Reviewer #1,

Thanks for your comments! The point-by-point responses are listed below.

*Comment:* In this paper, the authors proposed a method to quantify light absorption enhancement for black carbon (BC) aerosols by considering entropy and diversity. The authors indicated that the mass ratio (MR) of non-BC coating thickness to BC (MR) and particle-to-particle heterogeneity represent two key parameters in regulating the radiative absorption enhancements by BC. They introduced a BC mixing state index ( $\chi$ ) to quantify the dispersion of BC mixing states based on a binary system of BC and non-BC components.

They showed that the BC light absorption enhancement increases with  $\chi$  for the same MR, indicating that  $\chi$  can be employed as a factor to constrain the light absorption enhancement of ambient BC. This work proposed a novel framework to treat BC light absorption enhancement, which can be useful to study BC radiative effects in climate models. The paper was reasonably written, but some effort is still necessary to improve its readability. I recommend the publication of this paper in ACP, provided that the following issues have been adequately addressed.

*Reply:* Thanks for the helpful comments.

## Comment: Major points

(1) The title needs to be modified since it is unclear how their framework is obviously linked to "entropy and diversity measures"

*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) The authors argued that the BC light absorption enhancement is dominantly determined by two physical parameters, i.e., MR and  $\chi$ . However, there are several studies showing that the chemical properties of the coating materials, i.e., organic versus inorganic species, are also critical in regulating the morphology and optical properties. For example, coating of sulfuric acid has been shown to be more efficient in altering the BC morphology and light absorption (e.g., Variability in morphology, hygroscopic and optical properties of soot aerosols during internal mixing in the atmosphere, Proc. Natl. Acad. Sci. USA 105, 10291, 2008). Such an aspect needs to be discussed in the context of their proposed framework.

*Reply:* Thanks for the helpful suggestions. The reviewer gave a good aspect that should be concerned when dealing with the light absorption of the ambient BC-containing aerosols. We added some corresponding descriptions into the introduction part.

*Comment:* (3) It would be desirable that their proposed framework can also compared to other experimental studies, particularly those relevant to different chemical species (Enhanced light absorption and scattering by carbon soot aerosols internally mixed with sulfuric acid, J. Phys. Chem. 113, 1066, 2009; Effects of dicarboxylic acid coating on the optical properties of soot, Phys. Chem. Chem. Phys. 11, 7865, 2009).

**Reply:** Thanks for the helpful comment. The reviewer gave a perspective aspect that we should concern in our future works. Our study aims to propose a method to quantify the black carbon aerosol light absorption enhancement with mixing state index, which requires the information of particle resolved BC mass ratios. Unfortunately, these data were not measured in the refered experitemental studies.

*Comment:* (4) Also, I believe that their proposed framework deals exclusively with dry particles. Under atmospheric conditions, aerosols (particularly for those containing high level of inorganic species) likely experience hygroscopic growth at high relative humidity (RH), which inevitably impacts their morphology and optical properties. How such an issue could be addressed by their method.

**Reply:** Thanks for the comment. We calculated the BC absorption coefficient under different RH conditions with Mie theory and found that the BC light absorption enhancement is slightly related to the aerosol hygroscopic growth. The core diameter and coating thicknesses were 100 nm and 50 nm with a refractive index of the core and coating of 1.67 + 0.67i and 1.46 + 0i respectively. As for the  $\kappa$  for of shell, it is estimated using the parameterization scheme proposed by Yu et al. (2018) and mean results of the real-time measurement of the aerosol chemical compositions by an In situ Gas and Aerosol Compositions Monitor (TH-GAC3000, China). A mean value of 0.16 is derived and used here. The calculated absorption coefficient ( $\sigma_{abs}$ ) under different relative humidity (RH) are shown in Fig. R1. Results show that the  $\sigma_{abs}$  ranges between 40.5 and 36.5 when the RH ranges between 40% and 90%. Thus the  $\sigma_{abs}$  varies slightly within 5% under different RH conditions. Therefore, the light absorption enhancement of BC particles at dry conditions can be employed as a good approximate value of atmospheric conditions.



**Figure R1.** Calculated  $\sigma_{abs}$  under different RH values.

*Comment:* (5) A recent work showed BC-catalyzed sulfate formation (An unexpected catalyst dominates formation and radiative forcing of regional haze, Proc. Natl. Acad. Sci. USA 117, 3960, 2020), which is primarily responsible for their optical properties under polluted conditions. How would the BC aging processes, i.e., reactive (catalyzed) versus physical (condensation/partitioning) would impact their proposed framework?

**Reply:** Thanks for the comment. The BC-catalyzed sulfate formation and condensation/partitioning process would result in the different chemical compositions of the BC coating materials, which in theory would impact the refractive index of the coating and further influence the optical properties. However, once the refractive index of the coating material is determined by other methods, the relationship between the light absorption enhancement, mass ratio, and BC mixing states can be determined.

*Comment:* Minor suggestions. Improvement in English is still necessary. I identified some grammar errors below.

Comment: Line 13, replace "its" by "the".

*Reply:* Thanks for the comment. We replace "its" with "the".

Comment: Line 14, delete "thickness".

*Reply:* Thanks for the comment. We deleted 'thickness'.

*Comment:* Line 149, add a conjunction between two parallel sentences. *Reply:* Thanks for the comment. We revised the manuscript.

Reference:

Yu, Y., Zhao, C., Kuang, Y., Tao, J., Zhao, G., Shen, C., and Xu, W.: A parameterization for the light scattering enhancement factor with aerosol chemical compositions, Atmospheric Environment, 10.1016/j.atmosenv.2018.08.016, 2018.