Light absorption of black carbon aerosols strongly influenced by particle morphology distribution

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Abstract

Atmospheric black carbon (BC) is the most important aerosol contributor to global warming. However, there is a lack of understanding about the climate impact of BC aerosols because of systematic discrepancies between model and observation estimates of light absorption enhancements (Eabs) in atmospheric processes after emissions, and such discrepancies are transferred directly into large uncertainties of aerosol radiative forcing assessments. In this study, we quantify Eabs of atmospheric BC aerosols with diverse particle morphology distributions using a multi-dimensional aerosol model. We show that current widely used Mie method may overestimate BC Eabs by ~50% because variations in particle morphology are not considered. Although absorption calculation can be improved by including complex particle morphology and heterogeneity in composition, we find that neglect of the diverse particle morphology distributions in modeling may lead to 15% ~ 30% relative deviations on Eabs estimations of BC aerosol ensembles. The results thus imply that particle morphology distribution should be included in models to accurately represent the radiative effects of BC aerosols.

1. Introduction

Light absorption by black carbon (BC) aerosol is one of the largest sources of uncertainty in assessments of radiative forcing. Several hypotheses have been forwarded to account for uncertainty of BC radiative forcing by climate models, including the amount, lifetime, and regional transmission of BC and brown carbon (BrC) aerosols, and the coating-enhancement of ambient BC absorption [1, 2]. Quantifying global warming in relation to BC aerosols depends explicitly on predicting the absorption of ambient BC when it mixes with weakly-absorbing components [3, 4]. Previous laboratory measurements and modeling studies have shown that BC particles coated by non-BC materials can significantly enhance absorption per unit mass through the so-called ‘lens effect’ [5–13], however, this absorption enhancement (Eabs) was observed to be negligible in some ambient conditions [14]. Such a discrepancy indicates that the characteristics of atmospheric BC-containing aerosols are not adequately represented by current models. The possible source of the controversy is that measured BC particles may not be fully coated with a non-BC material in the ambient, which thus leads to underestimates of BC Eabs [15, 16]. To our knowledge, treatments of BC microphysics, such as mass-, moment- and sectional-based schemes [17, 18], are not able to explain this phenomenon in major climate models, as they have typically adopted a simplified approach to represent mixed BC particles with the core–shell spherical structure.

Recent studies have indicated that Eabs of BC-containing particles can be constrained by considering their complex particle morphologies and mixing states. Results of in situ measurements and laboratory studies have indicated that bare BC particles are aggregates of small carbon spherules. When a BC particle in the form of a loose fractal aggregate shrinks to a compact aggregate, it may have a substantially different Eabs [19]. In the atmosphere, bare BC particles tend to be coated with other aerosol components...
through coagulation and condensation of secondary aerosol compounds. Previous observations have indicated that the value of $E_{abs}$ can be determined by BC volume fractions within the individual particles [8, 9, 20–24]. In addition, previous modeling descriptions have shown that more realistic morphologies and mixing states of BC-containing particles are in better agreement with observed absorption for the individual particles, which suggests the use of an empirical formula for correcting the core–shell Mie method in the climate modeling [25–27]. However, these attempts to improve BC absorption estimates by considering their complex particle morphologies have been limited to the particle-level, and explorations have not yet been conducted to determine absorption variations in the BC aerosol ensemble with respect to how BC microphysics influence global climate effects.

This paper presents a multi-dimensional model integrating more realistic particle morphologies and mixing states for absorption estimates of BC aerosol ensembles rather than a single particle. We show the effect of particle morphology distribution on the light absorption of a population of BC aerosols and provides an analysis of the relative deviations in modeled atmospheric BC absorptions caused by simplified microphysics. A multi-dimensional aerosol model is firstly presented that integrates full physical and chemical distributions of BC-containing aerosol ensembles by four typical dimensions, namely, particle morphologies, sizes, multi-components (mixing states) and fractal parameters, providing a feasible representation for the prediction of atmospheric BC aerosol absorption. Experimental studies [28, 29] suggest that the morphology of BC-containing particles can be quantified into four categories: bare, partly coated, partially encapsulated, and heavily coated morphologies, and the particle morphology distribution is then determined by their respective proportions ($F_{m}$) in aerosol ensembles. The morphology distribution is introduced to indicate the percentage proportions of specific particle morphologies that can be observed easily using microscopy. All possible morphology distributions (176 852 cases by the depth-first search algorithm in this study, see section S1) are investigated assuming a resolution of 1% and a sum of 100% for four specific particle morphologies, which provides a benchmark dataset for use in the optical simulation of atmospheric BC aerosol ensembles. For each type of particle morphology, the distributions of size, multi-components, and fractal parameters can then be further determined to provide a more detailed description of BC aerosol ensembles. For the morphologically simplified models, recent study indicated that the particle composition diversity of BC populations can be simulated by a particle-resolved model that ignores the complex particle morphology of fractal aggregated structures [30, 31]. Further, the size-resolved model includes the size distribution, whereas the particle morphology and mixing states of BC aerosols are highly simplified. In this respect, the relative deviations in modeled atmospheric BC absorptions caused by these simplifications of microphysics are analyzed below.

2. Materials and methods

2.1 Multi-dimensional aerosol model integrated by physical and chemical distributions

BC aerosols ensembles were simulated using the multi-dimensional aerosol model presented in this study, which integrates the important physical and chemical distributions of the aerosol ensembles for a specific time and area. The BC-containing particles are sorted for four typical morphologies, and the numbers of different morphologies are counted. For each morphology, the size distribution of BC-containing particles are constructed by volume-equivalent diameter of BC component. Further, the multi-component distributions are constructed for the BC-containing particles with fixed morphology and volume-equivalent diameter of BC, indicating the heterogeneity in composition of BC aerosols. At last, the fractal dimension distribution are constructed for the BC-containing particles with fixed morphology, volume-equivalent diameter of BC and proportions of mixed components, indicating the compactness of the particle morphology. Modeling the individual BC-containing particles is described in section S2.

The size distribution is constructed by the numbers of the BC-containing particles with a fixed volume-equivalent diameter of the pure BC component ($D_{BC}$). Previous measurements have suggested that the volume-equivalent diameter of pure BC particles has a lognormal distribution from 50 nm to 300 nm [32–35]. The standard deviations of lognormal distributions in this study are assumed to be 0.001–1, which considers the major atmospheric conditions. The number distributions ($N(D_{BC})$) of size are described as following:

$$N(D_{BC}) = \frac{1}{D_{BC}\sigma \sqrt{2\pi}} \exp\left(-\frac{(\ln D_{BC}-\ln D_{m})^2}{2\sigma^2}\right)$$

where $\sigma$ is the standard deviation and $D_{m}$ is the peak volume-equivalent diameter of the pure BC component in the BC aerosol ensembles. The number distributions of multi-component and fractal dimensions are similar for the simulations.

The mixing of BC and non-BC components is described by the multi-component distribution of BC aerosol ensembles. The BC volume fraction ($F_{BC}$) is applied to indicate the fraction of the particle volume that is BC in the individual particles. Without non-BC coatings, the BC volume fraction of bare BC particles equals 1 ($F_{BC} = 1$). Recent studies [36] indicated that BC absorption is affected by particlescale diversity in its composition. In this study, the
The BC-containing particles are further classified by the fractal parameters of their aggregated structures. In this study, the BC monomer radii (a) are assumed to be constant at 0.025 μm, as suggested by the review of Bond and Bergstrom [39], and the monomer number of the BC aggregate (Ns) is calculated by the volume-equivalent diameter of the pure BC component (D_{BC}). Previous measurements and simulations have suggested that the fractal dimensions (Df) of bare and aged BC particles vary from approximately 1.8 to 3.0, with a fixed fractal pre-factor (k) of 1.2 [11, 40]. The fractal parameter distribution is simulated by assuming the varied fractal dimensions between 1.8 and 3.0 and the varied standard deviations between 0.001 and 1, including major cases in the atmosphere. A greater amount of a non-BC coating may lead to a more compact BC structure and thus to the larger averaged fractal dimensions in a population of BC aerosols. Details of the modeling parameters are shown in table S2 (available online at stacks.iop.org/ERL/15/094051/mmedia).

The effect of particle morphology distribution of light absorption of BC aerosol ensembles is mainly investigated in this study, thus for the BC aerosol ensembles with a fixed morphology distribution, the other distributions are varied for a wide range of possible conditions. The standard deviations in major cases of BC aerosol populations lie in the range of 0.001 and 1, and especially, the fixed value of physical properties can be normally simulated by the case of standard deviation equals 0.001. The value of Eab’s is calculated by the ratio of BC aerosol ensemble absorption to those of all bare. The sensitivity of size, multi-component and fractal parameter distributions are simulated and shown in figures S2–S4, respectively.

2.2. Optical calculation of BC aerosol ensembles

Cross sections of absorption and scattering (C_{abs} and C_{sc}) were calculated for the individual BC-containing particles, and the cross sections of entire aerosol ensembles (⟨C_{abs}⟩ and ⟨C_{sc}⟩) were integrated by the multi-dimensional distributions.

\[ \langle C_{abs} \rangle = \int \int \int C_{abs}(D_f, F_c, D_{BC}, F_m) \cdot N(D_f) \cdot N(F_c) \times N(D_{BC}) \cdot N(F_m) \, dD_f \, dF_c \, dD_{BC} \, dF_m \]  

(2)

The corresponding mass cross-sections (MAC and MSC, respectively) were further normalized and are defined as cross-sections per unit mass of BC. To provide an example of absorption, the normalization of an absorption cross section is divided by the BC mass as follows,

\[ \langle MAC \rangle = \frac{\langle C_{abs} \rangle}{\frac{4}{3} \pi (R_{BC})^3 \rho_{BC}} \]  

(3)

where ⟨R_{BC}⟩ and ρ_{BC} are the volume-equivalent BC radii integrated by the entire aerosol ensembles and the mass density of BC, respectively. The mass density of BC (ρ_{BC}) is assumed to be 1.8 g cm⁻³ according to a review of measurement by Bond and Bergstrom [39]. As shown in figure S5, the sensitivity of complex particle morphologies on BC optical properties at 0.532 μm and 1.064 μm is investigated.

2.3. Morphologically simplified models using the core–shell Mie method

The radiative properties of BC aerosols in climate models are commonly obtained based on the morphological simplification of homogenous sphere and single core–shell sphere. Their optical properties are generally calculated using the Lorenz–Mie–Debye theory and Mie Core–Shell model, when the morphology and fractal parameter distributions are not considered. However, large discrepancies have been measured and simulated between the aggregates and the equivalent sphere approximations due to their complex morphologies, mixing states, components and multiple scattering [22, 23, 41–43].

The volume-equivalent radius of BC particle (R_{BC}) is related to its aggregated morphology, according to the following equation,

\[ R_{BC} = \frac{1}{2} D_{BC} = \sqrt{N \cdot a} \]  

(4)

The volume-equivalent thickness of the non-BC shell (T_{non-BC}) of a single core–shell sphere is

\[ T_{non-BC} = \sqrt{\frac{N_c}{F_{BC}}} a - R_{BC} \]  

(5)

and the thickness of non-BC (T_{non-BC}) is zero for bare BC particles.

3. Results

Figure 1 shows how a complex BC population simulated by the multi-dimensional aerosol model, and shows that current models provide simplified simulations when one or more physical and chemical distributions of BC aerosol ensembles are not...
considered. Variations in the particle morphology distribution are shown in figure 1(b), each percentage proportion of the four morphologies (figure 1(a)) ranges from 0% to 100%, and their sum is 100% in the instantaneous atmosphere. The particle morphologies of atmospheric BC aerosols are complex, and they depend highly on the degree of aging, the ambient temperature and relative humidity. For example, previous measurements and simulations have suggested that the ratio of bare BC in aerosol ensembles tends to decrease after atmospheric aging, while the ratio of heavily aged BC conversely increases [44]. Some recent aggregate models have considered the complex particle morphologies of BC aerosols, however, the individual BC particles on different aging scales have been simulated rather than the actual particle populations [25–27]. The particle morphology play important role in optical calculations of BC particles, but the particle morphology variations in BC aerosol ensembles are not considered in current aggregate models. It is suggested that the particle morphology distribution is required to enable an exact description of BC aerosol ensembles, due to their significant differences in ambient environments.

Figure 2 shows that the light absorptions of BC aerosol ensembles are significantly influenced by their morphology distributions and suggests a negative correlation between the ratio of bare BC and total BC-containing aerosols ($F_{\text{bare}}$). Absorption calculations for the individual BC-containing particles with fractal aggregated structures were conducted using the superposition T-matrix method. The absorption cross-sections of the entire BC populations were integrated with those of individual particles with the multi-dimensional distributions, and their MAC values were further divided by the BC mass of the aerosol ensembles. Light absorptions of the BC aerosol ensembles with a fixed morphology distribution vary with different realizations due to the wide range of the other distributions when considering possible extreme conditions. As shown in figure 2(a), the averaged MAC values are sorted and range from $\sim 6.5 \text{ m}^2 \text{ g}^{-1}$ to $\sim 13.5 \text{ m}^2 \text{ g}^{-1}$, and the corresponding morphology distributions are shown in figures 2(b)–(e), including the percentage ratios of four typical particle morphologies. Observed MAC results of previous ambient and laboratory measurements support the range of these simulations [24, 33, 39, 45, 46]. When the aerosol ensembles consist entirely of bare BC particles, their simulated averaged MAC values are the lowest, and when aerosol ensembles are full of heavily coated BC, the averaged MAC values become the highest. BC particles freshly emitted from sources may have a bare morphology and they then become heavily coated over a period between hours to one week, in accordance with atmospheric aging. However, it is unrealistic to assume that the atmosphere is filled with only bare BC particles or the BC particles fully coated by non-BC components at a fixed coating scale. On the one hand, bare BC particles are being continuously and freshly emitted to the atmosphere, leading to coexist of BC particles with different morphologies; on the other hand, plenty of BC-containing particles are generated to be the partially encapsulated and partly coated morphologies during aging, which is not properly considered in current modeling. Therefore, understanding the proportions of various particle morphologies would provide a more reliable model representation of the real atmosphere.

The Eabs of entire BC aerosol ensembles is the mass absorption cross-sections of populated BC particles with fixed morphology distributions divided by those of bare BC particles. As shown in figure 2(a), the MAC results of BC aerosol ensembles are averaged by the possible cases with the fixed morphology distribution (yellow lines) and are affected by the distributions of size, multi-component and fractal dimension (gray region). Optical simulations of the fully aged BC aerosol ensembles dominated by heavily coated morphologies show that the lowest MAC results are $\sim 8 \text{ m}^2 \text{ g}^{-1}$ for certain extreme BC aerosol ensemble cases. The absorption enhancement (Eabs) is $\sim 1.25$, while the corresponding MAC values are calculated to be $\sim 6.3 \text{ m}^2 \text{ g}^{-1}$ for the aerosol ensembles with all bare BC. If the MAC of bare BC is assumed to be the widely used value ($7.5 \pm 1.2 \text{ m}^2 \text{ g}^{-1}$) suggested by Bond and Bergstrom [39], the possible Eabs is $\sim 1.05$. These possible values show agreement with the Eabs value ($\sim 1.06$) by previous observations in the ambient conditions [14]. The possible reasons for this are the participation of BC particles with partially encapsulated and partly coated morphologies [15], the non-BC coating consists of components with smaller real parts of refractive indices [47, 48], and the BC volume fractions of the entire aerosol ensembles are relative larger (e.g. $\sim 0.1$). In previous field measurements, the median value of the BC volume fraction of coated particles is measured to be $\sim 0.15$ with aerodynamic diameters ranging from 0.05 $\mu\text{m}$ to 0.3 $\mu\text{m}$ [49], indicating the possible situations for the small Eabs values. Therefore, it is necessary to describe the full physical and chemical properties of BC aerosol ensembles during optical modeling.

As shown in figures 2(f)–(g), the Eabs results of the simulations agree with the previous observations. The percentage ratios of typical BC-containing particle morphologies of two samples (figure 2(f)) and their Eabs values (figure 2(g)) were measured by Liu et al [30]. According to the observed morphology distributions of thermodenuded (T) and ambient (A) samples, the results of Eabs are simulated to be $1.54 \pm 0.35$ and $1.75 \pm 0.49$, respectively, and the measured Eabs of $1.67 \pm 0.42$ lie in the range of these simulations. The observed morphologies reappear in the modeling, although the thinly coated BC-containing particles observed in these measurements are assumed to be bare BC in the simulations, due
Figure 1. Schematic diagram of the multi-dimensional model for black carbon aerosols ensembles. The physical and chemical properties of BC-containing particles in the atmosphere are reconstructed using the multi-dimensional distributions: proportions of (a) four morphologies in (b) morphology distribution, (c) size distribution, (d) multi-component distribution and (e) fractal parameter distribution. The morphology distribution is applied first to classify BC particles and counts the proportions of the bare (blue), partly coated (yellow), partially encapsulated (pink), and heavily coated (green) morphologies. The red box in (b) shows an example of BC aerosol ensembles with a fixed morphology distribution (including 20% bare, 20% partly coated, 30% partially encapsulated, and 30% heavily coated particles). For each morphology, the size distribution of BC-containing particles are constructed by volume-equivalent diameter of BC in (c), further, the multi-component distributions are constructed for the BC-containing particles with fixed morphology and volume-equivalent diameter of BC in (d), and at last, the fractal dimension distribution are constructed for the BC-containing particles with fixed morphology, volume-equivalent diameter of BC and proportion of mixed components in (e). The lines and areas with the same colors in different sub-figures are not related.

The multi-dimensional aerosol model presented in this study may simulate BC absorption more realistically with four distributions, namely, particle morphology, fractal parameter, multi-component, and size. Three simplified representations were investigated using the same physical and chemical distributions of BC aerosol ensembles, employing: (1) an aggregate model that considers the aggregated structure of BC, multi-components, and volume-equivalent diameters but does not include the morphology distributions of populations, including the distributions of fractal parameter, multi-component and size; (2) a particle-resolved model that considers the size of BC and multi-components that can simulate the diversity in composition but does not treat complex morphologies by assuming single core–shell spherical structures, including the distributions of multi-component and size; (3) a size-resolved model that only varies in the volume-equivalent diameters of the BC particles and assumes that a single sphere...
Figure 2. Absorption of BC aerosol ensembles depending on the particle morphology distribution. (a), sorted mass absorption cross-sections (MAC) and corresponding percentage ratios of particle morphology distributions for bare (b), partly coated (c), partially encapsulated (d), and heavily coated (e) morphologies. The MAC results are averaged by the possible cases with the fixed morphology distribution (yellow lines in (a)) and are affected by the distributions of size, multi-component and fractal dimension (gray region in (a)). Model verification: (f), percentage ratios of particle morphologies are observed for the thermodenuded (T) and ambient (A) samples [30]. (g), observed Eabs and simulations of these two samples are compared.

represents individual particles, including size distributions of BC aerosol ensembles.

Figure 3(a) shows the relative deviations in light Eabs of BC aerosol ensembles between the multi-dimensional aerosol model and the particle-resolved model, where the ratio of bare BC to total BC aerosols ($F_{\text{bare}}$) is varied. In BC populations, $F_{\text{bare}}$ is the number of BC particles that are not mixed with other aerosol components divided by the total number of the BC-containing particles. This parameter, which exhibits a negative relationship with the MAC values of BC ensembles, can be feasibly estimated by the ambient observations. As shown in figure 3(a), the particle-resolved model may overestimate the magnitudes of Eabs for populated BC particles by \(\sim 50\%\) when $F_{\text{bare}}$ is larger than 0.6. In some extreme cases, the relative errors reach \(\sim 200\%\) due to the morphological simplifications. When major BC particles mixing with non-BC components they cause fewer relative deviations in the values of Eabs, which reaches \(\sim 20\%\) without bare BC particles in the aerosol ensembles. One possible reason for this is that the effect of complex morphology on the absorption of BC particles may be weakened by the larger volume of non-BC coatings in the individual BC-containing particles. Previous studies of the particle-level simulations have indicated that the relative deviation of light absorption between the aggregate and morphologically simplified models is \(\sim 30\%\) [50]. The quantitative results of the population-level simulations shown here demonstrate that the relative deviations of averaged Eabs values may vary with the ratio of bare BC in the morphology distributions.
Figure 3. Comparison of absorption enhancement (Eabs) between different modeling methods. (a) Variations in Eabs dependent on ratio of bare BC to total BC aerosols \( (F_{\text{bare}}) \) estimated using the multi-dimensional model and the particle-resolved model. Yellow line represents the averaged Eabs results of the possible cases with the fixed \( F_{\text{bare}} \) using the multi-dimensional model and affected by varied distributions of morphologies (constant bare, but varied coating states of BC-containing particles), size, multi-components, and fractal parameters, in yellow region. The green line and green region are those using the particle-resolved model. (b) Relative deviations in light Eabs between models considering different numbers of physical and chemical distributions of BC aerosol ensembles. The multi-dimensional aerosol model presented in this study considers the morphologies, fractal parameters, multi-components, and size distributions of BC populations, whereas these distributions are variously simplified by the other models, namely, the aggregate model, particle-resolved model, and size-resolved model.

Figure 3(b) depicts the relative deviations in Eabs of BC aerosol ensembles between the multi-dimensional aerosol model and the other simplified models. The morphology distributions of BC aerosol ensembles are not considered in the aggregate model, and the particles of BC mixing with non-BC components are assumed to be partly coated, partially encapsulated, heavily coated, and to have varied morphologies that are dependent on the BC volume fraction, respectively (see figure S1). The averaged relative deviation of Eabs between the multi-dimensional aerosol model and the aggregate models is \( \sim 25\% \), which shows the important role of morphology distribution when modeling representations of atmospheric BC populations and conducting absorption simulations. The multi-components that depend on BC volume fractions are included in the particle-resolved model, and when the complex particle morphology of BC-containing particles is ignored, relative deviations in Eabs values of \( \sim 45\% \) may occur. Only size distributions are included in the size-resolved model, which assumes a homogeneous
4. Discussion and conclusions

Light absorption of atmospheric BC aerosol ensembles is significantly influenced by the diversity of particle morphologies, and this is generally neglected in current climate modeling \[51, 52\]. The distribution of particle morphologies of BC populations offers a more complete description of their physical and chemical properties and provides a potential solution to solving the conflicting BC Eabs results. In the simulated results, the low observed Eabs values correspond to some certain typical BC aerosol ensemble cases. For example, the small Eabs values would be reproduced when major individual BC particles are not fully coated and exist with partially encapsulated and partly coated morphologies, because the absorption enhancement of heavily coated morphologies is significantly larger than those caused by the two morphologies. Moreover, the averaged BC volume fractions of the entire aerosol ensembles may be relatively large in the ambient, leading to the situations that non-BC coatings are not large enough in the individual BC-containing particles. In previous field measurements, the median value of the BC volume fraction of coated particles is measured to be \(\sim 0.15\) with aerodynamic diameters ranging from 0.05 \(\mu\)m to 0.3 \(\mu\)m \[49\], indicating the possible situations of ambient atmosphere. The simulations suggested that the small Eabs values may be performed by the cases with relative larger (e.g. \(\sim 0.1\)) BC volume fractions of the entire aerosol ensembles. Furthermore, if the non-BC coatings have smaller real parts of refractive indices, the lens effect of BC aerosols may be dramatically limited in these situations and result in the relatively small Eabs for BC aerosol ensembles. The simulations indicated that the effect of real parts of refractive indices on BC absorption enhancement is also obvious in figure S6. These possible reasons can be effectively described by the distributions of particle morphology and multi-components. Therefore, it is suggested that particle morphologies and mixing states should be observed synchronously with field experiments of BC absorption.

Our datasets indicate that current widely used Mie calculations may overestimate the Eabs of BC aerosols by \(\sim 50\%\), and these relative deviations vary with respect to the ratio of bare BC to total BC aerosols. In the cases of satellite-based observations of aerosol-optical properties, Andersson and Kahnert (2016) suggested that the simplifications of BC particle morphology in the optics model can introduce substantial errors (30% \(\sim 50\%\)) in assessment of BC radiative forcing and inversion of remote sensing \[53\]. BC particles are mainly emitted from the incomplete combustion of fossil fuels and biomass burning, they tend to become coated with non-BC particles during atmospheric processes. In the widely used Mie method, freshly emitted and aged BC particles are assumed to be single sphere and core–shell sphere, respectively. Although the coating state of BC-containing particles in many models is dynamic, the assumption of BC aerosol ensemble by an averaged parameter of coating state (e.g. core–shell ratio, or mass ratio) may be unrealistic in the ambient environment, because there are continual emissions of carbonaceous aerosols \[54\]. Recently, Fierce et al (2020) provided a modeling framework that reproduces measured absorption enhancements by BC-containing particles across field and laboratory measurements, suggesting that neglect of heterogeneity in particle composition is the dominant cause of previously reported discrepancies between models and observations \[36\]. It is suggested that BC absorption estimation can be improved by including the multi-component distributions in addition to size distributions. The presented multi-dimensional aerosol model is a model further considering complex particle morphologies for absorption estimation of BC aerosol ensemble rather than the single particles. In this study, \(F_{\text{bare}}\) can be applied to indicate the entire mixing states and complex particle morphologies of aerosol ensembles, which provides a strongly negative relationship with MAC values of BC populations. BC emissions and atmospheric process may be dramatically changed in different seasons and regions, leading to some remarkable variations in \(F_{\text{bare}}\) and corresponding light absorption. It is thus necessary to reassess the positive radiative forcing contribution from BC aerosols by considering accurate representations of real atmospheric BC aerosols using particle morphology distributions.

The multi-dimensional aerosol model may also provide a relatively complete description on regional microphysical processes. The relative difference of BC absorption between Mie calculations and the more realistic models considering particle morphology is remarkable in some cases due to the loose BC freshly emitted from diesel \[41\]. However, in some remote marine environments, the compact BC aggregates with large fractal dimensions were dominated. The corresponding numerical simulations indicated that the Mie calculations may provide a reasonable estimate (within 12%) of the radiative forcing for highly compact BC particles \[55\]. Therefore, more microscopy studies are acquired for different cases of BC emissions in the future, and the further constraint between the morphology distributions of BC aerosol ensembles and the regional atmospheric process would be helpful for the estimations of BC radiative forcing.
Author contributions

Y. Wu and T. Cheng designed the research; Y. Wu, and L. Zheng performed model simulations, data analysis and validation, Y. Wu and T. Cheng wrote the paper.

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Conflict of interest

The authors declare that they have no competing financial interests.

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