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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Contrasting physical properties of black carbon in urban Beijing between winter and summer" *by* Dantong Liu et al.

**Anonymous Referee #2**

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Overall, I find this to be a largely observational paper that provides some interpretation. I find it to be written in such a way that it is often difficult to follow. Most often this is because of a too-rapid going back and forth between winter/summer. I think that the authors should strongly consider trying to organize each section to fully describe each season, and then make comparisons. This would also really help in instances when they are trying to make specific points about specific seasons. I do also have some concerns about what the Esca-Dc plots mean when the Esca is < 1, and what this says about the uncertainty of the method overall. I agree with the first reviewer that the "what" is generally (although not always) clear, but the "why" is often lacking. I think the authors could do a better job at supporting their conclusions. There is a lot of very specific terminology used throughout and a summary table would be very



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helpful/welcome. I think that the measurements are, overall, of good quality and the analysis is likely robust, and thus the manuscript could ultimately be publishable. However, I do also think that the manuscript could do with some structural reorganization within sections, with some greater details, and with stronger connections between observations and interpretation. My specific comments follow below, with ** put next to those that I think are more crucial.

L34: The meaning of “dilution effect” is not clear.

L36: The meaning of this sentence about source apportionment methods is not clear. What does “physical method” mean? And how are these combined if they are, apparently, performed separately? (Perhaps the text addresses this, but the abstract is unclear.)

L42: What does “tended to dominate with moderate coatings” mean? These particles do not dominate the BC mass overall. Are words missing?

L100: The use of the word “novel” does not seem appropriate here. The techniques have been used previously. Perhaps a “novel combination” is appropriate, but even then I’m not certain as there are other studies that have looked at BC size distributions and composition.

References Liu et al. (2014a) and (2014b) are the same reference.

Fig. 1: The units on the emissions are not clear. It says Mg/m. Why per meter?

L157: It is unclear how the authors established that this “is the optimum metric to reflect. . .” In what way specifically was it optimum? How was this established specifically? How is varying by +/-10% the right value?

L181: What is the smallest coating amount that can be reliably determined, given the uncertainty of the method?

L195: I suggest the authors use a sub, rather than superscript for the MAC, such that

there is zero ambiguity as to whether this is an exponent.

L205: How was the PAX calibrated? How do the MAC values measured at 870 nm compare with those calculated at 550 nm?

L223: What does it mean to apply PMF in “real time”? Per the cited Wang paper, the PMF analysis was conducted using standard methods, which are certainly not applied in “real time”.

L233: I am not certain that the statement “As the bottom panels show, the site was mostly influenced by northerly air masses in winter. . .” is justified by the data. The figure shows that probably half of the total period was dominated by Plateau South air masses.

Fig. 3: The authors should consider using a different color scheme, especially one that does not put green and red next to each other.

L244: It would be helpful if the authors would elaborate on the meaning of the following sentence: “In summer, air masses from the western NCP showed lower RH which may result from the almost latitudinally homogenous distribution of higher temperatures.” How does a “homogenous distribution” translate to lower RH? I understand why higher temperatures at the point of measurement might. But higher temperatures elsewhere could, at least in theory, lead to increased evaporation of water.

L250: The authors state that rBC concentrations are higher in winter due to higher emissions. But boundary layers are often also lower in winter, leading to higher concentrations of primary pollutants. How can the authors separate these effects, or at least rule out boundary layer differences as an important reason for the wintertime increase? This should especially be rationalized with the authors statement above that wintertime saw more air masses from the northern plateau yet that the northern plateau airmasses were linked to periods of the lowest concentrations. And since the authors note boundary layer height differences below (L269). They do this below, but

they might either bring this discussion up or point the reader to the later discussion.

L256: When the authors state “This may be also. . .” the also makes me think that there has been some argument advanced already. But they do not seem to advance an argument before this point as to why the particles sizes from the northern plateau were smaller. Why would lower concentrations mean smaller sizes?

Fig. 5: If the authors were to show the averaged BC size distributions for the different air masses, perhaps as a supplemental figure, this would help the reader to understand the MMD histograms.

L264: It is not clear why “longer westerly transport” would result in larger rBC cores. This aspect needs to be justified.

** Section 4.1: I find that the authors bounce between summer/winter very quickly, and not always clearly. I suggest that this might be clearer if the authors were to fully present one season, and then the other, and then point out notable similarities/differences. As written, I find this more difficult to follow than it need be. This continues through many of the sections.

L270: Just to be clear, when the authors refer to BC being “concentrated” by the shrinking of the PBL at night, they are not implying that the shrinking itself concentrates the BC, correct? This is not physically what happens. Emissions that do occur at night are into a smaller atmospheric region and thus end up more concentrated.

**BC Sources: Is BC from coal expected to be chemically similar as BC from other sources? I could see reasons it might be quite different chemically, and therefore quite different optically. If coal combustion, especially residential coal combustion, is an important source of wintertime BC, could it be possible that the coating amount estimation method might have some trouble, as it uses an RI that was determined for BC from (most likely) vehicle combustion? Is there any evidence available in the literature to illustrate that BC from coal behaves similarly and that the methods applied





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are appropriate? Has any direct source testing been done? I am unaware of any SP2 source sampling of coal. Yet, we know that the properties of BC can influence the SP2 measurements (e.g. Laborde et al.)

L274: I am not sure about the statement that there were no “obvious” diurnal variations in the BC coatings. The figure suggests a reasonably evident increase around 10 am in winter and a small increase around 9 am in summer. And in winter the size seems to be notably higher at night than day. The variation is not huge, but it seems evident. The authors use this statement regarding no “obvious” variations to argue that things are “well mixed during both seasons.” This seems to be a bit of a stretch.

**L280: Is the conclusion that the size distributions are log normal robust over time? The two winter distributions shown suggest that if a campaign average were calculated one might need to use a multimode fit. Is this conclusion only true for a relatively narrow BC mass concentration range? That the MMD varies with BC concentration (Fig. 7b) suggests that the overall average distribution is not fittable by a single lognormal mode. Further clarification is needed. Also, are the fits given in Fig. 7b/c meaningful? These seem like arbitrarily chosen functions. Given the scatter in the data, one might think another functional form would work nearly as well. Are these just to show that the data vary? The authors might consider binning the data instead to illustrate this point.

**L295: The authors suggest coagulation might be responsible for the increase in BC size when the BC concentration is large. How can they exclude a shift in source? If they are going to speculate about one reason, they should speculate about the other reasonable interpretation (change in source). Also, arguments regarding coagulation would be strengthened if the authors could point to the fraction of the total particles that contain BC. Only BC-BC coagulation leads to growth. BC coagulation with non-BC particles does not lead to growth of the BC core. In many environments, BC is only a small fraction of the total particles. What is the situation here? I suggest this should be discussed. Especially, I don’t understand how the authors can argue that a shift in sigma during one season is likely due to changes in source but in another it is





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coagulation just because the direction of the shift is different. This requires the authors knowing a priori what the end members of mixing line are with respect to width and MMD. The weakest part here, in my opinion, is in the range of concentrations where the two seasons overlap (0.1-4 ug/m3) and where different behavior is observed. Why would coagulation drive behavior during one season in the overlapping region but not the other? Especially when the authors argue for a "greater complexity of sources" in winter. I suggest this paragraph needs substantial revision.

**L310: The discussion regarding the increase in coatings in the winter at higher BC concentrations would be, in my opinion, greatly strengthened if the authors also considered how the absolute concentrations of other PM species varied. Also, I find this discussion to be very weak in the context of the available data. The related paper (Wang et al.) uses PMF to analyze the coatings on BC. There are primary and secondary coating materials identified. How do these play into things? The discussion, as presented, is just statements of obvious factors that might impact coating amounts. But the authors could, and should, go beyond this, given the available data.

**Fig. 9: How should one interpret the large number of points below the Esca = 1 line? Presumably, no point should be below this line if the interpretation is robust. This is especially important for the "large uncoated BC" region, which is almost entirely in a range where signal should not exist. Also, what is the smallest core size for which a coating can be reliably determined? (Is there any mismatch between the incandescence and scattering lower size detection limits?) I do understand that there is uncertainty in the measurements, and that perhaps this is what contributes. But the particular patterns of the Esca-Dc relationship, which trend towards values of Esca < 1 as Dc increases in general, really make me question the robustness of the method and interpretation.

**Fig. S2: I do not understand this figure. The underlying distributions appear to have arbitrary sizes. I think these are somehow derived from the Esca-Dc relationship. But they are very oddly shaped, essentially unphysical. This figure is mentioned briefly as supporting conclusions regarding sources. But given that the shapes of the underlying

distributions are so very, very strange, I believe that it requires substantial additional discussion. These are most clearly not log normal, as stated by the authors.

L342: This should clarify that these are linear fits.

Section 4.4: It would be helpful to clarify that this is only for winter right at the start of this section.

Section 4.4: The authors need to clearly define what they consider good, moderate, poor, etc. correlations. A value of R2 > 0.6 is stated as both “high” and “moderate” and “tightly” for example. Use of consistent language would facilitate consistent interpretation.

**L355: I am finding it difficult to understand all the terminology. The authors are variously defining things by ranges (I-IV), types (fossil fuel, biomass burning, traffic), etc. An effort to really clarify all the terminology would be most welcome and would facilitate the readers understanding. This is especially true when the authors make statements such as that the fossil fuel and biomass burning have similar core sizes and coating contents. I am having a very difficult time understanding what, specifically, the authors refer to (especially when I look again at Fig. S2, where the distributions of the different types seem to vary quite greatly).

OOA: Per the complementary Wang et al. paper, the OOA2/BC ratio is notably larger than the OOA1/BC ratio. Yet the OOA2_BC factor is more correlated with “moderately” coated BC while the OOA1_BC is more correlated with “thickly” coated BC. Can these be reconciled?

L374: This sentence could be rewritten to make it clearer. Use of commas, at least, would help.

L382: The wintertime BC was, generally, anticorrelated with the MLH. It did not “follow” the MLH.

L401: The citation to Xu et al. (2000) just points out that NOx can be controlled from

coal combustion. It does not address the extent to which modern NOx controls are implemented today in and around Beijing. A more modern reference of direct relevance would be welcome.

L404: Since no large, uncoated BC mode was observed in London, and since this makes up a large fraction of the total, is it fair/relevant to compare the absolute fractions from this study to a study from London? I am not sure that it is. The authors might consider renormalizing, removing the large, uncoated BC mode from the statistics, if they wish to compare in this semi-quantitative way.

Fig. 9: Is each panel individually normalized to the maximum?

**Section 4.6: I am surprised to not see a discussion of how, perhaps, the small, uncoated BC is converted to thickly coated BC when the PM levels increase. It is evident from Fig. 12 that the sharp drop in the fraction of small, uncoated BC results largely from an increase in the thickly coated BC. What I find in this section is largely just a statement (or really, a series of statements) as to how the fractions of one type change with another. But there do not seem to be a lot of insights that actually come out of this section, in my opinion. I suggest that the authors focus more on development of insights rather than just a statement of relationships. Where they do try to develop insights, they really come off as speculative (e.g. L447) rather than fully developed.

Fig. 12/L455: if the calculations of absorption really account for single particle coating state, then I have a difficult time understanding how there is a linear translation between the particle fractional contributions (left axis) and the absorption contribution (right axis). This implies that there is a single, characteristic value for each BC type that the fractional contribution can be multiplied by. But this wouldn't seem to go with the single-particle analysis. Is the single particle analysis not applied at each point in time to understand the variability within the different classes, but instead applied as a class average? I think it is the latter based on the discussion, but this could be clearer.

L461: How was the MAC at 550 nm determined? The measurements were made at





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870 nm. I find this unclear. Also, people typically think of BC as having an MAC at 550 nm around 7.5 m2/g (see e.g. Bond et al. (2006)). Yet, the translation between the MAC and Eabs in Fig. 13 implies a smaller MAC was used with the measurements, and this is confirmed on L458, although there is a seemingly contradictory value on L465 (which is perhaps just demonstrating the inadequacy of Mie theory for calculation of MAC values.) Unless what the authors are doing is actually showing the measured Eabs and a modeled MAC. It is not clear what the authors have done here.

**Section 4.6 – Absorption: The authors report their MAC values, but provide very little interpretation. Some interpretation would be welcome. They cite the Zhang (2018b) paper as some support of the reasonableness of their observations. But, Zhang et al. (2018b) find Eabs values at the same wavelength that never fall 1.7 while here the authors find at about the same conditions values of 1.2. (Personally, I think there are substantial problems with the Zhang et al. (2018b) paper, but nonetheless there is an inconsistency that challenges the simple citing of this as support.) The total PM/BC ratio appears to increase with the PM1 concentration. Could there be additional brown carbon leading to the increase? Or do the authors think that the increase results from the coatings? Is this what they are trying to imply (but not stating directly) when they compare the observations to the calculations (although as I note above the origin of the observations at 550 nm is not clear)? I think the authors should be more explicit. Note also that there is only a “shadowing” effect (L470) if the coating is absorbing.

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