Reviewer #2,

Comment: This paper presents an analysis of ambient SP2 measurements at a site in Taizhou, China, to explain the range of observed black carbon absorption enhancements given a certain value of the mass ratio or non-BC coating material and BC. Motivated by the fact that previous studies show that the mass ratio and the absorption enhancement are only weakly related, the authors show that the range of absorption enhancement values at a given mass ratio can be explained by the mixing state of BC-containing particles (quantified by the mixing state metric χ). The paper presents an interesting analysis and fits within the scope of ACP. I have several comments that should be addressed before the paper is suitable for publication. I should note that the paper contains quite a few typos. I only flagged the typos that in my view hampered the understanding of the material, and I strongly recommend thoroughly proofread the revised version.

Reply: Thanks for the comments and suggestions. The point-by-point responses are listed below.

Comment: General comment: 1. To make the paper more impactful, I recommend that the authors could make more clear how their findings of the relationship of Eabs, MR, and χ can be applied in practice.

Reply: Thanks for the comment. The new finding of our study is that the mixing state index can contribute to improvements in the accuracy of simulating the BC radiative effects. In the particle-resolved simulation of ambient aerosols, the particle-to-particle heterogeneity of BC-containing aerosols can be resolved by simply introducing the BC mixing state index χ. Then the aerosol light absorption enhancement can be better constrained by MR and χ and then the radiative effects of BC can be estimated. Therefore, our framework can be employed in the model by simply introduce a BC mixing state index for better estimating the BC radiative effects.

We added these descriptions into the conclusion part of the manuscript.

Comment: Detailed comments: 1. Title: The title could be more descriptive of what the paper is actually about (relationship of absorption enhancement, mass ratio, and BC mixing state)

Reply: Thanks for the comment. We modified the title into "Method to Quantify the Black Carbon Aerosol Light Absorption Enhancement with Mixing State Index"

Comment:2. Abstract: Make clear that MR is used here as a bulk quantity of the population, rather than a per-particle quantity, i.e., MR here is the mass ratio of non-BC coating material in the population to BC in the population.

Reply: Thanks for the comment. We agree with the reviewer's opinion. We added some descriptions in the abstract to make clear the definition of MR. MR is the mass ratio of non-BC coating to BC in the population of BC-containing aerosols.

Comment: 3. Line 14: "coating thickness" should read "coating material"

Reply: Thanks for the comment. We revised the "coating thickness" into "coating material".

Comment: 4. Line 31: should read "lensing effect" (not "effects")

Reply: Thanks for the comment. We revised the "lensing effects" into "lensing effect".

Comment: 5. Line 82: Specify what is meant by "size-selected mixing states". I assume it means the distribution of BC core and non-BC coating thickness for a given total particle diameter?

Reply: Thanks for the comment. We agree with the reviewer's opinion and revised the "size-selected mixing states" into "size-selected distribution of BC core and non-BC coating thickness" in the corresponding text.

Comment: 6. Section 2.2: Add information on what size ranges the instruments can sample (and for which Eabs, MR, and χ is determined).

Reply: Thanks for the comment. As noted by Zhao et al. (2020), the SP2 can only detect these BC-containing aerosols with a core diameter larger than 84 nm. The DMA select the aerosol at the range between 13.3 nm and 749.9 nm. In the following discussion, the size-resolved distribution of BC core and coating thickness are constrained in the range between 84 and 749.9 nm.

The above information was added in section 2.2 in the manuscript.

Comment: 7. Line 92: Notation: This should be $\frac{d^2N}{\mathrm{dlogDp}\cdot\mathrm{dlogDc}}$ (second derivative). There are many other places in the paper where this needs to be corrected.

Reply: Thanks for the comment. We revised the corresponding texts in the manuscript.

Comment: 8. Line 113: "without thickness" should read "without coating"

Reply: Thanks for the comment. We replaced the "without thickness" with "without coating".

Comment:9. Line 116: Notation: Dp is the total diameter, so Dp = 0 doesn't make sense.

Reply: Thanks for the comment. It should be Dp=Dc here, and we replace the $\sigma_{abs,Dp=0}$ with $\sigma_{abs}(Dp = Dc)$.

Comment: 10. Line 112: Notation: Given that Dp and Dc are used as independent variables, I suggest writing $\sigma_{abs}(Dp, Dc)$ rather than putting Dp and Dc as index.

Reply: Thanks for the comment. We revised the text based on the reviewer's comment.

Comment: 11. Line 118: The use of the word "dispersion" sounds awkward. Suggest using "variability of BC mixing states" or simply "Quantifying BC mixing states".

Reply: Thanks for the comment. We revised the text.

Comment: 12. Line 139: H_α is the average mixing entropy of the population (not of each particle).

Reply: Thanks for the comment. We revised the text.

Comment: 13. Section 2.4: Note that a number of different (binary) species definitions for χ have been used in the literature, e.g. Ching et al. (2017) based their calculation on hygroscopic and non-hygroscopic species, Dickau et al. (2016) used volatile and nonvolatile species, Zheng et al. (2021) compared three different variants for χ, one of which was based on absorbing (BC) and non-absorbing species,and Yu et al. (2020) use a metric which is very related to this paper. It would be good to cite these studies here to provide context for this paper.

Ching, J., Fast, J., West, M. and Riemer, N., 2017. Metrics to quantify the importance of mixing state for CCN activity, ACP, 17, 7445-7458

Dickau, M., Olfert, J., Stettler, M.E., Boies, A., Momenimovahed, A., Thomson, K., Smallwood, G. and Johnson, M., 2016. Methodology for quantifying the volatile mixing state of an aerosol. *Aerosol Science and Technology*, *50*(8), pp.759-772.

Yu, C., Liu, D., Broda, K., Joshi, R., Olfert, J., Sun, Y., Fu, P., Coe, H., and Allan, J. D., 2020. Characterising mass-resolved mixing state of black carbon in Beijing using a morphology-independent measurement method, Atmos. Chem. Phys., 20, 3645–3661,

Zheng, Z., Curtis, J.H., Yao, Y., Gasparik, J.T., Anantharaj, V.G., Zhao, L., West, M. and Riemer, N., 2021. Estimating submicron aerosol mixing state at the global scale with machine learning and Earth system modeling. *Earth and Space Science*, *8*(2), p.e2020EA001500.

Reply: Thanks for the comment. We added these studies in second 2.4 in the manuscript.

Comment: 14. Line 157: “group bulks” sounds awkward. Suggest “populations”.

Reply: Thanks for the comment. We revised the text correspondingly.

Comment:15. Figure 2: This figure is confusing since in line 151, χ in this paper was defined only based on BC-containing particles (meaning that BC-free particles are not included in the calculation), while Figure 2 shows BC-free particles as examples. Please clarify and modify figure 2 as necessary.

Reply: Thanks for the comment. We modified figure 2. In the new figure, the amounts of BC are very small (mass ratio of the core and shell are 10^{-9}) and most of the aerosols are composed of the non-BC component. We added the BC core in Fig. 2 and some revisions are made in Table 1.

Comment: 16. Also, to make this figure easier to understand I suggest numbering the example populations according to the discussion in the text.

Reply: Thanks for the comment. We numbered the example populations in Figure 2.

Comment: 17. Figure 3: H_α and H_γ are redundant with D_α and D_γ but more difficult to interpret than the diversity metrics D_α and D_γ. Suggest removing the subpanels for H_α and H_γ from this figure.

Reply Thanks for the comment. We removed the subpanels for H_α and H_γ from the Figure 3.

Comment: 18. Figure 3: The temporal variability of the quantities shown here is interesting and deserves more in-depth discussion. For example, in line 187, it says that "D_α decreases with the MR". However, the figure shows D_α decreasing while MR is increasing. Please clarify and explain more clearly what process is responsible for these changes. Also, Figure 3a shows relatively low σ_{sca} values during the daytime while MR remains at a relatively constant level. Can you explain why this is?

Reply: Thanks for the comment. We tend to show that the D_γ decreases with the increasing of MR and made some revisions in the manuscript correspondingly.

The D_γ value is mainly determined by the total mass concentration ratio of the BC component to the non-BC component. It varies between 1 and 2 for different total mass concentration ratios. The D_γ increases when the mass ratio approaches 1. During the aging processing when the MR is larger than 1, it is reasonable that the D_γ is decreasing while the

MR is increasing from Fig 2. However, it is not clear why the D_α decrease with the MR.

Also, Figure 3a shows relatively low σ_{sca} during the daytime while MR remains at a relatively constant level. The bulk MR values are mainly determined by aging process of Bc and new emission of BC. The aging process of BC would lead to the increment of MR, while the new emission of BC would lead to the decrement of MR. In the day time, the decrement of MR due to new emission may be comparable to that of the increment of MR due to aging process. Thus, the MR can remain at a relative constant level.

Comment: 19. Line 208: "refractive index of χ --- What does this mean?

Reply: Thanks for the comment. We revised the text into "BC mixing states index χ".

Comment: 20. Figure 5: Suggest mentioning that the population shown for $\chi = 0.81$ is only one possible example. There are many other possible ways the particle composition can be arranged that would give the same mixing state index.

Reply: Thanks for the comment. We added some descriptions in section 3.3 correspondingly.

Zhao, G., Shen, C., and Zhao, C.: Technical note: Mismeasurement of the core-shell structure of black carbon-containing ambient aerosols by SP2 measurements, Atmospheric Environment, 243, 117885, 10.1016/j.atmosenv.2020.117885, 2020.