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 (χ) 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 χ for the same MR, indicating that χ 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 χ. 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 κ 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 (σ_{abs}) under different relative humidity (RH) are shown in Fig. R1. Results show that the σ_{abs} ranges between 40.5 and 36.5 when the RH ranges between 40% and 90%. Thus the σ_{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.

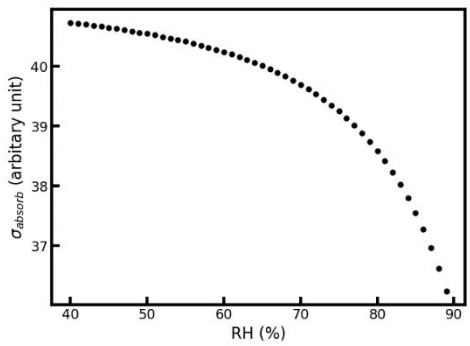

Figure R1. Calculated σ_{abs} under different RH values.

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.

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 $\frac{d^2N}{\mathrm{dlogDp \cdot dlogDc}}$ (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 χ = 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.

Comments: This papers uses a combination of an SP2 with a sizing instrument to deliver information concerning BC mixing state, which is topic of much interest us. We would like to take this opportunity to make the authors aware of a previous work we have published on the subject (Yu et al., 2020), which also used monodisperse SP2 measurements to generate the Riemer-style mixing state metrics, albeit classified using a CPMA rather than a DMA. Can the authors comment on how comparable the metrics produced by the two techniques are?

Reply: Thanks for the comment. Yu et al. (2020) provided the detailed single-particle level mixing state information, which can contribute to future studies concerning BC lifetime and transportation to help to constrain the simulation of BC radiative effects. The mixing state metrics proposed in our method are basically the same as that of Yu et al. (2020). The derived mixing state index from the CPMA-SP2 systems without any assumptions while that from the DMA-SP2 systems with the assuming that the effective density of the BC-containing coating material are the same as that of non-BC particles.

However, our manuscript mainly focus on the relationship between the mixing state index with light absorption enhancement and demonstrating that the mixing state index can be further used in model to quantify the BC light absorption enhancement. Our study offer new insights that the mixing state index can contribute to improvements in the accuracy of simulating the BC radiative effects.

We added some discussions in the manuscript.

Yu, C., Liu, D., Broda, K., Joshi, R., Olfert, J., Sun, Y., Fu, P., Coe, H., and Allan, J. D.:

Characterising mass-resolved mixing state of black carbon in Beijing using a morphology-independent measurement method, Atmospheric Chemistry and Physics, 20, 3645-3661, 10.5194/acp-20-3645-2020, 2020.

Method to Quantify the Black Carbon Aerosol Light Absorption Enhancement with ~~Entropy and Diversity Measures~~ Mixing State Index

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Abstract

Large uncertainties remain when estimating the warming effects of ambient black carbon (BC) aerosols on climate. One of the key challenges in modeling the radiative effects is predicting the BC light absorption enhancement, which is mainly determined by ~~its~~ the mass ratio of non-BC coating ~~thickness~~ material to BC in the population of BC-containing aerosols (MR). For the same MR, recent researches find that the radiative absorption enhancements by BC are also controlled by its particle-to-particle heterogeneity. In this study, the BC mixing state index (χ) is developed to quantify the dispersion of ambient black carbon aerosol mixing states based on binary systems of BC and other non-black carbon components. We demonstrate that the BC light absorption enhancement increases with χ for the same MR, which indicates that χ can be employed as a factor to constrain the light absorption enhancement of ambient BC. Our framework can be further used in the model to study the black carbon radiative effects on climate change.

1 Introduction

Black carbon (BC) aerosols absorb solar radiation, thus exert warming effects on the earth's energy system (Bond and Bergstrom, 2006;Bond et al., 2013). However, large uncertainties remain when quantifying the BC warming effects (Cui et al., 2016;Jacobson, 2010;Koch et al., 2009;Menon et al., 2002). Most of the BC particles were emitted from incomplete combustion of ~~bio~~ bio-fossil

fuel (Bond et al., 2013). After initially emitted, the BC particles would experience aging processing with some other non-BC components coated on the BC particles (Peng et al., 2017;Peng et al., 2016). During the aging processing, the light absorption of BC aerosols would increase, which is well known as "lensing effects" (Saleh et al., 2013;Saleh et al., 2014). One critical challenge in estimating the BC warming effects is quantifying the "lensing effects" of ambient BC aerosols (Liu et al., 2017).

The light absorption enhancement (E_{abs}), which is the ratio of light absorption of BC aerosols with the coating to that of bare BC particles, is proposed to quantify the "lensing effects". Comprehensive studies have been carried out to study the E_{abs} (Liu et al., 2017;Peng et al., 2016;Liu et al., 2015;Fierce et al., 2016;Fierce et al., 2020;Cappa et al., 2012). However, a large discrepancy remains between the results of E_{abs} from field measurements and laboratory studies. The measured E_{abs} of laboratory generated monodisperse BC particles can reach up to a factor of 2, which is consistent with the results from the Mie scattering model (Cappa et al., 2012;Cappa et al., 2019). However, some field measurement shows that the E_{abs} of ambient BC aerosols are relatively small, with 1.06 at California (Cappa et al., 2012), 1.07 in South China (Lan et al., 2013), and 1.10 in Japan (Nakayama et al., 2014), while the measured E_{abs} of ambient BC reaches 1.59 during summer time in Beijing (Xie et al., 2019).

Many factors, such as the morphology of the BC core, the position of BC core inside coating, the coating thickness, chemical properties of coating materials, and size distribution of the BC, would influence the E_{abs} of ambient BC aerosols. Wu et al. (2018) reported that the BC light absorption properties vary significantly for different morphology from the calculation of models. Laboratory studies also find that the light absorption properties of the BC core were tuned due to the change of the BC core morphology (Yuan et al., 2020). Comparing with the concentric spherical structure, the off-center coated BC aggregates would lead to up to a 31% reduction in E_{abs} by the multiple-sphere T-matrix method (Zhang et al., 2017). It has been well studied that the E_{abs} is highly related with the mass ratio of coating materials and BC core (MR) (Liu et al., 2014;Liu et al., 2017). The coating material are also critical in regulating the morphology and optical properties as the coating of sulfuric acid has been shown to be more efficient in altering the BC morphology and light absorption(Zhang

et al., 2008;Xue et al., 2009b, a). Zhao et al. (2019b) reported that the light absorption properties of ambient BC particles are influenced by BC mass size distribution. Besides, recently researchers found that the E_{abs} are also controlled by particle-to-particle heterogeneity (Fierce et al., 2016;Fierce et al., 2020). As shown in Fig.1, the E_{abs} of ambient aerosols for the same MR would vary by about 30%, which is consistency with the results of Fierce et al. (2020). However, there is no study, to our best knowledge, that constrains the uncertainties of the E_{abs} for the same MR.

In this study, we developed a BC mixing states index (χ) to quantify the dispersion of black carbon aerosol mixing states based on binary systems of BC and other non-black carbon components. We demonstrate that the BC E_{abs} increases with χ for the same MR based on the field measurement, which indicates that χ can be employed as a factor to constrain the E_{abs} properties of ambient BC.

2 Data and methods

2.1 Field measurement

The field measurements were conducted at a suburban site Taizhou (119°57' E, 32°35' N) from 26 May to 18 June. As shown in Fig. S1, the Taizhou site lies between two large cities of Nanjing and Shanghai, where the aerosols can be seen as representative that of the Yangtze River Delta area (Liu et al., 2020). More details of the field measurements can refer to Zhao et al. (2019a). During the field measurement, we placed all of the instruments in a container where the temperature was carefully controlled between 22 and 26 ℃. A PM_{10} impactor, which is about 5 meters above the ground, was mounted on the top of the container. The sample aerosols were drawn from the impactor and then dried by a Nafion dryer tube.

The size-resolved BC ~~mixing states~~core distribution and non-BC coating thickness were measured by using a differential mobility analyzer (DMA, model 3081, TSI, USA) in tandem with a single-particle soot photometer (SP2, Droplet Measurement Technologies, USA). Detailed information on the DMA can refer to Zhao et al. (2019c). SP2 can measure the BC mass concentration from the incandescence signals emitted by the BC particle, which is heated to around 6000 K by laser with a wavelength of 1064 nm (Zhao et al., 2020b). Along with the measurement of size-resolved BC ~~mixing states~~distributions, a nephelometer (Aurora 300, Ecotech, Australia)

(Müller et al., 2011) was employed to measure the aerosol scattering coefficient (σ_{sca}) at the wavelength of 525 nm.

2.2 BC mixing states from DMA-SP2 system

In this study, the SP2 was placed after the DMA to measure the size-selected distribution of BC-core and non-BC coating thickness~~mixing states of the quasi-monodisperse aerosols~~. The schematic instrument setup is shown in Fig. S1 and the details can refer to part 1 in the supplementary material. After careful calibrations of the SP2 (part 2.1 in the supplementary material), transformations of the measured signals to BC mass concentrations (part 2.2 in the supplementary material), and multiple charging corrections (part 2.3 in the supplementary material), the BC-containing number concentration distribution under different total diameter (Dp) and BC core diameter (Dc) can be calculated, as shown in Fig. S4 (b). The details of the calculation of size-resolved distribution of BC ~~mixing states~~core and coating thickness from the DMA-SP2 system can refer to Zhao et al. (2020a). The measured size-resolved distribution of BC ~~mixing states~~core and coating thickness as in Fig. S4(b) were used for further analysis. It should be mentioned that the measured number distribution of BC-containing aerosols is two dimensional ($\frac{d^2N}{\mathrm{dlogDp \cdot dlogDc}}$ ~~$\frac{dN}{\mathrm{dlogDp \cdot dlogDc}}$~~). As noted by Zhao et al. (2020b), the SP2 can only detect these BC-containing aerosols with 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.

2.3 Calculating the aerosol optical properties

2.3.1 Calculating the aerosol absorption coefficient for a given Dp and Dc

A Mie scattering model (Bohren and Huffman, 2007) was employed to calculate the aerosol absorption coefficient (σ_{abs}). When calculating the σ_{abs} of single particle, the Mie scattering model requires the diameter of the core, the coating thickness, the refractive index of the core, and the refractive index of the shell. The refractive index of the core adopted here is 1.67+0.67i, which is the calculated mean value by comparing the measured light absorption and calculated light absorption properties (Zhao et al., 2020a). The refractive index of the shell is chosen to be 1.46+0i, which is

assumed to be as that of the non-BC component measured by the DMA-SP2 system (Zhao et al., 2019a;Zhao et al., 2019c). With the above information, the σ_{abs} values at a given Dp and a given Dc can be calculated.

2.3.2 Calculating the aerosol bulk absorption coefficient

We calculate the single-particle σ_{abs} of different Dp and Dc with the given refractive index of core and shell and then the ambient aerosol σ_{abs} distributions at different Dp and Dc ($\frac{d^2\sigma_{abs}}{dlogDp \cdot dlogDc}\frac{d\sigma_{abs}}{dlogDp \cdot dlogDc}$) can be calculated by multiplying the number concentrations of the BC-contained aerosols ($\frac{d^2N}{dlogDp \cdot dlogDc}\frac{dN}{dlogDp \cdot dlogDc}$). By integrating the $\frac{d^2\sigma_{abs}}{dlogDp \cdot dlogDc}\frac{d\sigma_{abs}}{dlogDp \cdot dlogDc}$ over different Dc values, the ambient aerosol σ_{abs} distribution along with different Dp ($\frac{d\sigma_{abs}}{dlogDp}$) can be calculated. The total σ_{abs} of the ambient BC-containing aerosols can be calculated by integrating the $\frac{d\sigma_{abs}}{dlogDp}$ over different Dp values.

2.3.3 Calculating the aerosol E_{abs}

Along with calculating the ~~$\sigma_{abs,Dp,Dc}$~~ $\sigma_{abs}(Dp, Dc)$ of single-particle for different Dp and Dc, we calculate the corresponding light absorption ($\sigma_{abs}(Dc, Dc)$~~$\sigma_{abs,\ Dc,\ Dc}$~~) value for Dc without thickness. The corresponding total light absorption of all measured BC-contained aerosols without ~~thickness~~coating can be calculated by integrating the calculated $\sigma_{abs}(Dc, Dc)$~~$\sigma_{abs,\ Dc,\ Dc}$~~ among different Dp and Dc weighted with $\frac{d^2N}{dlogDp \cdot dlogDc}\frac{dN}{dlogDp \cdot dlogDc}$. Thus the ambient BC particles without coating ($\sigma_{abs}(Dp = Dc)$~~$\sigma_{abs,Dp=0}$~~) can be calculated. The bulk ambient aerosol E_{abs} can thus be calculated with $E_{abs} = \frac{\sigma_{abs}}{\sigma_{abs}(Dp=Dc)\sigma_{abs,\ Dp=0}}$.

2.4 Quantifying ~~the dispersion of~~ BC mixing states

In this study, the mass-weighted mixing state index for BC-containing particles (χ) is developed to investigate the distribution of non-BC material across the BC-containing particle population, which is essentially the same as that of Yu et al. (2020). As for BC particles with known Dp and Dc, the mass concentration of BC core and coating material can be calculated with the effective density of BC core and coating material. The effective density of the BC core is calculated in detail in section 2.2 in the supplement. The effective density of the coating material is assumed to be the same

as the measured effective density of non-BC aerosols by using a centrifugal particle mass analyzer (version 1.53, Cambustion Ltd, UK) in tandem with a scanning mobility particle sizer system (Zhao et al., 2019a) and a mean value of 1.5 g/cm^3 was used here.

For each of the particle *i (i=1,2,.., N* is the measured BC-containing aerosol number concentration), we can calculate its mass ratio of BC with

$$p_{i,BC} = \frac{m_{i,BC}}{m_i}, \quad (1)$$

where $m_{i,BC}$ is the mass concentration of BC and m_i is the total mass concentration of particle *i*. The mass portion of BC can be calculated as

$$p_{BC} = \frac{m_{BC}}{m_{tot}}, \quad (2)$$

were m_{BC} (the total mass concentration of BC) and m_{tot} (total mass of BC-containing aerosols) can be calculated as $m_{BC} = \sum_{i=1}^{N} m_{i,BC}$, $m_{tot} = \sum_{i=1}^{N} m_i$. The MR is calculated as:

$$\mathrm{MR} = \frac{(m_{tot} - m_{BC})}{m_{BC}}, \quad (3)$$

The mass portion of particle *i* to total BC-containing aerosols is calculated as

$$p_i = \frac{m_i}{m_{tot}}. \quad (4)$$

With the definition above, we can calculate the mixing entropy of particle *i* (H_i) by:

$$H_i = -\left(p_{i,BC} ln(p_{i,BC}) + (1 - p_{i,BC}) ln(1 - p_{i,BC})\right), \quad (5)$$

the average mixing entropy of ~~each particle~~the population by:

$$H_\alpha = \sum_{i=1}^{N} p_i H_i, \quad (6)$$

And the population bulk mixing entropy by:

$$H_\gamma = -\left(p_{BC} ln(p_{BC}) + (1 - p_{BC}) ln(1 - p_{BC})\right). \quad (7)$$

Then the average particle species diversity can be calculated by

$$D_\alpha = e^{H_\alpha}, \quad (8)$$

And the bulk population species diversity can be calculated by

$$D_\gamma = e^{H_\gamma}, \quad (9)$$

With the above information, the dispersion of BC particle mixing states can be defined as:

$$\chi = \frac{D_\alpha - 1}{D_\gamma - 1}. \quad (10)$$

The basic idea of quantifying the BC particle mixing states is the same as that of Riemer and West (2013) and Riemer et al. (2019), their framework mainly focuses on the bulk ambient aerosols with about five species (Bondy et al., 2018;Ye et al., 2018). A number of different (binary) species definitions for χ have been used in the literature. Ching et al. (2017) used this index to study the impact of mixing of hygroscopic and non-hygroscopic species on cloud condensation nuclei. Dickau et al. (2016) quantified the volatile and nonvolatile species mixing characters. 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. ssOur developed χ is a reduced parameter that only concerns the BC-containing aerosols with two species of BC component and non-BC coating materials.

3. Results and Discussions

3.1 BC mixing states diagram

A mixing state diagram as shown in Fig. 2 was employed for better understanding the dispersion of BC mixing states. Nine different ~~group bulks of~~ aerosols populations were given and summarized in Table 1. For each group, we include six BC-containing particles with different mass concentrations of BC core and non-BC coating material.

For group 1, the amounts of BC are very small (near zero) and most of the aerosols are composed of the non-BC component. The D_α and D_γ values are 1.00 and 1.00 respectively. These groups can also be described as all of the particles are pure BC particles without coating.

For groups 2, 3, and 4, the mass concentration ratios of the BC component to the non-BC component are 1:5, 2:4, and 3:3 respectively. All of the D_α values are 1.00 for groups 2, 3, and 4 because the BC particles are externally mixed. The corresponding D_γ values are 1.56, 1.89, and 2.00 respectively. For these three groups, the χ values are all 0.00.

For groups 4, 5, 6, and 7, the mass concentration ratios of the BC component to the non-BC component are all 1:1 while the BC component is mixed to a different extent. It is easy to conclude that the BC particles of group 7 are most well mixed among these four groups. The corresponding χ values are 0, 0.26, 0.83, and 1.0 for group 4, 5, 6, and 7, respectively.

As for groups 8 and 9, the mass concentration ratios of the BC component to the non-BC component are 1:6.1. The D_γ values are 1.5 and the D_α values are 1.5 and 1.35 respectively.

From the different group, the average particle species diversity 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. The bulk population species diversity D_α ranges between 1 and D_γ. It denotes the diversity of different BC-containing particles.

3.2 Overview of the measurement

Fig.S6 gives the time series of our field measurements results. During the field measurement, the σ_{sca} varies between 29 and 1590 Mm^{-1}. The ranges of H_α, H_γ, D_α, D_γ, and χ are 0.10~0.55, 0.42~0.64, 1.32~1.72, 1.52~1.91 and 0.62~0.82 respectively.

For a better understanding of the characteristics of the above parameters, we only present the time series of these parameters during a pollution period between 27, May and 30, May in Fig. 3. As shown in Fig. 3, the MR increased from about 2 to 4 when the σ_{sca} increased from 300 to 1200 Mm^{-1}, which indicates that some secondary aerosol components were coated on the BC particles when the ambient air is more polluted. During the aging processing, the H_α decreased from 0.51 to 0.38 and H_γ decreased from 0.63 to 0.49. The D_α decreases ~~with the MR~~ from 1.66 to 1.48. The D_γ decreases with the MR from 1.86 to 1.66, which is consistent with the results in section 3.1 that the D_γ~~D_α~~ should decrease with the MR when the MR is larger than 1. The χ varies between 0.68 and 0.79. It is worth noting that the χ is not well correlated with the pollution conditions.

The daily variation of σ_{sca}, which is highly related to the development of the boundary layer, reaches its maximum value of 525 Mm^{-1} at 6:00 AM and a minimum value of 150 Mm at 7:00 PM. The daily variation of MR is largest at 5:00 AM with a mean value of 3.16 and reaches its minimum value of 2.56 at 7:00 PM. The daily variation of MR was mainly influenced by aging processing and anthropogenic activities. During the daytime, the newly emitted BC particles due to anthropogenic activities have low MR and the measured mean MR is low than that at night. The D_α values, which are anti-correlated with MR, show the opposite trend with MR. As for χ, it is smaller in the daytime

than that at night. The lower χ values at daytime mainly resulted from the mixing of newly emitted BC particles due to anthropogenic activities and some pre-existed aged BC particles.

3.3 Relationship between the χ and E_{abs} from measurement

For each of the measured group of size-resolved distribution of BC core and coating thickness~~BC mixing states~~, we calculated the corresponding MR, χ, and E_{abs}. And the relationship between the MR and absorption enhancement is summarized in Fig. 5. It should be noted that the shown BC population is only one of possible examples with χ equaling 0, 0.81, and 1 respectively. There are many other possible ways the particle composition can be arranged that would give the same mixing state index.

Overall, the BC E_{abs} increase with MR, which is consistent with the previous knowledge. For a given value of MR, E_{abs} varies by about 20%, especially for these conditions with MR larger than 1.0. When MR is larger than 1.0, the E_{abs} increase with the χ. Relationship between the E_{abs} and χ is rather complex when MR is smaller than 1.0. However, only 448 of 6948 groups (6.4%) of the measured MR values are smaller than 1. Therefore, for most of the conditions, the measured E_{abs} should increase with χ, which indicates that the ~~refractive index of~~BC mixing state index χ can be employed as a factor to constrain the E_{abs} of ambient aerosols.

A schematic diagram as shown in Fig. 6 to denotes the relationship between the E_{abs} and χ. From Fig. 6, we calculated the E_{abs} and χ under differ MR and then compared the E_{abs} of different bulk aerosols. The first group contains two particles with both the MR equaling 8. The corresponding χ is 1.00 and E_{abs} is 1.60. Another group of particles contains two particles with MR equaling 1 and 15, respectively. Thus the second group of particles has a mean MR of 8. The calculated corresponding χ and E_{abs} are 0.79 and 1.42 respectively. Thus, the E_{abs} tend to increase with χ for the same MR, which is mainly resulted from that the increasing ratio of E_{abs} (the slope of E_{abs} to MR) decrease with MR.

It is worth noting that the increasing ratio is almost the same when the MR is in the range of 0 and 3. Therefore, the E_{abs} doesn't tend to increase with the χ when the MR was less than 1, which is consistent with our study as shown in Fig. 6.

3.4 Relationship between the χ and E_{abs} from simulation

A Mont-Carlo simulation was carried out for a better understanding of the relationship between χ and E_{abs}. During the simulation, the number of BC-containing particles was assumed to be 30. For each of the BC particle, the core diameter of the BC particle was randomly generated with a geometric mean diameter of 130.7 nm and a geometric standard deviation of 1.5, which is the mean measurement results of the BC core distribution during the field measurement (Zhao et al., 2020b). The corresponding MR of the BC particle is assumed to be in the range between 0.0 (pure BC particles without coating) and 78.0 (particles with a core diameter of 130 nm and a total diameter of 560 nm). For each of the group of particles, the corresponding aerosol bulk MR, E_{abs} and χ can be calcualted. The simulations were conducted for 10^7 times, and the calculated mean and standard deviation of E_{abs} under different MR and χ are summarized in Fig. 7 (a) and (b).

From Fig. 7 (a), the calculated E_{abs} tend to increase with MR for each of the given χ, which is consistent with the previous knowledge of the BC light absorption properties. Then the MR is smaller than 2, the calculated E_{abs} does not seem to increase with the χ, which is consistent with the analyzed results from section 3.3 and Fig. 6. When the MR is larger 2, the E_{abs} tend to increase with the χ. The larger the MR is, the E_{abs} is more sensitive to χ. Two reasons may lead to this phenomenon. One reason is that that calculated slope of E_{abs} to MR for one particle as shown in Fig. 6 decreases with the MR. Another reason is that the calculated E_{abs} range increase with MR when the χ changes between 0 and 1 as shown in Fig. 5.

As for the uncertainties of simulated E_{abs}, it tends to increase with the MR, which is consistent with the previous discussions that the E_{abs} the range tends to increase with MR. Overall, the calculated standard deviations of E_{abs} are all the way smaller than 10% for different MR and χ. Therefore, the calculated E_{abs} can be well constrained by χ.

4 Conclusion

Larger uncertainties remain when estimating the warming effects of ambient BC aerosols due to the poor understanding of the ambient BC light absorption enhance ratio. Previous studies find that the light absorption of ambient aerosols was mainly determined by the morphology of the BC core, the position of the BC core inside coating, the coating thickness, and the size distribution of the BC.

We find that there are more than 20% of uncertainties for the same measured mean coating thickness, i.e. the same measured MR based on the field measurement of the size-resolved distribution of BC core and coating thickness~~BC mixing states~~. However, there were no-study, to our best knowledge, that attempts to constrain the uncertainties.

In this study, we developed the BC mixing states index χ based on the mass concentrations of BC components and non-BC material of each BC-containing particle. Results show that the light absorption enhancement ratio E_{abs} tend to increase the χ for the same measured MR. Therefore, our developed parameter χ, which reflects the dispersion of the BC mixing states, can be employed as an effective parameter to constrain the light absorption enhancement of ambient BC-containing aerosols.

The new finding of our study is ~~Our study offer new insight~~ 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 introduce 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.

Data availability. The data involved is available in the manuscript.

Author contributions. Gang Zhao wrote the manuscript. Chunsheng Zhao, Min Hu, Tianyi Tan, Song Guo, Zhijun Wu, Yishu Zhu and Gang Zhao discussed the results.

Competing interests. The authors declare that they have no conflict of interest.

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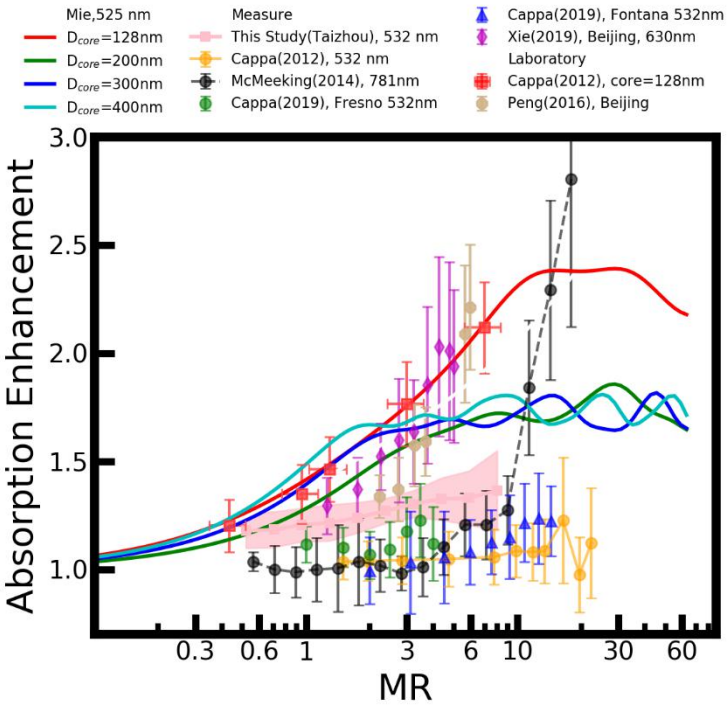

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447 Figure 1. The measured E_{abs} of BC particles from different ambient measurements, including this

448 work (in pink), and lab studies.

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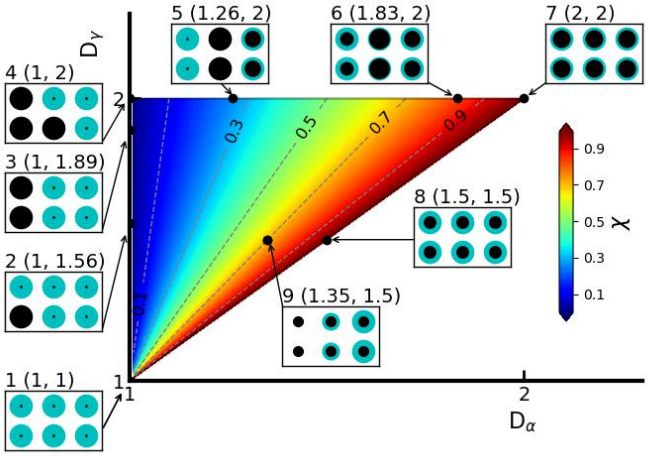
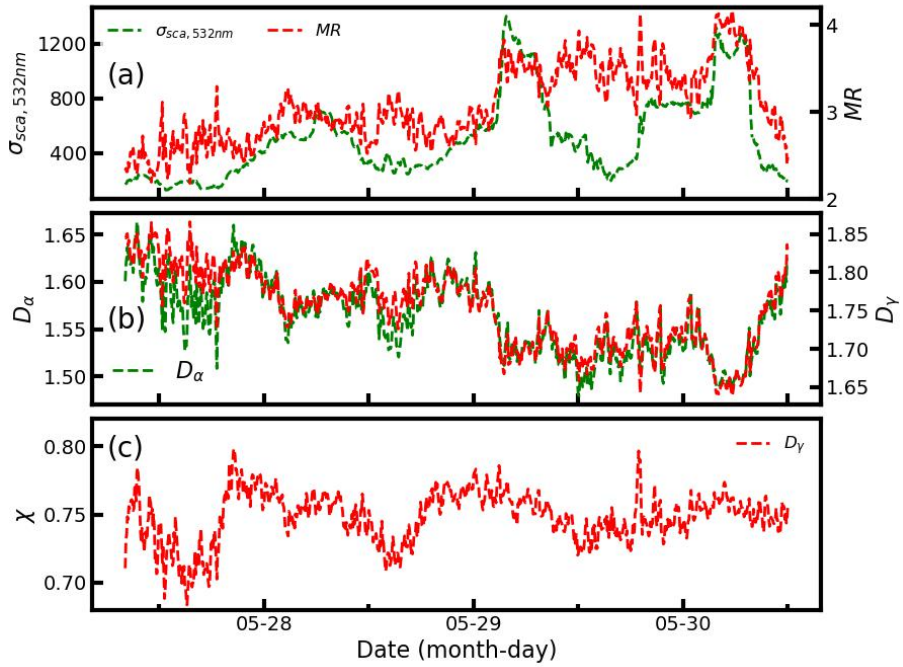


Figure 2. Mixing states diagram to illustrate the relationship between D_α, D_γ, and χ. Each species consists of six particles, and the colors of black and cyan represent the BC and non-BC components.



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455 **Figure 3.** Measured time series of (a) σ_{sca} and MR, (b) ~~H_α and H_γ, (c)~~ D_α and D_γ, and (~~d~~c) χ.

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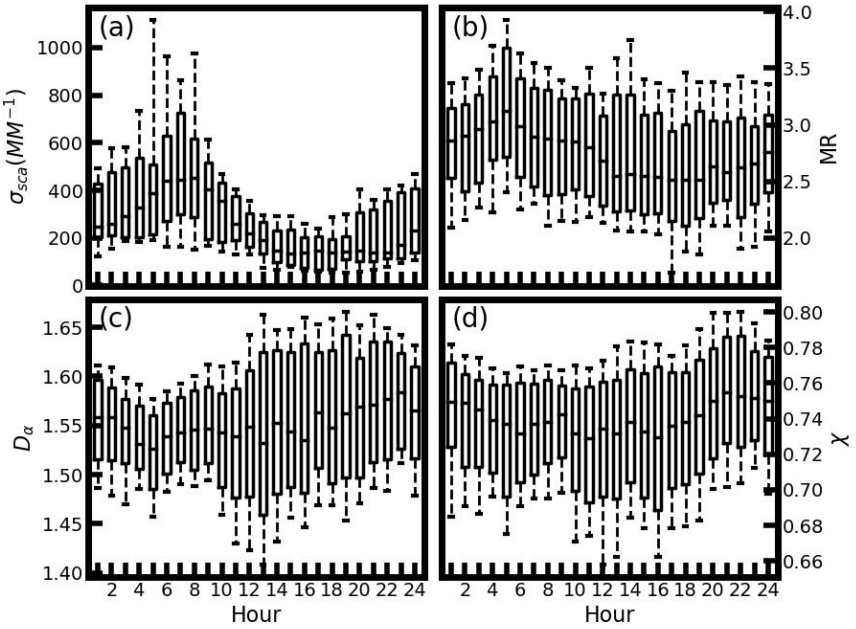

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458 **Figure 4.** Daily variation of the measured (a) σ_{sca}, (b) MR, (c) D_α, and (d) χ.

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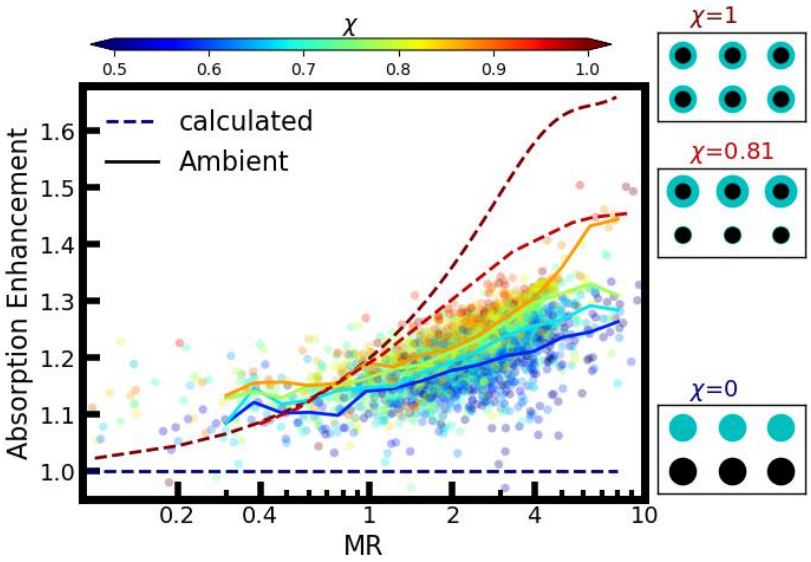


Figure 5. Relationship between the BC E_{abs} and the measured mass ratio of the BC-containing aerosols coating material to BC under different χ conditions. Four solid lines from bottom to up corresponding to the measured ambient size-resolved BC mixing states data with χ ranges of 0.575~0.625, 0.625~0.675, 0.675~0.725, and 0.725~0.775. The dotted line corresponds to the χ of 0.0 (blue), 0.81 (light red), and 1.0 (dark red), respectively.

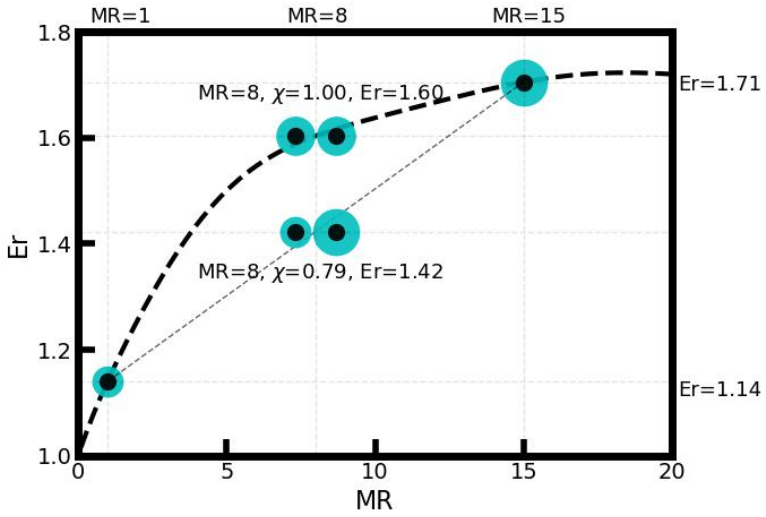
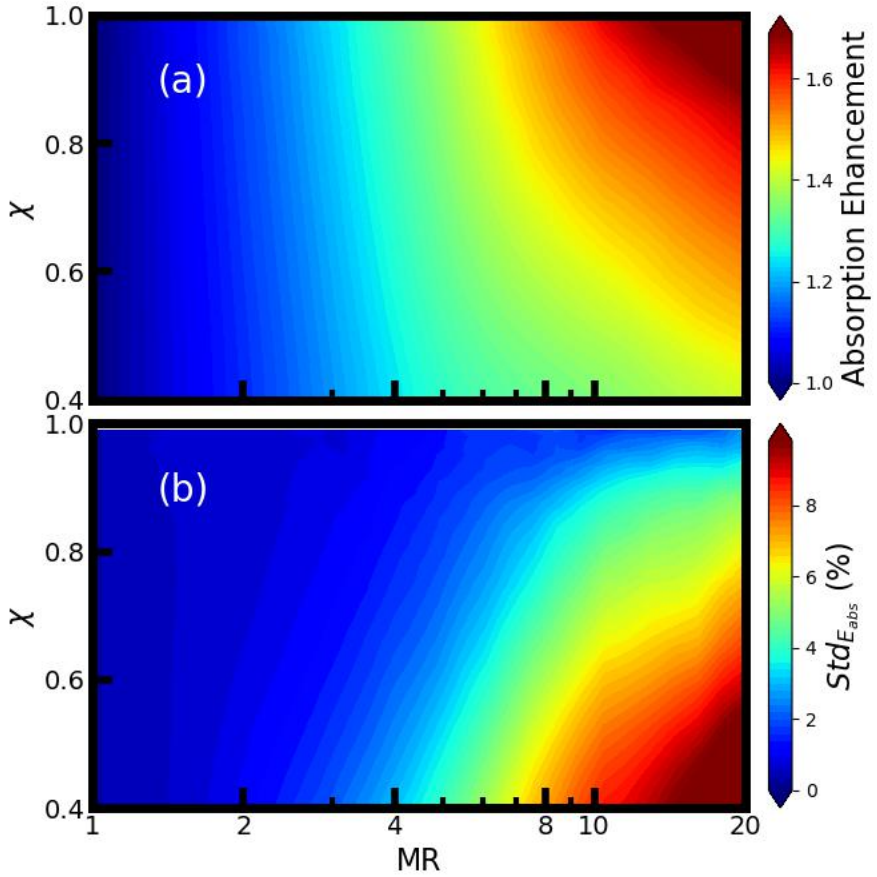


Figure 6. Schematic diagram that denotes the relationship between χ and Er.



471

472 **Figure 7.** The calcaulted (a) mean E_{abs} values and (b) standard deviations of the E_{abs} values for
473 different MR and χ.

474

ID	(D_α, D_γ)	χ	P1[*1]	P2[*1]	P3[*1]	P4[*1]	P5[*1]	P6[*1]	Tot[*1]
1	(1.00, 1.00)	-	(10^{-9},1)	(10^{-9},1)	(10^{-9},1)	(10^{-9},1)	(10^{-9},1)	(10^{-9},1)	(6·10^{-9},1)
2	(1.00,1.56)	0	(1,10^{-9})	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(1, 5)
3	(1.00, 1.89)	0	(1,10^{-9})	(1,10^{-9})	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(2,4)
4	(1.00, 2.00)	0	(1,10^{-9})	(1,10^{-9})	(1,10^{-9})	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(3,3)
5	(1.26, 2.00)	0.26	(2,10^{-9})	(2,10^{-9})	(10^{-9},2)	(10^{-9},2)	(10^{-9},1)	(1,1)	(6, 6)
6	(1.83, 2.00)	0.83	(1,3)	(1,3)	(3,1)	(3,1)	(2,2)	(2,2)	(12,12)
7	(2.00, 2.00)	1.00	(1,1)	(1,1)	(1,1)	(1,1)	(1,1)	(1,1)	(6,6)
8	(1.5, 1.50)	1.00	(1,6.1)	(1,6.1)	(1,6.1)	(1,6.1)	(1,6.1)	(1,6.1)	(6, 36.6)
9	(1.35, 1.50)	0.70	(1,10^{-9})	(1,10^{-9})	(1,6.1)	(1,6.1)	(1,12.2)	(1,12.2)	(6, 36.6)

475 **Table 1.** Detail information of the BC particles shown in Fig.2

476 [*1] Mass of the BC component of and non-BC component (arbitrary unit).

Method to Quantify the Black Carbon Aerosol Light Absorption Enhancement with Mixing State Index

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Abstract

Large uncertainties remain when estimating the warming effects of ambient black carbon (BC) aerosols on climate. One of the key challenges in modeling the radiative effects is predicting the BC light absorption enhancement, which is mainly determined by the mass ratio of non-BC coating material to BC in the population of BC-containing aerosols (MR). For the same MR, recent researches find that the radiative absorption enhancements by BC are also controlled by its particle-to-particle heterogeneity. In this study, the BC mixing state index (χ) is developed to quantify the dispersion of ambient black carbon aerosol mixing states based on binary systems of BC and other non-black carbon components. We demonstrate that the BC light absorption enhancement increases with χ for the same MR, which indicates that χ can be employed as a factor to constrain the light absorption enhancement of ambient BC. Our framework can be further used in the model to study the black carbon radiative effects on climate change.

1 Introduction

Black carbon (BC) aerosols absorb solar radiation, thus exert warming effects on the earth's energy system (Bond and Bergstrom, 2006;Bond et al., 2013). However, large uncertainties remain when quantifying the BC warming effects (Cui et al., 2016;Jacobson, 2010;Koch et al., 2009;Menon et al., 2002). Most of the BC particles were emitted from incomplete combustion of bio-fossil fuel

(Bond et al., 2013). After initially emitted, the BC particles would experience aging processing with some other non-BC components coated on the BC particles (Peng et al., 2017;Peng et al., 2016). During the aging processing, the light absorption of BC aerosols would increase, which is well known as "lensing effect" (Saleh et al., 2013;Saleh et al., 2014). One critical challenge in estimating the BC warming effects is quantifying the "lensing effects" of ambient BC aerosols (Liu et al., 2017).

The light absorption enhancement (E_{abs}), which is the ratio of light absorption of BC aerosols with the coating to that of bare BC particles, is proposed to quantify the "lensing effects". Comprehensive studies have been carried out to study the E_{abs} (Liu et al., 2017;Peng et al., 2016;Liu et al., 2015;Fierce et al., 2016;Fierce et al., 2020;Cappa et al., 2012). However, a large discrepancy remains between the results of E_{abs} from field measurements and laboratory studies. The measured E_{abs} of laboratory generated monodisperse BC particles can reach up to a factor of 2, which is consistent with the results from the Mie scattering model (Cappa et al., 2012;Cappa et al., 2019). However, some field measurement shows that the E_{abs} of ambient BC aerosols are relatively small, with 1.06 at California (Cappa et al., 2012), 1.07 in South China (Lan et al., 2013), and 1.10 in Japan (Nakayama et al., 2014), while the measured E_{abs} of ambient BC reaches 1.59 during summer time in Beijing (Xie et al., 2019).

Many factors, such as the morphology of the BC core, the position of BC core inside coating, the coating thickness, chemical properties of coating materials, and size distribution of the BC, would influence the E_{abs} of ambient BC aerosols. Wu et al. (2018) reported that the BC light absorption properties vary significantly for different morphology from the calculation of models. Laboratory studies also find that the light absorption properties of the BC core were tuned due to the change of the BC core morphology (Yuan et al., 2020). Comparing with the concentric spherical structure, the off-center coated BC aggregates would lead to up to a 31% reduction in E_{abs} by the multiple-sphere T-matrix method (Zhang et al., 2017). It has been well studied that the E_{abs} is highly related with the mass ratio of coating materials and BC core (MR) (Liu et al., 2014;Liu et al., 2017). The coating material are also critical in regulating the morphology and optical properties as the coating of sulfuric acid has been shown to be more efficient in altering the BC morphology and light absorption(Zhang

et al., 2008;Xue et al., 2009b, a). Zhao et al. (2019b) reported that the light absorption properties of ambient BC particles are influenced by BC mass size distribution. Besides, recently researchers found that the E_{abs} are also controlled by particle-to-particle heterogeneity (Fierce et al., 2016;Fierce et al., 2020). As shown in Fig.1, the E_{abs} of ambient aerosols for the same MR would vary by about 30%, which is consistency with the results of Fierce et al. (2020). However, there is no study, to our best knowledge, that constrains the uncertainties of the E_{abs} for the same MR.

In this study, we developed a BC mixing states index (χ) to quantify the dispersion of black carbon aerosol mixing states based on binary systems of BC and other non-black carbon components. We demonstrate that the BC E_{abs} increases with χ for the same MR based on the field measurement, which indicates that χ can be employed as a factor to constrain the E_{abs} properties of ambient BC.

2 Data and methods

2.1 Field measurement

The field measurements were conducted at a suburban site Taizhou (119°57' E, 32°35' N) from 26 May to 18 June. As shown in Fig. S1, the Taizhou site lies between two large cities of Nanjing and Shanghai, where the aerosols can be seen as representative that of the Yangtze River Delta area (Liu et al., 2020). More details of the field measurements can refer to Zhao et al. (2019a). During the field measurement, we placed all of the instruments in a container where the temperature was carefully controlled between 22 and 26 ℃. A PM_{10} impactor, which is about 5 meters above the ground, was mounted on the top of the container. The sample aerosols were drawn from the impactor and then dried by a Nafion dryer tube.

The size-resolved BC core distribution and non-BC coating thickness were measured by using a differential mobility analyzer (DMA, model 3081, TSI, USA) in tandem with a single-particle soot photometer (SP2, Droplet Measurement Technologies, USA). Detailed information on the DMA can refer to Zhao et al. (2019c). SP2 can measure the BC mass concentration from the incandescence signals emitted by the BC particle, which is heated to around 6000 K by laser with a wavelength of 1064 nm (Zhao et al., 2020b). Along with the measurement of size-resolved BC distributions, a

nephelometer (Aurora 300, Ecotech, Australia) (Müller et al., 2011) was employed to measure the aerosol scattering coefficient (σ_{sca}) at the wavelength of 525 nm.

2.2 BC mixing states from DMA-SP2 system

In this study, the SP2 was placed after the DMA to measure the size-selected distribution of BC-core and non-BC coating thickness. The schematic instrument setup is shown in Fig. S1 and the details can refer to part 1 in the supplementary material. After careful calibrations of the SP2 (part 2.1 in the supplementary material), transformations of the measured signals to BC mass concentrations (part 2.2 in the supplementary material), and multiple charging corrections (part 2.3 in the supplementary material), the BC-containing number concentration distribution under different total diameter (Dp) and BC core diameter (Dc) can be calculated, as shown in Fig. S4 (b). The details of the calculation of size-resolved distribution of BC core and coating thickness from the DMA-SP2 system can refer to Zhao et al. (2020a). The measured size-resolved distribution of BC core and coating thickness as in Fig. S4(b) were used for further analysis. It should be mentioned that the measured number distribution of BC-containing aerosols is two dimensional ($\frac{d^2N}{\mathrm{dlogDp \cdot dlogDc}}$). As noted by Zhao et al. (2020b), the SP2 can only detect these BC-containing aerosols with 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.

2.3 Calculating the aerosol optical properties

2.3.1 Calculating the aerosol absorption coefficient for a given Dp and Dc

A Mie scattering model (Bohren and Huffman, 2007) was employed to calculate the aerosol absorption coefficient (σ_{abs}). When calculating the σ_{abs} of single particle, the Mie scattering model requires the diameter of the core, the coating thickness, the refractive index of the core, and the refractive index of the shell. The refractive index of the core adopted here is $1.67+0.67i$, which is the calculated mean value by comparing the measured light absorption and calculated light absorption properties (Zhao et al., 2020a). The refractive index of the shell is chosen to be $1.46+0i$, which is assumed to be as that of the non-BC component measured by the DMA-SP2 system (Zhao et al.,

2019a;Zhao et al., 2019c). With the above information, the σ_{abs} values at a given Dp and a given Dc can be calculated.

2.3.2 Calculating the aerosol bulk absorption coefficient

We calculate the single-particle σ_{abs} of different Dp and Dc with the given refractive index of core and shell and then the ambient aerosol σ_{abs} distributions at different Dp and Dc ($\frac{\mathrm{d}^2\sigma_{abs}}{\mathrm{dlogDp}\cdot\mathrm{dlogDc}}$) can be calculated by multiplying the number concentrations of the BC-contained aerosols ($\frac{d^2N}{\mathrm{dlogDp}\cdot\mathrm{dlogDc}}$). By integrating the $\frac{\mathrm{d}^2\sigma_{abs}}{\mathrm{dlogDp}\cdot\mathrm{dlogDc}}$ over different Dc values, the ambient aerosol σ_{abs} distribution along with different Dp ($\frac{\mathrm{d}\sigma_{abs}}{\mathrm{dlogDp}}$) can be calculated. The total σ_{abs} of the ambient BC-containing aerosols can be calculated by integrating the $\frac{\mathrm{d}\sigma_{abs}}{\mathrm{dlogDp}}$ over different Dp values.

2.3.3 Calculating the aerosol E_{abs}

Along with calculating the $\sigma_{abs}(Dp, Dc)$ of single-particle for different Dp and Dc, we calculate the corresponding light absorption ($\sigma_{abs}(Dc, Dc)$) value for Dc without thickness. The corresponding total light absorption of all measured BC-contained aerosols withoutcoating can be calculated by integrating the calculated $\sigma_{abs}(Dc, Dc)$ among different Dp and Dc weighted with $\frac{d^2N}{\mathrm{dlogDp}\cdot\mathrm{dlogDc}}$. Thus the ambient BC particles without coating ($\sigma_{abs}(Dp = Dc)$) can be calculated. The bulk ambient aerosol E_{abs} can thus be calculated with $\mathrm{E}_{abs} = \frac{\sigma_{abs}}{\sigma_{abs}(Dp=Dc)}$.

2.4 Quantifying BC mixing states

In this study, the mass-weighted mixing state index for BC-containing particles (χ) is developed to investigate the distribution of non-BC material across the BC-containing particle population, which is essentially the same as that of Yu et al. (2020). As for BC particles with known Dp and Dc, the mass concentration of BC core and coating material can be calculated with the effective density of BC core and coating material. The effective density of the BC core is calculated in detail in section 2.2 in the supplement. The effective density of the coating material is assumed to be the same as the measured effective density of non-BC aerosols by using a centrifugal particle mass analyzer (version 1.53, Cambustion Ltd, UK) in tandem with a scanning mobility particle sizer system (Zhao et al., 2019a) and a mean value of 1.5 g/cm^3 was used here.

For each of the particle *i (i=1,2,.., N* is the measured BC-containing aerosol number concentration), we can calculate its mass ratio of BC with

$$p_{i,BC} = \frac{m_{i,BC}}{m_i}, \quad (1)$$

where $m_{i,BC}$ is the mass concentration of BC and m_i is the total mass concentration of particle *i*. The mass portion of BC can be calculated as

$$p_{BC} = \frac{m_{BC}}{m_{tot}}, \quad (2)$$

were m_{BC} (the total mass concentration of BC) and m_{tot} (total mass of BC-containing aerosols) can be calculated as $m_{BC} = \sum_{i=1}^{N} m_{i,BC}$, $m_{tot} = \sum_{i=1}^{N} m_i$. The MR is calculated as:

$$\mathrm{MR} = \frac{(m_{tot} - m_{BC})}{m_{BC}}, \quad (3)$$

The mass portion of particle *i* to total BC-containing aerosols is calculated as

$$p_i = \frac{m_i}{m_{tot}}. \quad (4)$$

With the definition above, we can calculate the mixing entropy of particle *i* (H_i) by:

$$H_i = -(p_{i,BC} ln(p_{i,BC}) + (1 - p_{i,BC}) ln(1 - p_{i,BC}), \quad (5)$$

the average mixing entropy of the population by:

$$H_\alpha = \sum_{i=1}^{N} p_i H_i, \quad (6)$$

And the population bulk mixing entropy by:

$$H_\gamma = -(p_{BC} ln(p_{BC}) + (1 - p_{BC}) ln(1 - p_{BC}). \quad (7)$$

Then the average particle species diversity can be calculated by

$$D_\alpha = e^{H_\alpha}, \quad (8)$$

And the bulk population species diversity can be calculated by

$$D_\gamma = e^{H_\gamma}, \quad (9)$$

With the above information, the dispersion of BC particle mixing states can be defined as:

$$\chi = \frac{D_\alpha - 1}{D_\gamma - 1}. \quad (10)$$

The basic idea of quantifying the BC particle mixing states is the same as that of Riemer and West (2013) and Riemer et al. (2019), their framework mainly focuses on the bulk ambient aerosols

with about five species (Bondy et al., 2018;Ye et al., 2018). A number of different (binary) species definitions for χ have been used in the literature. Ching et al. (2017) used this index to study the impact of mixing of hygroscopic and non-hygroscopic species on cloud condensation nuclei. Dickau et al. (2016) quantified the volatile and nonvolatile species mixing characters. 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. ssOur developed χ is a reduced parameter that only concerns the BC-containing aerosols with two species of BC component and non-BC coating materials.

3. Results and Discussions

3.1 BC mixing states diagram

A mixing state diagram as shown in Fig. 2 was employed for better understanding the dispersion of BC mixing states. Nine different aerosols populations were given and summarized in Table 1. For each group, we include six BC-containing particles with different mass concentrations of BC core and non-BC coating material.

For group 1, the amounts of BC are very small (near zero) and most of the aerosols are composed of the non-BC component. The D_α and D_γ values are 1.00 and 1.00 respectively. These groups can also be described as all of the particles are pure BC particles without coating.

For groups 2, 3, and 4, the mass concentration ratios of the BC component to the non-BC component are 1:5, 2:4, and 3:3 respectively. All of the D_α values are 1.00 for groups 2, 3, and 4 because the BC particles are externally mixed. The corresponding D_γ values are 1.56, 1.89, and 2.00 respectively. For these three groups, the χ values are all 0.00.

For groups 4, 5, 6, and 7, the mass concentration ratios of the BC component to the non-BC component are all 1:1 while the BC component is mixed to a different extent. It is easy to conclude that the BC particles of group 7 are most well mixed among these four groups. The corresponding χ values are 0, 0.26, 0.83, and 1.0 for group 4, 5, 6, and 7, respectively.

As for groups 8 and 9, the mass concentration ratios of the BC component to the non-BC component are 1:6.1. The D_γ values are 1.5 and the D_α values are 1.5 and 1.35 respectively.

From the different group, the average particle species diversity 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. The bulk population species diversity D_α ranges between 1 and D_γ. It denotes the diversity of different BC-containing particles.

3.2 Overview of the measurement

Fig.S6 gives the time series of our field measurements results. During the field measurement, the σ_{sca} varies between 29 and 1590 Mm^{-1}. The ranges of H_α, H_γ, D_α, D_γ, and χ are 0.10~0.55, 0.42~0.64, 1.32~1.72, 1.52~1.91 and 0.62~0.82 respectively.

For a better understanding of the characteristics of the above parameters, we only present the time series of these parameters during a pollution period between 27, May and 30, May in Fig. 3. As shown in Fig. 3, the MR increased from about 2 to 4 when the σ_{sca} increased from 300 to 1200 Mm^{-1}, which indicates that some secondary aerosol components were coated on the BC particles when the ambient air is more polluted. During the aging processing, the H_α decreased from 0.51 to 0.38 and H_γ decreased from 0.63 to 0.49. The D_α decreases from 1.66 to 1.48. The D_γ decreases with the MR from 1.86 to 1.66, which is consistent with the results in section 3.1 that the D_γ should decrease with the MR when the MR is larger than 1. The χ varies between 0.68 and 0.79. It is worth noting that the χ is not well correlated with the pollution conditions.

The daily variation of σ_{sca}, which is highly related to the development of the boundary layer, reaches its maximum value of 525 Mm^{-1} at 6:00 AM and a minimum value of 150 Mm at 7:00 PM. The daily variation of MR is largest at 5:00 AM with a mean value of 3.16 and reaches its minimum value of 2.56 at 7:00 PM. The daily variation of MR was mainly influenced by aging processing and anthropogenic activities. During the daytime, the newly emitted BC particles due to anthropogenic activities have low MR and the measured mean MR is low than that at night. The D_α values, which are anti-correlated with MR, show the opposite trend with MR. As for χ, it is smaller in the daytime than that at night. The lower χ values at daytime mainly resulted from the mixing of newly emitted BC particles due to anthropogenic activities and some pre-existed aged BC particles.

3.3 Relationship between the χ and E_{abs} from measurement

For each of the measured group of size-resolved distribution of BC core and coating thickness, we calculated the corresponding MR, χ, and E_{abs}. And the relationship between the MR and absorption enhancement is summarized in Fig. 5. It should be noted that the shown BC population is only one of possible examples with χ equaling 0, 0.81, and 1 respectively. There are many other possible ways the particle composition can be arranged that would give the same mixing state index.

Overall, the BC E_{abs} increase with MR, which is consistent with the previous knowledge. For a given value of MR, E_{abs} varies by about 20%, especially for these conditions with MR larger than 1.0. When MR is larger than 1.0, the E_{abs} increase with the χ. Relationship between the E_{abs} and χ is rather complex when MR is smaller than 1.0. However, only 448 of 6948 groups (6.4%) of the measured MR values are smaller than 1. Therefore, for most of the conditions, the measured E_{abs} should increase with χ, which indicates that the BC mixing state index χ can be employed as a factor to constrain the E_{abs} of ambient aerosols.

A schematic diagram as shown in Fig. 6 to denotes the relationship between the E_{abs} and χ. From Fig. 6, we calculated the E_{abs} and χ under differ MR and then compared the E_{abs} of different bulk aerosols. The first group contains two particles with both the MR equaling 8. The corresponding χ is 1.00 and E_{abs} is 1.60. Another group of particles contains two particles with MR equaling 1 and 15, respectively. Thus the second group of particles has a mean MR of 8. The calculated corresponding χ and E_{abs} are 0.79 and 1.42 respectively. Thus, the E_{abs} tend to increase with χ for the same MR, which is mainly resulted from that the increasing ratio of E_{abs} (the slope of E_{abs} to MR) decrease with MR.

It is worth noting that the increasing ratio is almost the same when the MR is in the range of 0 and 3. Therefore, the E_{abs} doesn't tend to increase with the χ when the MR was less than 1, which is consistent with our study as shown in Fig. 6.

3.4 Relationship between the χ and E_{abs} from simulation

A Mont-Carlo simulation was carried out for a better understanding of the relationship between χ and E_{abs}. During the simulation, the number of BC-containing particles was assumed to be 30. For each of the BC particle, the core diameter of the BC particle was randomly generated with a

geometric mean diameter of 130.7 nm and a geometric standard deviation of 1.5, which is the mean measurement results of the BC core distribution during the field measurement (Zhao et al., 2020b). The corresponding MR of the BC particle is assumed to be in the range between 0.0 (pure BC particles without coating) and 78.0 (particles with a core diameter of 130 nm and a total diameter of 560 nm). For each of the group of particles, the corresponding aerosol bulk MR, E_{abs} and χ can be calcualted. The simulations were conducted for 10^7 times, and the calculated mean and standard deviation of E_{abs} under different MR and χ are summarized in Fig. 7 (a) and (b).

From Fig. 7 (a), the calculated E_{abs} tend to increase with MR for each of the given χ, which is consistent with the previous knowledge of the BC light absorption properties. Then the MR is smaller than 2, the calculated E_{abs} does not seem to increase with the χ, which is consistent with the analyzed results from section 3.3 and Fig. 6. When the MR is larger 2, the E_{abs} tend to increase with the χ. The larger the MR is, the E_{abs} is more sensitive to χ. Two reasons may lead to this phenomenon. One reason is that that calculated slope of E_{abs} to MR for one particle as shown in Fig. 6 decreases with the MR. Another reason is that the calculated E_{abs} range increase with MR when the χ changes between 0 and 1 as shown in Fig. 5.

As for the uncertainties of simulated E_{abs}, it tends to increase with the MR, which is consistent with the previous discussions that the E_{abs} the range tends to increase with MR. Overall, the calculated standard deviations of E_{abs} are all the way smaller than 10% for different MR and χ. Therefore, the calculated E_{abs} can be well constrained by χ.

4 Conclusion

Larger uncertainties remain when estimating the warming effects of ambient BC aerosols due to the poor understanding of the ambient BC light absorption enhance ratio. Previous studies find that the light absorption of ambient aerosols was mainly determined by the morphology of the BC core, the position of the BC core inside coating, the coating thickness, and the size distribution of the BC. We find that there are more than 20% of uncertainties for the same measured mean coating thickness, i.e. the same measured MR based on the field measurement of the size-resolved distribution of BC

core and coating thickness. However, there were no-study, to our best knowledge, that attempts to constrain the uncertainties.

In this study, we developed the BC mixing states index χ based on the mass concentrations of BC components and non-BC material of each BC-containing particle. Results show that the light absorption enhancement ratio E_{abs} tend to increase the χ for the same measured MR. Therefore, our developed parameter χ, which reflects the dispersion of the BC mixing states, can be employed as an effective parameter to constrain the light absorption enhancement of ambient BC-containing aerosols.

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 introduce 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.

Data availability. The data involved is available in the manuscript.

Author contributions. Gang Zhao wrote the manuscript. Chunsheng Zhao, Min Hu, Tianyi Tan, Song Guo, Zhijun Wu, Yishu Zhu and Gang Zhao discussed the results.

Competing interests. The authors declare that they have no conflict of interest.

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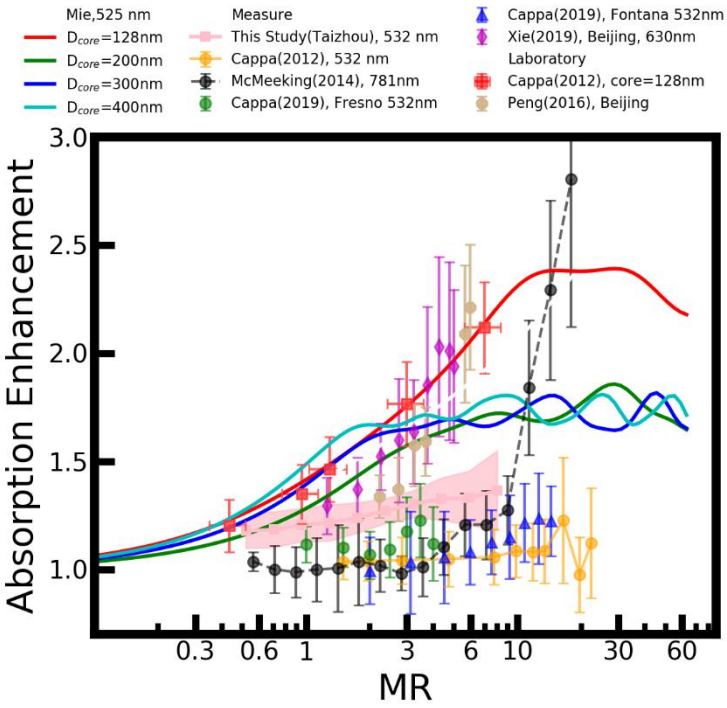

441

442 Figure 1. The measured E_{abs} of BC particles from different ambient measurements, including this

443 work (in pink), and lab studies.

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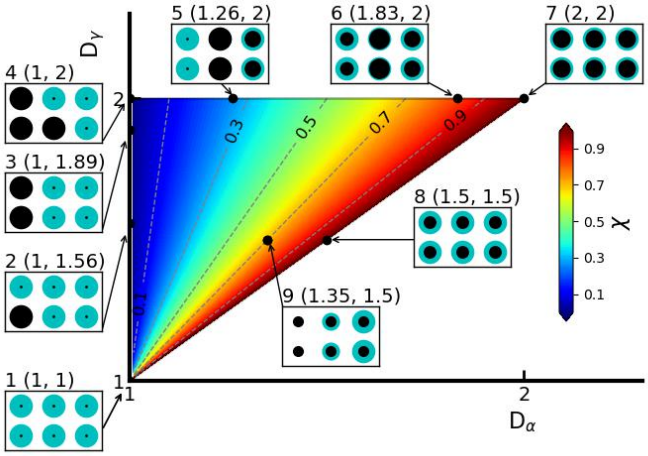

445

446 **Figure 2.** Mixing states diagram to illustrate the relationship between D_α, D_γ, and χ. Each species

447 consists of six particles, and the colors of black and cyan represent the BC and non-BC components.

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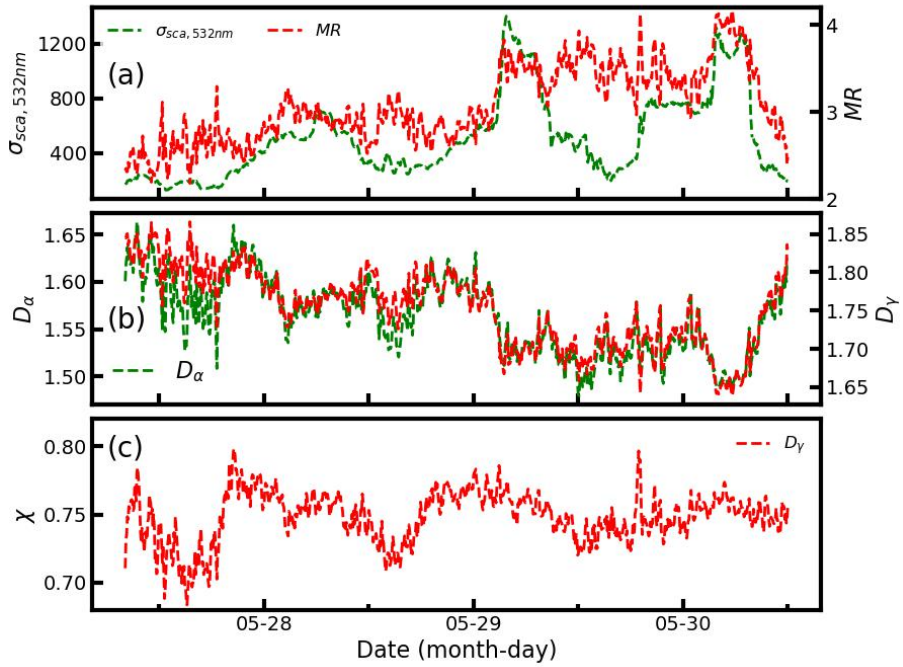

449

450 **Figure 3.** Measured time series of (a) σ_{sca} and MR, (b) D_α and D_γ, and (c) χ.

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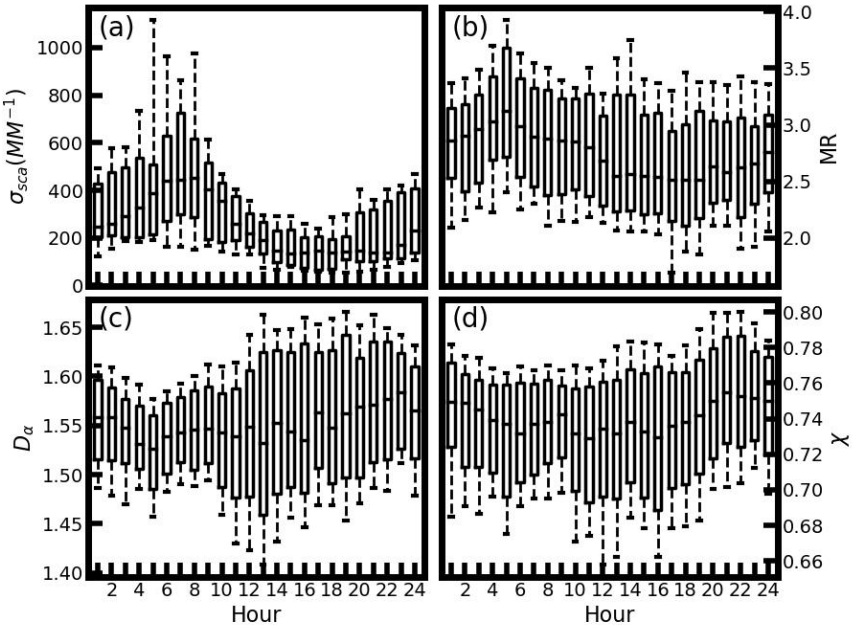

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453 **Figure 4.** Daily variation of the measured (a) σ_{sca}, (b) MR, (c) D_α, and (d) χ.

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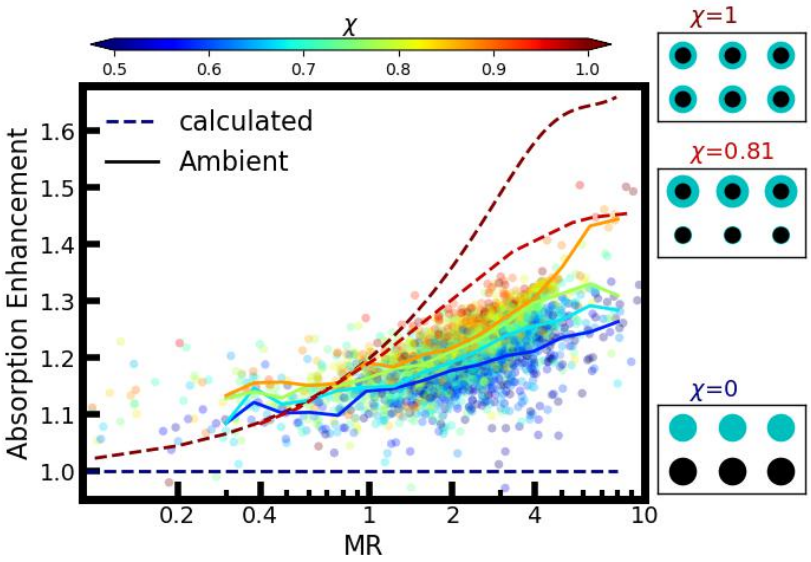
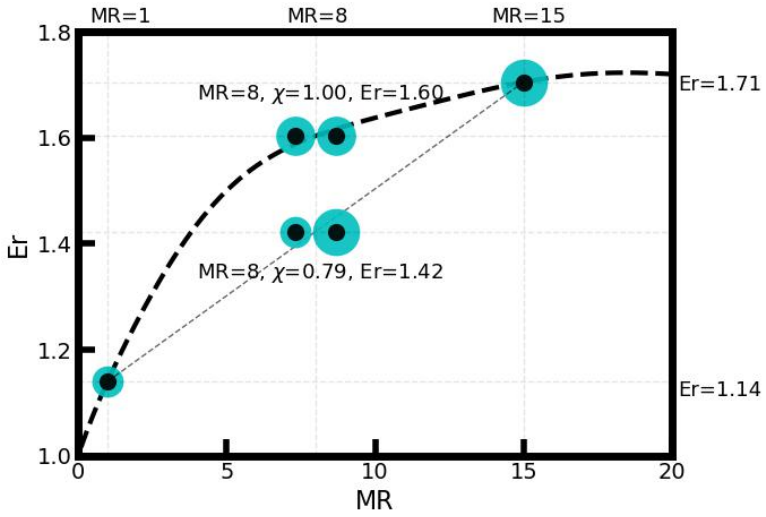


Figure 5. Relationship between the BC E_{abs} and the measured mass ratio of the BC-containing aerosols coating material to BC under different χ conditions. Four solid lines from bottom to up corresponding to the measured ambient size-resolved BC mixing states data with χ ranges of 0.575~0.625, 0.625~0.675, 0.675~0.725, and 0.725~0.775. The dotted line corresponds to the χ of 0.0 (blue), 0.81 (light red), and 1.0 (dark red), respectively.



463

464 **Figure 6.** Schematic diagram that denotes the relationship between χ and Er.

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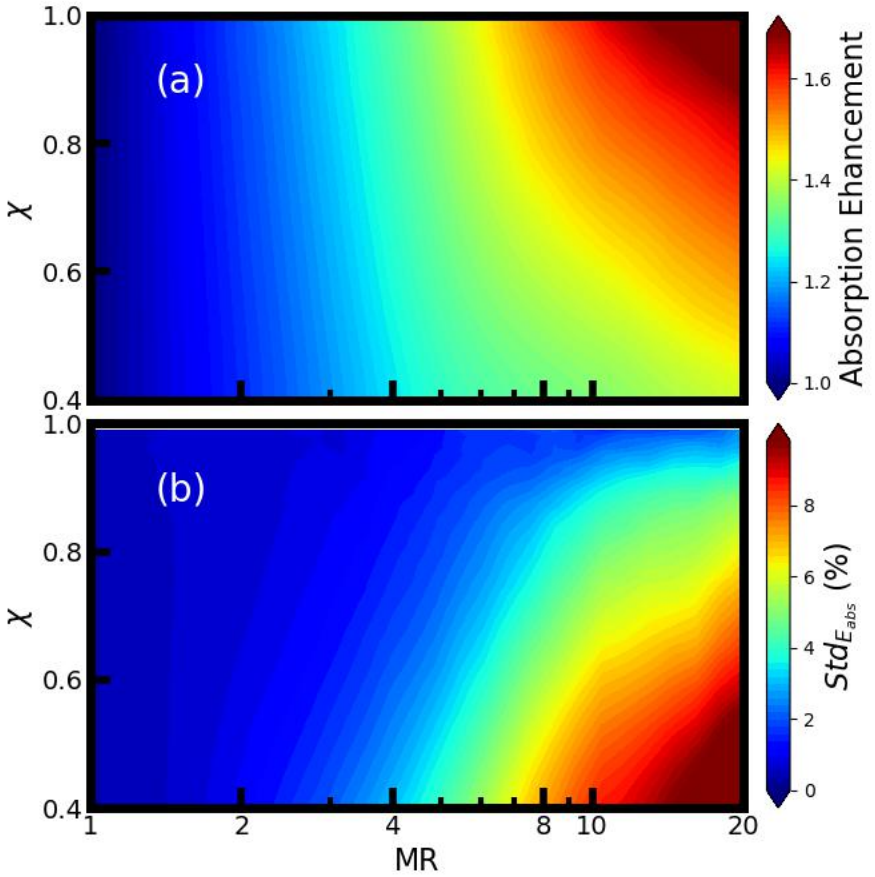

Figure 7. The calcaulted (a) mean E_{abs} values and (b) standard deviations of the E_{abs} values for different MR and χ.

469

ID	(D_α, D_γ)	χ	P1[*1]	P2[*1]	P3[*1]	P4[*1]	P5[*1]	P6[*1]	Tot[*1]
1	(1.00, 1.00)	-	(10^{-9},1)	(10^{-9},1)	(10^{-9},1)	(10^{-9},1)	(10^{-9},1)	(10^{-9},1)	($6{\cdot}10^{-9}$,1)
2	(1.00,1.56)	0	(1,10^{-9})	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(1, 5)
3	(1.00, 1.89)	0	(1,10^{-9})	(1,10^{-9})	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(2,4)
4	(1.00, 2.00)	0	(1,10^{-9})	(1,10^{-9})	(1,10^{-9})	(10^{-9}, 1)	(10^{-9}, 1)	(10^{-9}, 1)	(3,3)
5	(1.26, 2.00)	0.26	(2,10^{-9})	(2,10^{-9})	(10^{-9},2)	(10^{-9},2)	(10^{-9},1)	(1,1)	(6, 6)
6	(1.83, 2.00)	0.83	(1,3)	(1,3)	(3,1)	(3,1)	(2,2)	(2,2)	(12,12)
7	(2.00, 2.00)	1.00	(1,1)	(1,1)	(1,1)	(1,1)	(1,1)	(1,1)	(6,6)
8	(1.5, 1.50)	1.00	(1,6.1)	(1,6.1)	(1,6.1)	(1,6.1)	(1,6.1)	(1,6.1)	(6, 36.6)
9	(1.35, 1.50)	0.70	(1,10^{-9})	(1,10^{-9})	(1,6.1)	(1,6.1)	(1,12.2)	(1,12.2)	(6, 36.6)

470 **Table 1.** Detail information of the BC particles shown in Fig.2

471 [*1] Mass of the BC component of and non-BC component (arbitrary unit).