Reply to the comments of anonymous reviewer #2 on manuscript Entitled " Mixing characteristics of refractory black carbon aerosols determined by a tandem CPMA-SP2 system at an urban site in Beijing"

We appreciate very much the patient and insight comments and recommendations of the reviewer in improving this paper and our future research. Here, we will response to all the comments one by one as follows:

General comments:

The paper reports the microphysical properties and aging/ mixing state of rBC particles during summer time in Beijing. The research site is mostly influenced by traffic emissions from the surrounding highways and is well representative of the Beijing urban outflow. Ambient aerosol were measured using the single particle soot photometer (SP2) for 2 weeks (30 May to 13 June 2018). Complementary measurements of O_3, NO_2 and $PM_{2.5}$ were performed, however, the measurement techniques were not specified in the methodology section, which I recommend to do so.

Reply: The concentrations of $PM_{2.5}$ and gaseous pollutants were from a state control air quality site, provided by the China National Environmental Monitoring Centre. The state control air quality site was 2.5 km from our observation site. We think the air quality condition of the state control site was similar with the air quality condition of our observation site in such close distance. And we added the position of the state control air quality site in Fig. 1b and the measuring instrument type for every pollutants (line 180-183).

There were two case studies that the authors refers as 'clean' and 'polluted' for which the rBC properties were determined. Moreover, during these periods, a dedicated experiment was performed by coupling a DMA or CPMA with the SP2 in order to determine the effective density, morphology and absorption enhancement of rBC particles due to coatings. The methods used in this study are valid, however, the measurement setup is questionable. For example, the authors do not mention whether the aerosol particles are dried before detection. The particle size depends on relative humidity (RH) that can strongly influence the results. Note that the RH is much higher in the "polluted" case.

Reply: We add a specific description about the tandem system in a new section to make it more clear, line (145-176). The air sample was dried before it entered into the tandem system. Thus, the RH wouldn't influence the particle size in this study (Fig. 2a).

Moreover, I do not agree in using the terms "clean" and "polluted" applied for the two periods. The clean period is rather influenced by the fresh traffic emissions.

Reply: The clean and polluted periods were defined according to the Chinese air quality standard which mainly focus on the mass concentration of $PM_{2.5}$. We agree there is still substantial rBC emission in clean periods. However, there were more pollutant gases and particle matters in polluted periods which may influence the rBC-containing particles' coating and morphology. For example, Zhang et al. (2018) found the D_p/D_c value of rBC-containing particles tended to be larger in the episodes with higher $PM_{2.5}$ mass concentration. That's Why we conducted the tandem experiments separately in clean and polluted episodes.

The description of the tandem experiment is not well described and difficult to understand. Since this is one of the highlights of the paper, it deserves a dedicated section on the methodology containing precise information of the measurement period, the atmospheric conditions during the experiment (what kind of air masses were sampled?) and the purposes of doing this. I suggest to have a dedicated section (after section 2.1) in the methodology for the case studies. A table containing the main results of this comparison can be also helpful.

Reply: Thanks for the advice, we added a new section to describe the tandem system, (line 145-176).

Overall, I suggest improvements of the writing. In my opinion, the discussion of the results are not presented in a precise way and the figure notes are quite vague and lacking information. For all of them, I recommend to give more details, using the full name of the variables.

Reply: Thanks for the comments, we will improve our writing in the following version. The figure notes and legends have been improved.

Specific comments:

L149: "In this study, the SP2's low detection bound was set to Dc = 70 nm". Please re-phrase.

Reply: The sentence has been changed to

The former : "In this study, the SP2’ s low detection bound was set to D_c = 70 nm." (line 149)

Now: "For rBC with Dc< 70 nm, the detect efficiency of SP2 significantly dropped below 60%. Thus, rBC with Dc< 70 nm was not considered in this study." (line 124-126)

L152: Why 1.17 factor was used?

Reply: The corrected concentration is : (the measured concentration) / (1-15%) which is the same as (the measured concentration) * 1.17. We have changed the expression:

By extrapolating a lognormal function fit to the observed mass distribution, we found that rBC particles outside the detection range caused an ~15% underestimation of the rBC mass concentration. To compensate, the mass concentration of rBC was corrected by dividing by a factor of 0.85 during the measurement. (line 126-129)

L159: replace "owing".

Reply: The sentence has been rewritten.

The former: "For rBC-containing particles, the scattering cross section significantly decreased owing to the evaporation of the nonrefractory coating as the rBC core absorbed energy" (line 159)

Now: "However, for rBC-containing particles, the particles will evaporate during the measurement since rBC can absorb the laser energy, which results in a decrease in the rBC-containing particles' sizes and thus a decrease in the $\sigma_{measured}$." (line 108-110)

L171: "The coating density was set to 1.5 g/cm3". Please re-phrase.

Reply: The sentence has been rewritten.

The former: "The coating density was set to 1.5 g/cm^3". (line 171)

Now: "Knowing M_p and M_{rBC}, the scattering cross section of rBC-containing particles can be calculated through Mie-theory with refractive indices of 2.26-1.26i of rBC and 1.48i of coatings, by assuming a core-shell structure and the coating density of 1.5 g/cm^3." (line 172-174)

L209: remove "that".

Reply: We have removed “that” in the text.

L211-212: Which MAC value did you assume for calculating the BC mass? L212: “Overestimation” of? Incomplete sentence.

Reply: The discussion about MAAP have been removed in the new manuscript since we found the coatings weren’t the cause of the bias of the rBC mass concentration measured by MAAP and SP2

L218: Rephrase “during which time”.

Reply: The sentence have been rewritten.

The former: “A heavy rainfall event occurred from 0300 – 0700LST on June 7, during which time the mass concentration of $PM_{2.5}$ decreased from 65 to 10 μg/m3 and the mass concentration of rBC decreased from 2.63 to 0.2 μg/m^3. (line 218)

Now: “ On June 7, a heavy rain fall event occurred, most of the major pollutants decreased due to significant wet scavenging. The mass concentration of $PM_{2.5}$ decreased from 65 to 10 μg/m3 and the mass concentration of rBC decreased from 2.63 to 0.2 μg/m^3 from 0300–0700 LST on June 7.” (line 190-192)

L243 – 260: This whole paragraph discussing “after rain” case should be more concise. It is a bit confusing with presenting several dates. Try to group them.

Reply: Thanks for the advice, we have rewritten the paragraph and make the paragraph more concise. (line 216-221)

L244: What was the decrease in MMD on June 4 in numbers? Is it consistent with the event on June 8?

Reply: The MMD decreased from 186 nm to 170 nm on June 4 and decreased from 183 nm to 159 nm on June 8 (Figure 5). The more decrease of MMD on June 8 may be the result of heavier rain event (Figure 3). We have added the specific MMD number in the paragraph. (line 216)

L258: Are the southerly winds representative for the Beijing outflow? And the northerly winds?

Reply: Beijing outflow is mainly affected by the southerly wind and the northerly winds. The north of Beijing is a clean region with little emission while the south of Beijing is one of the most polluted regions in China. We add a detailed discussion about the MMD characteristic when Beijing was affected by the northerly winds to make the result more comprehensive.

Since the air mass from the north is always clean, the local rBC emissions may be the main contributors to the total rBC concentration in the northerly wind period. Thus, the MMD may be more influenced by local emissions and show a weak correlation with the wind speed during northerly wind periods. (line 230-234)

L265: Investigation period.

Reply: We have changed the expression

L273: “Episode 1”, specify the time interval.

Reply: We have specified the time interval in the text.

The Dp/Dc distributions for the two episodes before the tandem CPMA/DMA-SP2 experiments are shown in Fig. 7. Episode 1 (June 7 2200 LST – June 8 1200 LST) occurred after a heavy rain period and is representative of a clean condition. Episode 2 (June 11 2300 LST – June 12 1200 LST) was characterized with the highest Dp/Dc value (1.4) and the highest $PM_{2.5}$ concentration value (120 μg/m^3). (line 246-249)

L274: "During episode 1, the Dp/Dc distribution exhibited a single peak at 1.05. However, during episode 2, two Dp/Dc distribution peaks were found". What point do you want to make here?

Reply: We want to exhibit the different D_p/D_c distribution during episode 1 and episode 2. We have changed the expression which may be more understandable and concise.

The Dp/Dc exhibited a unimodal distribution during episode 1 and a clear bimodal pattern during episode 2 as shown in the upper panel of Fig. 7. (line 249-250)

L285-286: Tends vs. tended.

Reply: Thanks, "tends" seems to be more appropriate.

L304-305: There are more recent studies on the microscopy of BC.

Reply: It's true. We update the reference now. (line 282)

L341: "The median MR values of the pollution day were all larger than those on the clean day for the four Mp points. This result demonstrated that rBC had more coating material during the pollution day than the clean day." Couldn't it be related to the higher RH?

Reply: A higher RH can truly increase the water content in the rBC-containing particles and thus the size as well as M_R. However, we have dried the rBC-containing particles before the tandem system as shown in Fig. 2(a) to avoid the influence of RH. We will write a new section to introduce the tandem system including the drying process to avoid causing misunderstanding of readers.

L380: "indicating in-cloud nucleation scavenging may be a more efficient mechanism for rBC-containing particles". Do you mean removal mechanisms?

Reply: Yes, we will add "removal" to make it more clear.

Fig. 1: has four panels. It is helpful for the readers if there is a full description of the measurements (from the top to the bottom) and possibly how they were collected. Moreover, I recommend to indicate the two studied cases in the figure. The same applies for figures 5.

Reply: A new section which is used to describe the tandem system has been added. (line 145-176) and the two studied cases have been indicated in the figure notes of Fig. 2. (line 546-547)

The Fig. 3 has been improved to denote the time when the tandem system was conducted.

Fig. 5: Y axis in log scale and increased range. What are the units of dM/dlogDc? The values seems too high!

Reply: Thanks for the advice, we have changed the figure. The unit of dM/dlogD_c has been changed to μg m^{-3}.

Fig.7: In the two upper panels, the integral of the area below the curve seems to be larger than 1, is it really the normalized dN/dlogDp? Moreover, the arrows indicating the clean and polluted periods are not

precise.

Reply: The former normalized dN/dlogDp was obtained by letting the maximum value of the histogram be 1. We have changed the calculation to let the integral of the area be 1.

The arrows have been in bold to make them more clear and extra guide line have been added to denote the clean and polluted periods.

Fig. 8: Where are the Ox measurements from?

Reply: The concentrations of $PM_{2.5}$ and gaseous pollutants were from a state control air quality site, provided by the China National Environmental Monitoring Centre. The state control air quality site was 2.5 km from our observation site. We think the air quality data is similar in such close distance. And we added the position of the state control air quality site in Fig. 1b. (line 186-189)

Fig. 9: Relationship between effective density and mobility dimeter of?

Reply: We have made the figure note more specific. Now:

Relationship between effective density and mobility diameter of rBC-containing particles. The black circle and triangle denote the fresh rBC-containing particles (Dp/Dc = 1) measured in clean day and polluted day in this study. Other markers denote the data from previous research. (line 576-579)

Fig. 11: MR = mass ratio of non-refractory matter to rBC. Add the full name to the figure description.

Reply: We have followed your advice. (line 584)

Figure S1: Does the 'after experiment' calibration have only one data point? Try to use different markers so that both measurements are visible on the plot.

Reply: Yes, there is only one data point. The purpose of this calibration is to determine the laser intensity of SP2. In fact, the DMT company suggests the calibration of the scattering signal only needs one data point. Thus, we only did calibration of scattering signal using PSL with diameter of 240 nm after the experiment which showed good consistency with the calibration before the experiment (varied within 3%) demonstrating the stability of the instrument.

Fig. S7: This figure is important to understand the origin of air masses for the two study cases. However, there is no information of the age or the starting point of the back trajectories. Also please adjust the scale of the plot.

Reply: We have drawn this figure again and specifically describe the calculation of the trajectories. (Section 2 in Supplementary, line 45-54).

Zhang, Y. X., Zhang, Q., Cheng, Y. F., Su, H., Li, H. Y., Li, M., Zhang, X., Ding, A. J., and He, K. B.: Amplification of light absorption of black carbon associated with air pollution, Atmospheric Chemistry and Physics, 18, 9879-9896, 10.5194/acp-18-9879-2018, 2018.