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Method to Quantify the Black Carbon Aerosol Light Absorption Enhancement with Entropy and Diversity Measures

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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 mass ratio of non-BC coating thickness to BC (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

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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 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, 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). Zhao et al. (2019b) reported that the light

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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 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, a nephelometer (Aurora 300,

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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 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 BC mixing states from the DMA-SP2 system can refer to Zhao et al. (2020a). The measured size-resolved BC mixing states 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 ($\frac{\mathrm{dN}}{\mathrm{dlogDp \cdot dlogDc}}$).

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+0.67*i*, 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+0*i*, 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.

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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 ($\frac{d\sigma_{abs}}{dlogDp \cdot dlogDc}$) can be calculated by multiplying the number concentrations of the BC-contained aerosols ($\frac{dN}{dlogDp \cdot dlogDc}$). By integrating the $\frac{d\sigma_{abs}}{dlogDp \cdot dlogDc}$ 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 $\sigma_{abs,Dp,Dc}$ 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 without thickness can be calculated by integrating the calculated $\sigma_{abs,\ Dc,Dc}$ among different Dp and Dc weighted with $\frac{dN}{dlogDp \cdot dlogDc}$. Thus the ambient BC particles without coating ($\sigma_{abs,Dp=0}$) can be calculated. The bulk ambient aerosol E_{abs} can thus be calculated with $E_{abs} = \frac{\sigma_{abs}}{\sigma_{abs,\ Dp=0}}$.

2.4 Quantifying the dispersion of BC mixing states

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.

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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:

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

the average mixing entropy of each particle 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

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

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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). Our 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 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

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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, which is consistent with the results in section 3.1 that the 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.

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

For each of the measured group of size-resolved 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. 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 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

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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 BC mixing states. However, there were no-study, to our best knowledge, that attempts to constrain the uncertainties.

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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.

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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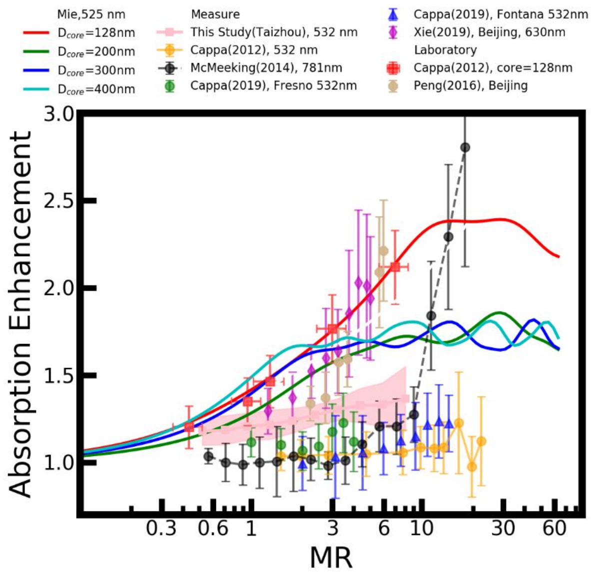

Figure 1. The measured E_{abs} of BC particles from different ambient measurements, including this work (in pink), and lab studies.

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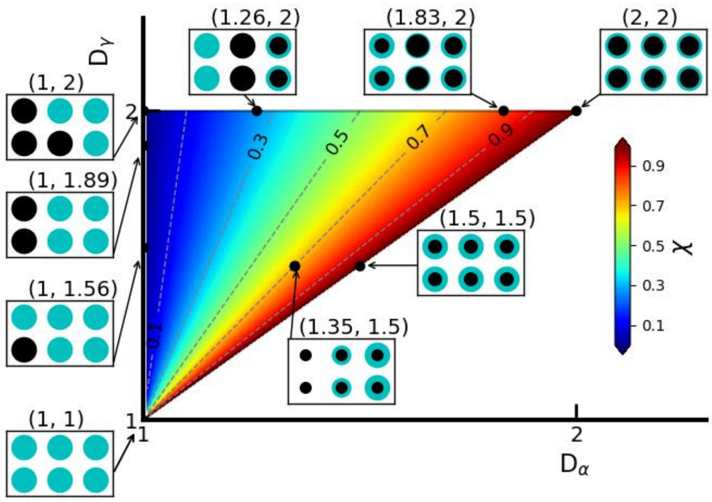

Figure 2. Mixing states diagram to illustrate the relationship between D_α, D_γ, and χ. Each species consists of six particles, and the colors of black and cyan represent the BC and non-BC components.

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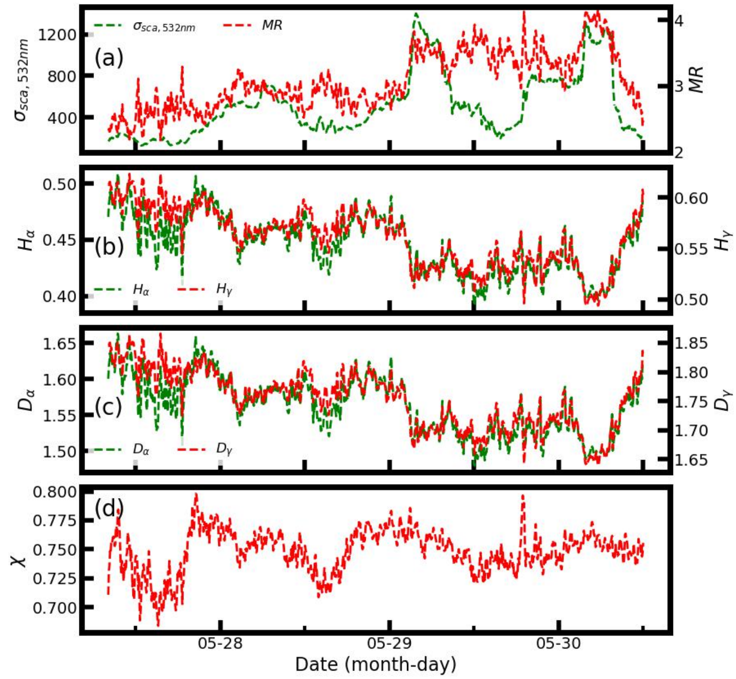

Figure 3. Measured time series of (a) σ_{sca} and MR, (b) H_α and H_γ, (c) D_α and D_γ, and (d) χ.

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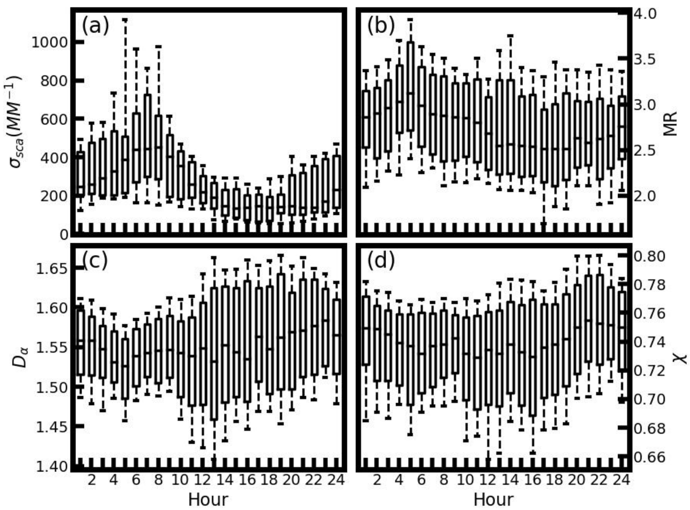

Figure 4. Daily variation of the measured (a) σ_{sca}, (b) MR, (c) D_α, and (d) χ.

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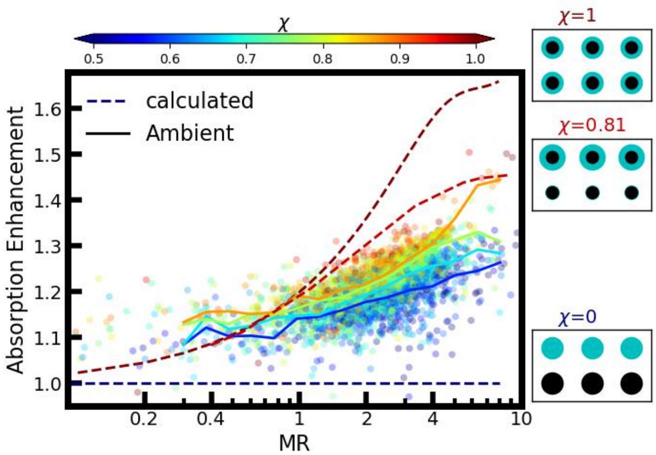

Figure 5. Relationship between the BC E_{abs} and the measured mass ratio of the BC-containing aerosols coating material to BC under different χ conditions. Four solid lines from bottom to up corresponding to the measured ambient size-resolved BC mixing states data with χ ranges of 0.575~0.625, 0.625~0.675, 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.

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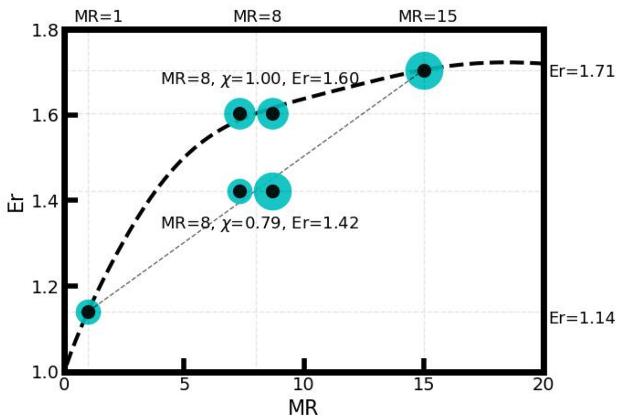

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416 **Figure 6.** Schematic diagram that denotes the relationship between χ and Er.

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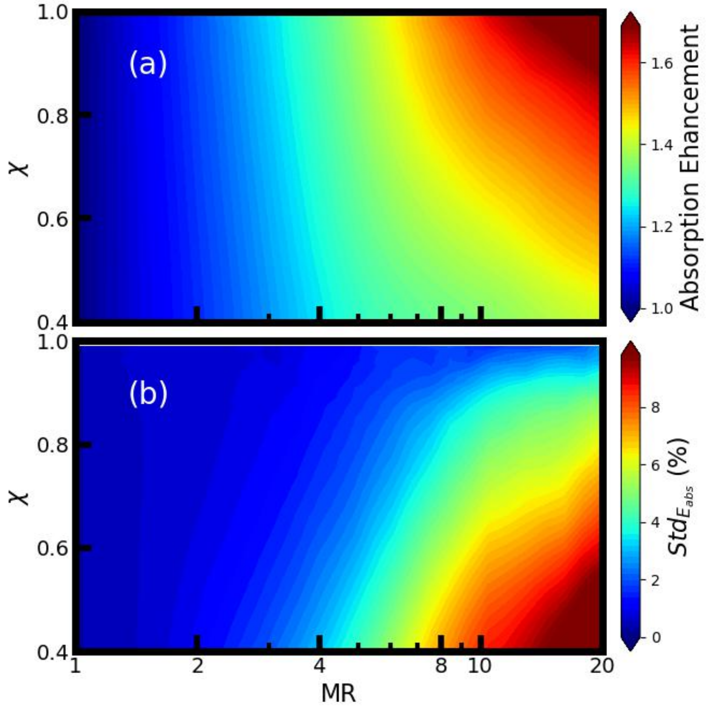

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419 **Figure 7.** The calcaulted (a) mean E_{abs} values and (b) standard deviations of the E_{abs} values for

420 different MR and χ.

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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)	-	(0, 1)	(0, 1)	(0, 1)	(0, 1)	(0, 1)	(0, 1)	(0, 6)
2	(1.00,1.56)	0	(1,0)	(0, 1)	(0, 1)	(0, 1)	(0, 1)	(0, 1)	(1, 5)
3	(1.00, 1.89)	0	(1,0)	(1,0)	(0, 1)	(0, 1)	(0, 1)	(0, 1)	(2,4)
4	(1.00, 2.00)	0	(1,0)	(1,0)	(1,0)	(0, 1)	(0, 1)	(0, 1)	(3,3)
5	(1.26, 2.00)	0.26	(2,0)	(2,0)	(0,2)	(0,2)	(1,1)	(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	(1,0)	(1,0)	(1,6.1)	(1,6.1)	(1,12.2)	(1,12.2)	(6, 36.6)

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

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