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Comment on acp-2021-336

Anonymous Referee #2

Referee comment on "Method to quantify black carbon aerosol light absorption enhancement with a mixing state index" by Gang Zhao et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-336-RC2, 2021

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 publications. 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 to thoroughly proofread the revised version.

General comment:

1. To make the paper more impactful, I recommend that the authors could make more clear how their finding about the relationship of $E_{\rm abs}$, MR and χ can be applied in practice.

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)

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.

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

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

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?

6. Section 2.2: Add information on what size ranges the instruments are able to sample (and for which $E_{\rm abs}$, MR and χ is determined).

7. Line 92: Notation: This should be $\frac{d^2N}{d \log D_{\rm p} d \log D_{\rm c}}$ (second derivative). There are many other places in the paper where this needs to be corrected.

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

9. Line 116: Notation: $D_{\rm p}$ is the total diameter, so $D_{\rm p} = 0$ doesn't make sense.

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

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

12. Line 139: H_{α} is the average mixing entropy of the population (not of each

particle).

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 non-volatile 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.

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

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.

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

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.

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_{\alpha} decreases with the MR". However, the figure shows D_{\alpha} decreasing while MR is increasing. Please clarify and explain more clearly what process is responsible for these changes.

Also, Figure 3a shows relatively low $\sigma_{\sca}$ values during the daytime while MR remains at a relatively constant level. Can you explain why this is?

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

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.