarly, a couple of examples below:

Response: Thanks for the comment. Following the reviewer's suggestion, we

have modified some discussion on the causes of BC coating and thus light absorption enhancement in the revised manuscript to make them more appropriate. Here, to the discussions/statements mentioned by the reviewer is as follows.

(a) Page 8, line 18: "due to more secondary component formation", is it possible that more primary components were also emitted under the more polluted condition and coated onto the BC core during the aging process?

Response: Thanks to the reviewer for raising this concern. We agree with the reviewer. The statement has been revised as "*In terms of BC-containing particles with a certain rBC core size, their D_p/D_c ratio and E_{ab} were greater under higher PM_1 concentrations, which could be attributed to more coating materials on BC surface under more pollution environment. The increase of both primary and secondary components under more polluted conditions was favorable to BC aging by coagulation and condensation, which happen mostly between BC and non-BC species.*"

(b) Page 10, lines 19-28: I do not understand why the authors are looking at the temporal trend of O3 to evaluate local photochemical processes. The trend of O3 could mean weak production, could mean strong ozonolysis (which could be dark reaction, i.e., nothing with photochemistry), or could just mean cloudy days thus no sunlight. This is not a sound reason for "weak local aging".

Response: Thanks to the reviewer to point this out. We agree with the reviewer that the decrease in O3 concentration can not fully support weaken local aging. In this study, we focused on the effect of regional transport on BC aging process. We just roughly discussed the chemical process during BC aging. The chemical process of BC aging under polluted environment in china is complex, which involved photochemical oxidation and heterogeneous chemical production (Zheng et al. 2015). We will investigate the chemical process of BC aging under polluted environment in future.

In the revised manuscript, we have toned down the related discussion on the

chemical process of BC aging, as "*When PM_1 concentrations were higher than ~120 μg m^{-3}, O_3 concentrations decreased to ~2 ppb. Zheng et al. (2015) has demonstrated the weakened importance of photochemistry in the production and aging of secondary aerosols in Beijing under polluted conditions due to decrease of oxidant concentrations. This indicated that the photochemical processing in BC aging may be weakened under higher polluted levels (i.e., PM_1>120μg m^{-3}). Noted that photochemical processing is not the only possible pathway in BC aging process and other pathways were not discussed in this study. For example, high concentrations of aerosols under polluted environment may compensate the adverse photochemical conditions for BC aging.*"

2. Minor issues

(1) The authors sometimes used "BC-containing particles" while sometimes "BC particles" and "BC" to name the same term, the BC-cored and other materials coated particles. Please try to be consistent throughout the manuscript, otherwise it will be confusing, e.g., Page 2, the "BC" of line 12, and the first "BC" of line 22, they did not actually have the same meaning.

Response: Thanks for the comment. Throughout the manuscript, we have revised the terms to keep them consistent.

(2) Page 1, line 14: It "is" well known…

Response: Thanks. We have revised it.

(3) Page 2, line 5: both emissions of BC and the coating materials -> emissions of both BC and the coating materials;

Response: Thanks. We have revised it.

(4) Page 2, line 22: lens effect -> lensing effect. Same problem throughout the paper, e.g., Page 5, line 24, and Page 8, line 10, etc.

Response: Thanks. We have revised them.

(5) Page 2, line 23: results -> result;

Response: Thanks. We have revised it.

(6) Page 3, line 25: the particles were not "collected" by the diffusion dryer, please correct;

Response: Thanks for the comment. We have revised the sentence as "*Ambient aerosol particles were collected by a PM_1 cyclone and then passed through a diffusion silica gel dryer…..*"

(7) Page 4, line 25: not "RIs and RIc", here it should be RIs only.

Response: Thanks. We have revised it.

(8) Page 5, line 10: underestimate -> underestimation;

Response: Thanks. We have revised it.

(9) Page 5, equation (4) and equation (6): what is the difference between mrBC and CrBC?

Response: Thanks. The m_{rBC} represents the mass of a single rBC core (see the statement in the page 4, line 21 in the manuscript), and the C_{rBC} is the rBC mass concentration (see the statement in the page 6, line 3 in the manuscript).

(10) Page 7, line 21: that -> those;

Response: Thanks. We have revised it.

(11) Page 7, lines 22-24: The logic of this sentence is not 100% correct. Dp increases, which could be the increase of either Dc or coating materials, or both. The authors mentioned "simultaneous increase in the rBC mass concentration" exactly in the following sentence, which makes this sentence reads really weird. Same problem applies to the texts following Figure S8, that the authors only suggested the "18-fold" increase of σab, and will need to provide more evidence on the "simultaneous increase" in both rBC and the coating materials.

Response: Thanks. Following the reviewer's suggestion, we have added the Fig. R3 (new Fig. S8 in the revised manuscript) to support "simultaneous increase in rBC mass concentration and the amount of coating materials". Correspondingly, the sentences in Page 7, lines 22-24 were revised as "*Moreover, the D_p exhibited sustained growth from ~180 nm to ~400 nm during a pollution episode, which could be a consequence of the increase in either D_c or coating materials, or both. Figure S6a shows a slight change in D_c with pollution development. However, the coating thickness of BC-containing particles increased with PM1 concentration (Fig. S10a). Therefore, the sustained growth of D_p during a pollution episode was dominated by more coating materials under more polluted conditions. Figure S10 shows the simultaneous increase in the rBC mass concentration and the amount of coating materials on the BC surface, which could significantly enhance the light absorption of BC-containing particles.*", and the statement following Fig. S8 (new Figure S11 in the revised supplement) in the revised supplement was revised as "*The simultaneous increase in the rBC mass concentration and the amount of coating materials shown in Fig. S10 revealed that the increase of $\sigma_{ab,880nm}$ (~18 fold from ~10 μg m^{-3} of PM_1 to ~230 μg m^{-3} of PM_1) could be attributed to simultaneous increase in the rBC mass concentration and the amount of coating materials on the BC surface.*"

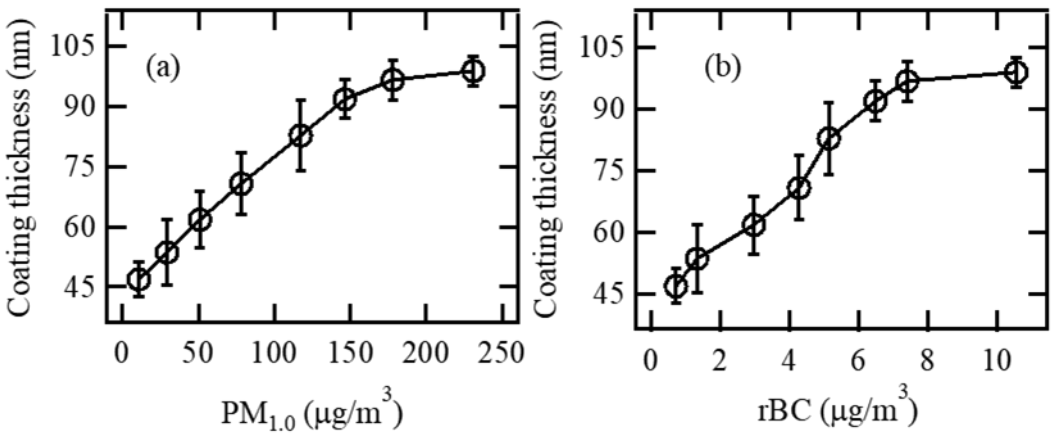

Figure R3 (Fig. S10 in the revised manuscript). Variations in the coating thickness of BC-containing particles with the (a) PM_1 and (b) rBC mass concentrations.

(12) Page 8, line 30: change rates -> changing rates;

Response: Thanks. We have revised it.

(13) Page 9, line 3: at our study -> in our study;

Response: Thanks. We have revised it.

(14) Page 9, line 21: (A) BC aging and (B) BC internally mixed with other components, it is hard to say A is the consequence of B, or vice versa;

Response: Thanks. We have revised the sentence as "*BC aging in the atmosphere, namely BC internally mixing with other aerosol components, is associated with atmospheric transport (Gustafsson and Ramanathan, 2016).*"

(15) Page 12, line 3: capacity or capability? Please note this is not the only place.

Response: Thanks. Throughout the manuscript, we have changed "capacity" into "*capability*".

(16) Page 12, lines 14-15: decreased by significantly -> decreased significantly;

Response: Thanks. We have revised it.

(17) Table 1, the unit is "μg m-3", not "μg cm-3";

Response: Thanks. We have revised it.

(18) Figure 2: Eab is not light absorption capability, it should be enhancement;

Response: Thanks. We have revised it.

(19) SI: page 4, line 13: what is "larges of coating materials"?

Response: Thanks. We have changed "larges of coating materials" into "*significantly larger in volume of coating materials than that of rBC cores*".

References:

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