Reply to the comments of anonymous reviewer #3 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:

First, a full and careful proofreading is necessary to catch all the grammar and word choice errors. I have listed a bunch, but am not confident I listed them all here. In fact, after a while, I gave up listing them because this was taking too much time. Please correct these errors before sending out for review again.

Reply:Great thanks to the reviewer to point out the grammar errors. We have changed the error places and checked the manuscript several times again.

Second, there needs to be better quantification of the SP2 calibrations. To give this dataset importance in terms of the big picture, better uncertainties are necessary. The conclusions all sound reasonable as far as I can tell; but they need to be mathematically rigorous as well.

Reply: We have determined the uncertainty of our SP2 and the error bars have been added to the calibration figures. Also, the uncertainties of some key parameters have been evaluated.

A final big comment, there are many places in the paper that need more detail and clarifications. See my many comments listed below. In general, a better description of the tandem experiments and of the models used to calculate absorption enhancements are necessary. Many other places need rewording for clarity.

Reply: We have reorganized the method section and added a part to describe the tandem system including the configuration, time, the air mass during the tandem system (Section 2.32 and line 145-176 in the manuscript). The description of the calculation of absorption enhancement (Section 3 in supplementary) is added in the supplementary. Many parts of the manuscript have been rewritten to make it clearer.

Specific comments:

Line 21 - Is “enhancing rate” standard terminology? I don’t know what the units are (0.013 what per hour?).

Reply: 1) We changed the expression of “enhancing rate” to “growth rate”.

2) D_p/D_c is a dimensionless quantity, so there shouldn’t be a unit for 0.013. We change to report the growth rate of D_p instead of D_p/D_c to make it clearer for readers. The growth rate of D_p now has a unit of “nm”. (line 21)

Line 21 - Should the “x” be a subscript in “Ox”?

Reply: Thanks, we have changed the mistake. (line 21)

Line 65 - Is the "18.97%" number really accurate to two decimal places?

Reply: We directly cited this value from the work of (Qin and Xie, 2012), so we think it's not appropriate to change the value which we cited.

Line 95 - I don't understand the sentence beginning with "Anthropogenic".

Reply: We have changed the expression to make it clearer. What we mean is the emission in the campus is little, which led little influence of the observation site. Now:

Anthropogenic emissions from the experimental campus were negligible. Thus, this site can well represent the urban conditions in Beijing. (line 89-90)

Line 99 - A few more details would be nice on Fig 2a - what is the residence time in the diffusion dryer? Did you check if there were any particle losses in the dryer? When switching configurations, how long did you wait for the sampling to stabilize?

Reply: 1) Good advice, we would like to check such loose and correct the concentration according to the probable particle loose in the diffusion dryer. Unfortunately, our SP2 has some problems now and have been sent to the manufacturers for repairing and we can't do the test at present.

2)We waited a long time (about half an hour) after changing the regular single SP2 observation to tandem system measurement. And waiting for about 2 min every time we change the setpoint of DMA/CPMA to let the system stabilize, we have added these details in the method section. (line 156, line 168).

Line 104-105 - Should show error bars/scatter in the data on Fig S1. Also should quantify how constant the laser was during the study. Also, be careful of wording – two data points does not ensure that the laser was constant during the study, just that the beginning and end points were similar. Without more data, it even looks like intensity may have been drifting in one direction.

Reply: 1) There is a parameter called YAG power which is recorded in the housekeeping file in SP2 and reflects the laser intensity. We found the YAG power was 4.8±0.1 during the observation indicating the stable condition of the laser.

2) Yes, we agree that two data points does not ensure that the laser was constant. We reword the expression and use YAG power to prove the laser was nearly constant during the study. Now:

The calibration of the scattering channel and incandescence channel was also conducted after the observation, the calibration coefficient varied little (<3%) and the YAG power (laser intensity index recorded by SP2) fluctuated with 4.8±0.1 indicating the stable condition of SP2 during the observation period. (line 119-121)

Line 115 - Is it useful? (Don't use "could be".)

Reply: We have changed the expression.

Line 117 - Can you estimate uncertainty from your own calibration of the SP2? You should be able to, especially with a CPMA.

Reply: Yes, we have determined the uncertainty of our own SP2 and reported in the manuscript. (line 105)

Line 121 - Is Fig S2 really necessary? Line 123 - Is it really necessary to quote the CPMA force balance equation? What does your study do with this equation specifically? Line 127 - Is the comment about superiority of the CPMA relative to the APM necessary? What value does this statement add to your study specifically?

Reply: We have removed the Fig S2 and simplified the introduction of CPMA to make the paper more concise.

Line 136 - More precisely, the peak LII signal is what is used, not the entire LII signal.

Reply: Yes, we have changed the expression. Now:

The LII peak-M_{rBC} relationship is thus obtained (Fig. S1). (line 104)

Line 137 - Why did you use a spline fit when earlier you state that there is a linear relationship between peak height and rBC mass?

Reply: It's nearly linear relationship between LII peak height and rBC mass, but not perfectly linear. The DMT company suggested their custom to use a spline fit. The coefficient of the spline fit is exhibited in Fig S1. In fact, the coefficient of x^2 is very small.

Figure S3 - Should show error bars showing the scatter in the data and uncertainty in the particle mass from the CPMA. Also, if there is no calibration equation, how do you use these data?

Figure S5 - Again, would be nice to see error bars showing scattering/uncertainty in the data.

Reply: We have followed the advice of the reviewer. Now, the error bar and calibration equation are exhibited on the figure. (Fig S1-S3)

Line 138 - What does "approximately" mean? You should quantify these fits. And be more clear - these are spline fits like in Fig S3? My same comments apply to Fig S4 as above for S3 (include error bars, etc.).

Reply: The coefficient of DMA-SP2 calibration and CPMA-SP2 calibration varied little (<3%). For conciseness, we don't mention the DMA-SP2 calibration since the coefficient used in this study came from the CPMA-SP2 calibration.

Line 152 - Why multiply by 1.17 and not 1.15?

Reply: The corrected concentration is: (the measured concentration) / (1-15%) or (the measured concentration) * 1.17. We have changed the expression to "dividing by a factor of 0.85". We have changed the expression, now:

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 128-129)

Line 159 - This whole section should probably be edited for clarity. Specifically here, I don't understand "dividing by laster intensity". Line 161 - Again, clarity - reword "the data before a length"

Reply: We have reworded the method section to make it clearer.

Line 173 - How did you determine which was the most proper refractive index to use? Supplemental - What is "RCT"?

Reply: We have removed this part and directly use the refractive index from the previous research.

Line 182 - To be clear, the M_{rBC} is what is measured by the SP2, correct? Section 2.3.3 needs some work for clarity.

Reply: Yes, it's directly measured by SP2. The method section has been rewritten.

Line 192 - Again, there is no quantification of how well the current study compares with previous studies. It looks like your data points are systematically higher than the polynomial fit by Gysel. You should quantify the relationship and tell the reader what it means for your study.

Reply: Our results are ~7% higher than the poly-fit of Gysel but lower than the results from Moteki and Kondo. These differences may be result of different characteristics of Aquadag with varied lot and different instrument condition (such as the uncertainty of SP2).

Line 195 - What is the purpose of Section 2.3.5? Need more details. Where exactly do the parameters going into the Mie model come from? The Cabs variables should be defined in Table S1.

Reply: We have rewritten the method section and added a new section in the supplementary to describe the optical calculation. (Section 3 in supplementary)

Line 202 - What instruments measured the gaseous pollutants? Line 207 - What measured total PM2:5 mass? Was this measurement behind the cyclone? If so, was the cyclone's cut size at 2.5 microns, or something higher? These details might effect your measurement.

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 with the air quality of our observation site in such close distance. And we added the position of the state control air quality site in Fig. 1b. (line 180-183)

Line 210 - Should provide details on the MAAP in the method section. Line 213 - What do you mean

"may be affected by coating"? With the instrumentation you have, you should be able to unambiguously determine if the coatings are the reason for the discrepancy. That analysis could be an important part of this work.

Reply: Thanks. It's a good advice to examine whether the coatings are the reason for the discrepancy. We did the test and found there wasn't strong relationship between the coating thickness and the discrepancy between MAAP and SP2. Thus, we remove the part of MAAP in the manuscript.

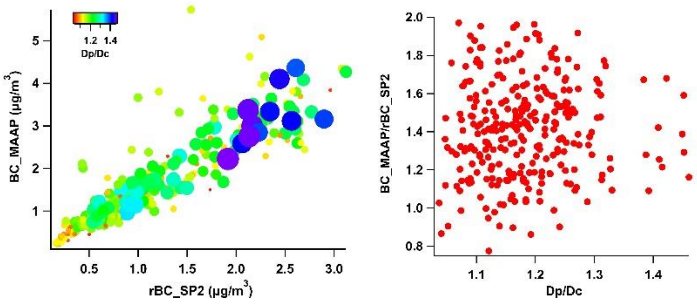

Figure 1 (a) The relationship between the mass concentration of BC (rBC) measured by MAAP and SP2, the color denoted the coating thickness. (b) The relationship between the coating thickness and the ratio of mass concentration measured by SP2 and MAAP.

Line 213 - Maybe start a new paragraph with the sentence beginning with "During"? This paragraph is a bit haphazard and should be rewritten probably.

Reply: Thanks, we changed the expression of this paragraph.

Line 216 - You don't specifically reference the other parts of Fig 3 - you should.

Reply: We try to describe more in revised version but not too much because the main purpose of this paper is to explore the properties of rBC. This Figure aimed to provide a basic observation condition for the readers.

Fig S9 - Need numbers of your scale bars. Line 223 - Why is June 13 not shown in Fig S9?

Reply: We have drawn Fig S9 again and added the backward trajectories of June 13 on Fig S9. Now: Figure S5 in supplementary.

Line 223 - To this point, I still don't understand what "the tandem CPMA-DMA-SP2 experiment" is. There are a lot more details and description needed in the Methods.

Reply: We have rewritten the method and added a new section to describe the tandem experiment. (line 145-176)

Line 276 - It actually looks like the increase was on June 12, not June 13.

Reply: Yes, it's on June 12. The episode 2 that we referred here is just June 12. We conducted the tandem experiment on June 13. So, the D_p/D_c value from single SP2 measurement is only available on June 12. And we report the D_p/D_c before the tandem experiment. The expression of this part has been modified. Now:

The D_p/D_c 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 clean conditions. Episode 2 (June 11 2300 LST – June 12 1200 LST) was characterized by the highest D_p/D_c value (1.4) and the highest $PM_{2.5}$ concentration value (120 μg/m^3) during the observation period. (line 246-249)

Line 277 - Where does 63% come from?

Reply: 63% is cited from (Zhang et al., 2018). The air quality in Beijing is easily influenced by the regional pollution transportation in pollution conditions (Wu et al., 2017;Li et al., 2017). We just use this number in the previous research to demonstrate our inference that the rBC-containing particles with Dp/Dc = 1.8 in the right peak of the bimodal distribution may be the result of transportation from pollution region.

Line 295 - Do you have any idea the magnitude/emission rate of fresh rBC in Beijing? If so, you could use that number for a closure study.

Reply: It's a good advice. It's possible to estimate the true growth rate of D_p by simultaneously considering the fresh rBC emission and the "apparent" D_p growth rate. And this true growth rate value is important in the atmospheric model. Unfortunately, the emission data about the aging degree of rBC from different emission sectors is still lacking. We may conduct laboratory experiments to determine the rBC aging degree from varied rBC sources and try to estimate the true D_p growth rate in the future.

Line 305 - This sentence is worded as if the Li et al 2003 study took images of the rBC from this study, which is obviously not correct. Were any new microscopy images taken from the current study period?

Reply: There was no new microscopy images taken from current study. The literature cited here is to support the argument that bare rBC is in a fractal structure. We have changed the expression to make it clearer and added some new literatures. Now:

This significant discrepancy indicates bare rBC was in a fractal structure consistent with the previous research from electron microscopic image that bare rBC was in a fractal chain-like structure (Li et al., 2003;Adachi and Buseck, 2013;Wang et al., 2017). (line 285-286)

Line 325 - What ambient measurements? From Peng et al 2016? These effective densities are nothing like what you report in the previous section.

Reply: Yes, it's from Peng et al 2016, we cite this literature in order to support the argument that the rBC-containing particles tend to become more compact with the coating increasing. The effective densities are the parameters in Peng's literature to support this argument. However, we think this sentence may confuse the readers and thus we changed the expression in the manuscript. Now:

Different techniques have been used to explore the morphology of rBC-containing particles in ambient and laboratory measurements (Zhang et al., 2008;Peng et al., 2016;Pagels et al., 2009). It is generally agreed that the morphology of rBC-containing particles will become more compact with the aging process or with increasing coating thickness. (line 310-312)

Line 363 - Did you find the large uncertainty? Or did Liu et al 2017? Discuss more.

Reply: Yes, both of our study and Liu found the uncertainty, typically in the external and transit stage because of the assumption in the morphology-dependent model. The section about the light absorption have been rephrased.

Line 366 - What is the "morphology dependent model"? I am very confused by the whole Section 4.2.2.

Reply: We rephrase this section and specifically describe the morphology-dependent model in the supplementary.

Line 380 - More efficient than what?

Reply: What we mean is the wet scavenging may be a more efficient removal mechanism for larger rBC-containing particles. We have changed the expression. Thanks for reminding.

Technical corrections

Great thanks for the patient and careful comments about the technical corrections from the reviewer, we have corrected the technical corrections pointed by the reviewer and carefully checked the manuscript again and again. Thanks again for the reviewer for improving this paper.

Line 44 - should be "into the atmosphere"
Line 45 - need a comma after condensation
Reply: Now: After being emitted into the atmosphere, BC particles tend to mix with other substances through coagulation, condensation, and other photochemical process, which significantly changes BC's cloud condensation nuclei activity as well as its light absorption ability (Liu et al., 2013;Bond and Bergstrom, 2006). (line 41-44)

Line 83-84 - reword: the data are not analyzed in the discussion section, they are presented
Reply: Now: A tandem experiment combining a centrifugal particle mass analyzer (CPMA, Cambustion Ltd.) and a differential mobility analyzer (DMA, model 3085A, TSI Inc., USA) with a SP2 were performed during two typical cases, focusing on BC-containing particles' microphysical properties. (line 76-78)

Line 86-87 - reword "incandescent signal emissions"
Reply: Now: After a rBC-containing particle crosses the beam, it is heated to incandesce by sequentially absorbing the laser power. (line 93-95)

Line 87 - What is "this rule"?

Reply: What we mean is we would use rBC as the abbreviation in the following section. Now:

For the SP2, the mass concentration of BC was measured on the basis of incandescent signal emissions; therefore, refractory black carbon (rBC) was used. (line 81-82)

Line 95 - Fig 1b, specifically

Reply: Thanks, we have changed. (line 89)

Line 104 - reword: the laser intensity is not constant by performing PSL calibrations

Reply: We use a YAG power index in the housekeeping file to support the stability of our SP2. Yes, it's not constant. We rephrase the expression, now:

The calibration of the scattering channel and incandescence channel was also conducted after the observation, the calibration coefficient varied little (<3%) and the YAG power (laser intensity index recorded by SP2) fluctuated with 4.8±0.1 indicating the stable condition of SP2 during the observation period. (line 119-121)

Line 148 - unified should be unity; low should be lower

Reply: Now:

For large particles, the SP2 detection efficiency was approximately unity and decreased gradually for smaller rBC particles (Fig. S3). (line 123-124)

Line 151 - without should be outside

Reply: Now:

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. (line 126-128)

Line 156 - is intracavity a noun?

Line 164-165 - reword "description ... described"

Line 170 - the densities are not defined in the text nor in Table S1

Line 174 - add "respectively" to the sentence

Line 179 - reword

Reply: These sentences have been deleted in the method section in the new manuscript.

Line 205 – reword

Reply: Former: O_3 dominant pollution occurred at 1400 LST on June 2, with a maximum of 145 ppbv, reflecting high atmospheric oxidant levels and strong photochemistry during the observation.

Now: The maximum O_3 concentration appeared at 1400 LST on June 2 with a value of 145 ppbv, reflecting high atmospheric oxidant levels and strong photochemistry during the observation. (line 185)

Line 207 - Don't use "this" as the subject of a sentence.

Reply: Former:" The mass concentration of rBC was 1.21 ± 0.73 μg/m^3 on average, accounting for 3.5 ± 2.4% of $PM_{2.5}$ on an hourly basis. This was comparable to the previous filter-based measurement in Beijing, with an average fraction of 3.2% in the summer of 2010 (Zhang et al., 2013)."

Now: The mass concentration of rBC was 1.21 ± 0.73 μg/m^3 on average, accounting for 3.5 ± 2.4% of $PM_{2.5}$ on an hourly basis, which was comparable to the previous filter-based measurement in Beijing, with an average fraction of 3.2% in the summer of 2010 (Zhang et al., 2013). (line 186-188)

Line 209-210 - reword, I don't think this is actually a sentence
Reply: It has been deleted.

Line 232 - MED or MMD?
Reply: It's MED. Liu et al. (2014) only reported the average MED and standard deviation of MED in winter and summer but not MMD, but these MED value also show a summer-low-winter-high trend.

Line 242 - combination should be combined
Reply: Now:
The diurnal cycle reached a peak plateau between 0300–0700 LST and it decreased gradually in the afternoon, which was controlled by the combined effects of the development of a planetary boundary layer (PBL) variation and on-road rBC emissions. (line 213-215)

Line 243 – reword
Reply: Former: "Significant change of rBC's mass size distribution occurred on June 7, corresponding to the heavy rain period. After the heavy rain event, the MMD decreased sharply to 159 nm. This decreasing trend of MMD also occurred on June 4 during another rainfall event."
Now: After the two rain events (June 4 and June 7) as shown in Fig. 3, the MMD decreased significantly from 186 nm to 170 nm and from 183 nm to 159 nm separately. (line 216-217)

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