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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterising Mass-resolved Mixing State of Black Carbon in Beijing Using a Morphology-Independent Measurement Method" *by* Chenjie Yu et al.

Anonymous Referee #1

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This paper presents results from the APHH campaign in Beijing where mixing state of black carbon-containing particles was experimentally determined using a combination of a centrifugal mass analyzer and a single particle soot photometer. To quantify "black carbon mixing state", 2D number distributions were produced, the mass ratio of non-BC and BC material was used, as well as the mixing state parameter chi.

This paper represents an important step forward in deriving mass-fraction-based metrics from single-particle measurements. I commend the authors for accomplishing this, as it is a key prerequisite for eventually being able to compare these measurements to mixing-state-aware models. The data analysis reveals interesting and novel insight

into the seasonable variability of black carbon mixing state in a polluted environment such as Beijing. Overall this work warrants publication in ACP, but before it can be accepted, several issues need to be addressed. I have four major comments regarding the presentation and interpretation of the results, and a number of minor comments, mainly pertaining to the clarity of notation and terminology.

Major comments:

1. The usage of the mixing state chi is confusing in the paper. Due to the measurement technique, it is only the mixing state of BC-containing particles that can be evaluated, i.e. the method is blind to a potentially existing subpopulation that only contains non-BC material. This is an important point, and it is explained in the paper, but it comes too late. I recommend introducing the specific use of chi already in the introduction, following the overview of previous studies that used chi as a metric for mixing state (note that each of those studies have their own take on chi). The authors might consider calling their chi "χ_{BC}" or similar, to emphasize this point.

2. Related to comment 1, the interpretation of chi is also inconsistent throughout this paper. In the abstract and a few times in the text, it is correctly mentioned that chi (as defined here) "quantifies whether the coating is evenly distributed across the rBC-containing particle population". – I agree with this interpretation, and suggest that the authors explain this in the text more precisely, and stick to this interpretation throughout the text. I.e. a high value of chi means that all particles have the a very similar BC mass fractions (the same mass fraction for chi=100%) compared to a low value, where the distribution of BC mass fractions is very uneven across the population.

I strongly suggest not to use language like "A decrease of chi was observed [. . .] which indicates a period of more externally mixed rBC" (p. 11, line 8). The phrase "Externally mixed" is not meaningful here. In fact, I believe you can describe your results without even mentioning the terms internally and externally mixed.

3. Having established the meaning of chi in your study, it is important to recognize that

the diversity metrics as used here are mass-weighted. Therefore, the overall chi for the whole population will represent mainly the conditions for the large particles. It would be interesting to see how chi varies between small and large particles. Is it possible to calculate a size-resolved chi?

4. Use of the terminology “core-coating”. The authors emphasize that their measurement method is morphology-independent, but still frequently use the terms “BC core” and “coating”. I think these terms should be avoided because it invokes the traditional BC core-shell assumption, which may or may not apply in reality (and which is actually not made by the authors!).

Minor comments:

1. The abstract is too long. I recommend shortening it.

2. p. 1, line 17: Which year was the campaign in?

3. p. 1, line 25: “positively associated” - Do you mean “correlated”?

4. p. 2, line 9: Reference to Ramanathan and Carmichael (2008) is more than 10 years. Suggest to replace with a more recent reference.

5. p. 3, line 20: “measure the different mass ratio” – Should read “measure the mass ratio”?

6. p. 4, line 25: The size ranges for the detection limits are confusion. Do you mean 200-720 nm for the mixed particles (i.e. referring to the scattering signal) and 70-850 nm for the BC-component (traditionally called the BC core)? Please clarify. In this context, Figure 5 shows that the distributions extend to smaller diameters than 200 nm/70 nm. Do you also extrapolate towards the small sizes (not only the large sizes)?

7. p. 5, section 2.5: This is beyond the scope of this paper, but possibly an avenue for future research: Since you know the size distribution of the non-refractory particle matter from the AMS, and you know how much NR material is associated with BC,

can you infer the size distribution of the subpopulation that does not contain BC by combining the AMS with the CPMA-SP2 measurements? This would yield a complete picture of aerosol mixing state.

8. p. 6, line 22: This is an example where Major comment 2 applies. The terms internal/external mixing are not useful here. Chi as applied to the subpopulation of BC containing particles quantifies how evenly the BC and non-BC mass fractions are distributed. (with a focus on the large particles).

9. p. 8, line 16: “For better quantification” – what does that mean? To reduce noise?

10. p. 9, line 1: “It would appear that .. “ – can you please clarify? Do you mean that the summer distribution is similar to winter/moderate or winter/light pollution, or that it is not similar?

11. p. 9: equation 6 and 7: Suggest finding more succinct variable names for “single particle MR” and “bulk MR”. Even MR_i and MR_{bulk} would be an improvement here.

12. p. 9, equation 7: Since $m_{p,i}$ refers to an individual particle, and the summation is over all particles, there should not be the factor N_i. In other words, isn't $N_i = 1$?

13. p. 10, line 8: “due to the presence of large numbers of large rBC-particles ... the bulk MR value increased rapidly.” This seems the wrong way round, since $m_{BC,i}$ is in the denominator for Bulk MR.

14. p. 10, line 27: should be Figure 9. Is there anything that could be gained by comparing the bulk MR with the population average of the single particle MR?

15. p. 10: Figure 9: What does MrBC mean? Caption: Should read “evolution”.

16. p. 10, line 32: Can you please add a reference, why coal burning should lead to thickly coated rBC-containing particles?

17. p. 11: Comparing to Healy et al. (2014), it would be important to note that they defined chi based on the 5 species system (SO4, NO3, NH4, OA, BC). So those chi

values and the one from this study are not directly comparable.

18. p. 11: Figure 10, top panel: Specify that it's bulk diversity for the y label.

19. p. 12, line 6: "there was a strong correlation. . ." Can you quantify this more precisely?

20. p. 12 Discussions: This section is also a good example that should be rephrased in light of Major comment 2. I think the main point of figure 12 is that during summer, for a narrow range of bulk MR a wide range of chi is observed, meaning even for similar bulk MR, there is a variation how the "coating material" is distributed (note that in summer chi is not always high). During winter, there seems to be a positive correlation between bulk MR and chi, meaning that the NR material is more evenly distributed when bulk MR is high. The explanation of why this summer/winter difference happens, however, is not clear. It would be important to improve this discussion.

21. p. 13: Implications: I appreciate the authors effort to connect their results to the modeling community, but The statement "it may be appropriate to assume internal mixing of BC in the summer" is over-reaching in my opinion. It would at least require an estimate of calculating absorption enhancement for the conditions shown here to substantiate. Water uptake (which will be modulated by the composition of the NR material) further impacts this impact.

I think an important contribution of this study is that it provides a framework to produce the 2D distributions as shown in Figure 4 (mass-based, including an objective method of extrapolation). This is data that mixing-state aware models (including, but not limited to particle-resolved models) can compare to.

22. Axis labels are generally too small

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