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Radiative properties of coated black carbon aerosols impacted by their microphysics

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ABSTRACT

Black carbon (BC) aerosols have substantial effects on the climate, whereas there still are significant uncertainties on coated BC radiative properties. Numerical investigations on radiative properties of polydisperse BC aggregates partially coated by non-absorbing organics, which are calculated by the exact multiple-sphere T-matrix method, are presented. The objective of our study is to evaluate the impacts of particle microphysics, including BC coating volume fraction, shell/core ratio, BC morphology, and size distribution, on partially coated BC radiative properties, as well as their absorption and scattering enhancements. Particle size distribution and shell/core ratio affect radiative properties of partially coated BC significantly, while BC coating volume fraction is an insensitive parameter. The absorption enhancement of BC particles due to partially coated by organics varies from ~1.0 to ~2.0, whereas their scattering enhancement can reach up to ~100. The BC coating volume fraction seems to be responsible for BC absorption enhancement, and with BC coating volume fraction becoming larger, the absorption enhancement becomes more sensitive to particle size distribution and shell/core ratio. However, the dependence of BC scattering enhancement on shell/core ratio is larger than size distribution and BC coating volume fraction, and the dependences of scattering enhancement to size distribution and BC coating volume fraction becomes stronger as shell/core ratio becomes larger. Our study gives a further understanding of the influences of particle microphysics on partially coated BC radiative properties, and it may be helpful for model and parameter simplifications.

1. Introduction

Black carbon (BC) aerosols, originating from incomplete fossil fuel combustion and biomass burning, have profound effects on regional and global climate [1–3]. As the second most important contributor to global warming after CO_2, BC contributes significant positive climate radiative forcing [4]. Currently, our understanding of BC radiative properties, which are determined by their complicated microphysics, such as BC morphology and mixing state, is still limited, resulting in one of the largest uncertainties in assessing aerosol radiative forcing [5].

Freshly emitted BC aerosols are in general hydrophobic and externally mixed with other particle components [6,7]. During the aging process, BC tends to be coated by water-soluble constituents, including heterogeneous reactions with gaseous oxidants [8,9], coagulation with preexisting particulates [10,11], and condensation of sulfate, nitrate and organics [12,13]. In addition to mixing state, BC morphology undergoes considerable restructuring, and becomes compact [7,14]. With significant variations of particle microphysics during the aging, the radiative properties of BC aerosols can alter dramatically, for instance, BC absorption can be enhanced observably due to coating [e.g., 15,16]. In aerosol-climate models, the concentric spherical core-shell model is widely used to obtain the radiative properties of aged BC particles based on the core-shell Mie theory [17]. Nevertheless, this simplified model may introduce significant errors, and its effectiveness still need to be validated on

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the basis of the comparison of its radiative properties with those of more realistic ones.

The understanding of aged BC radiative properties can be carried out based on numerical investigation, and observed irregular BC coating morphologies in ambient atmosphere can generally be classified into three types: fully coated, partially coated, and externally attached [e.g., 18–20]. Many researches have been performed to study radiative properties of BC or fully coated BC particles considering the effects of coating morphology and mixing state. The radiative properties of coated BC with complex morphologies can be computed utilizing the discrete dipole approximation (DDA) method, which is flexible in dealing with particles with arbitrary geometry and compositions [21,22]. With DDA, Kahnert et al. study the radiative properties of BC aggregates encapsulated into sulfate coating [23] and validate a core-grey shell model with a more realistic particle model [24]. Adachi et al. [25] apply DDA to simulate radiative properties of BC embedded within host particles, and find that the shapes and positions of BC inside its host particles have an important effect on the radiative properties. Scarnato et al. [26] also examine the effects of internal mixing and aggregate morphology on BC radiative properties using DDA. Dong et al. [27] introduce irregular sulfate coating to fractal BC aggregates and show the morphological effects on the radiative properties of BC particles. Kahnert presents radiative properties, including backscattering depolarization ratio, of BC fully coated by sulfate based on comparison of closed cell model with coated aggregate model [28]. Those simulations do build particle models quite close to realistic aerosols, whereas the numerical calculations for the radiative properties are neither simple nor efficient. Meanwhile, Cheng et al. [29] present the effects of morphology on the radiative properties of coated BC aggregates at different aging status based on the efficient multiple-sphere T-matrix method (MSTM). Based on the MSTM, Liu et al. investigate the spherical coating on radiative properties of BC aggregates [30]. These studies generally show the results of monodisperse coated BC particles and indicate that the radiative properties of coated BC are substantially influenced by their coating microphysics.

Although above significant numerical investigations have been performed to study radiative properties of BC or fully coated BC particles, generally limited research focuses on the effects of particle microphysics on partially coated BC radiative properties. Ishimoto et al. propose a shape model of coated BC particles, including partially coated BC, derived from artificial surface tension, and create a dataset of their radiative properties [31]. Kangieβer and Kahnert develop a model with tunable transition from film-coating to spherical-shell coating to investigate the impact of this transition on the linear depolarization ratio [32]. Meanwhile, more and more observations relevant to the microphysics of coated BC particles are carried out recently. Based on the measurements of BC spherical equivalent core diameter (D_c) and coating thickness with a single-particle soot photometer (SP2), the shell/core ratio D_p/D_c (particle diameter divided by BC core diameter) values of 1.1–2.1 and 2.1–2.7 are observed in London [33] and Beijing [34], respectively. On the basis of the observations of BC particles emitted from biomass burning, China et al. [19] quantify that ~50% are heavily coated (embedded), ~34% are partially coated, ~12% have inclusions and only ~4% are bare. The partially coated morphologies are found to be dominated for aged BC particles at a remote marine free troposphere site in Portugal [18]. These mixing-state observation of coated BC indicates that partially coated morphologies are important for coated BC in ambient air, whereas the discussions of radiative properties of partially coated BC with numerical investigations are not enough. As more recent observations are shown, it becomes urgent to do a more reliable simulation of the radiative properties of partially coated BC particles impacted by their microphysics. In addition, from application insights, it is difficult to reconstruct BC radiative properties with all detailed microphysical parameters with a simplified model, and one of the reasons is due to high computational cost. Nonetheless, it is possible to find a simplified model with reduced parameters requiring detailed variable values for a specified problem after knowing the effects of the microphysics. Although significant numerical investigations have been performed to study optical properties of BC or fully coated BC particles, the researches focusing on the effects of particle microphysics on partially coated BC radiative properties are generally limited, which significantly limits our ability to understand the model-observation discrepancies and to carry out accurate radiative transfer calculations.

Here, numerical investigations of radiative properties of polydisperse BC aggregates partially coated by organics are systematically presented. An exact MSTM is employed to numerically calculate the radiative properties of partially coated BC aggregates with various coating microphysics. The aim is to evaluate the effects of particle microphysics, including BC coating volume fraction, shell/core ratio, BC morphology, and size distribution, on radiative properties of partially coated BC particles, which hopefully improve our refining estimates of aerosol radiative forcing. The scattering and absorption enhancements of BC aerosols due to partial coating of organics are also investigated.

2. Methodology

2.1. Models of BC particles

It is observed that fresh BC particles look like loose cluster-like aggregates with numerous similar-sized spherical monomers [e.g., 35], and the fractal aggregates are successfully applied to construct these BC geometries based on the well-known statistical scaling law [e.g., 36]:

$$N = \mathrm{k}_0\left(\frac{R_g}{a}\right)^{D_f}, \tag{1}$$

$$R_g = \sqrt{\frac{1}{N}\sum_{i=1}^{N} r_i^2}, \tag{2}$$

where the monomer number of an aggregate (N), monomer radius (a), fractal prefactor (k_0) and fractal dimension (D_f) describe the morphology of a BC aggregate. R_g is the gyration radius, measuring the overall spatial size of an aggregate, and r_i indicates the distance between the ith monomer and the mass center of the aggregate. This study keeps employing the fractal aggregates to model homogeneous BC particles, and the k_f of BC is assumed to be 1.2 [36]. Two D_f values of 1.8 and 2.8, representing lacy and compact BC aggregates [29,37], are chosen for comparison. The radius a of BC aggregate monomers can alter over a range of 10–25 nm [38], whereas the monomer number N is observed to vary up to approximately 800 [39]. We choose N of 200 as examples for accumulation particles, as BC is observed to be mostly in the accumulation mode, following Zhang et al. [15]. With given fractal parameters, the BC aggregates can be generated based on a tunable particle-cluster aggregation algorithm from Skorupski et al. [40].

While the fractal aggregates are widely employed to model homogeneous BC particles, the models for simulating inhomogeneous aged BC are relatively complicated. This study considers the BC aggregates coated with organics, and the organics are treated as homogenous spheres that are coated to the BC fractal aggregates for the sake of simplicity. Some observations of individual aged BC particles actually do show this spherical coating geometry [e.g., 7,37,41], and the simple spherical coatings on BC particles have similar effects on radiative properties to those with more complex coating structures [27,42]. To describe the coating state of BC-

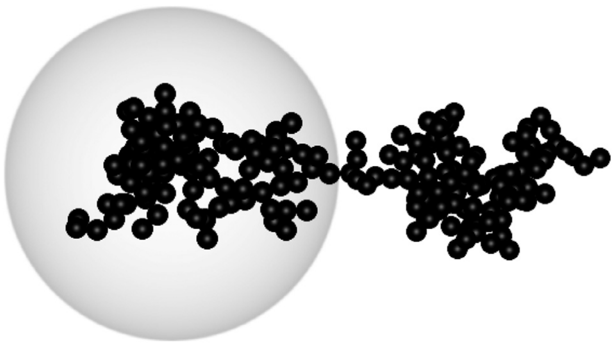

Fig. 1. Sketch map of partially coated black carbon geometry. An example of fractal black carbon aggregates, containing 200 monomers, is partially coated by non-absorbing organics with coated volume fraction of black carbon being 0.5.

organics internal mixtures, the BC coating volume fraction (F) is defined following the equation below:

$$F = \frac{V_{BC\,inside}}{V_{BC}}, \tag{3}$$

where $V_{BCinside}$ is the volume of BC monomers inside organics, and V_{BC} is the volume of the overall BC aggregate (i.e., the volume of BC monomers both inside and outside the coating). Obviously F measures how much BC is enclosed in the coating organics. The externally attached and fully coated BC aggregates typically have $F = 0$ and $F = 1$, respectively, while the partially coated BC particles show a BC coating volume fraction of $0 < F < 1$.

To build this inhomogeneous internal-mixed particle, we first generate a BC fractal aggregate and a spherical coating of organics with given parameters. Then, the coating sphere of organics is placed to coat the BC aggregate from one side to the other until an expected F is obtained. To avoid overlapped spheres after being coated, some BC aggregate monomers are slightly moved, whereas their aggregate geometry is still kept. It should be noted that these BC monomer movements to avoid overlapping have caused a slight D_f alteration of the BC aggregate, and the sketch map of partially coated BC aggregate is shown in Fig. 1.

2.2. Numerical simulation of aged BC radiative properties

Given that the model of coated BC aggregates is constructed with a spherical coating and no overlapped spheres, this study takes the advantage of the multiple-sphere T-matrix method [43,44]. The MSTM, which is in the framework of the T-matrix method, is employed to exactly calculate the random-orientation radiative properties of BC aggregates coated by organics in this study. This robust MSTM has already been popularly utilized for numerous numerical investigations of optical properties of fractal aggregates [e.g., 45,46].

For ambient atmospheric applications, bulk aerosol radiative properties are meaningful, which are averaged over a certain particle size distribution. This study considers an ensemble of BC aggregates with different sizes but the same coating fraction of organics, and a lognormal size distribution is assumed with the form following:

$$\mathrm{n}(r) = \frac{1}{\sqrt{2\pi}\, r \ln(\sigma_g)} \exp\left[-\left(\frac{\ln(r) - \ln(r_g)}{\sqrt{2}\ln(\sigma_g)}\right)^2\right], \tag{4}$$

where σ_g is the geometric standard deviation, and r_g is the geometric mean radius [e.g., 21,47]. Coated BC aggregates follow this lognormal size distribution, and r is the radius of a sphere with the same volume as that of the whole partially coated BC aggregate (i.e., equivalent volume sphere). We only consider accumulation coated BC aerosols, since BC is observed to be mostly in the accumulation mode. In this study, r_g is assumed to be 0.075 µm [48], while σ_g of 1.59 is considered [49]. With a particle size distribution given, the bulk radiative properties of coated BC can be calculated following the equations of:

$$\langle C_{sca} \rangle = \int_{r_{\min}}^{r_{\max}} C_{sca}(r) n(r) d(r) \tag{5}$$

$$\langle C_{abs} \rangle = \int_{r_{\min}}^{r_{\max}} C_{abs}(r) n(r) d(r) \tag{6}$$

$$\langle SSA \rangle = \frac{\langle C_{sca} \rangle}{\langle C_{ext} \rangle} \tag{7}$$

and

$$\langle g \rangle = \frac{\int_{r_{\min}}^{r_{\max}} g(r) C_{sca}(r) n(r) d(r)}{\int_{r_{\min}}^{r_{\max}} C_{sca}(r) n(r) d(r)} \tag{8}$$

As the accumulation mode is considered, the radius range is set as 0.05–0.5 µm in steps of 0.005 µm for the averaging. Note that the exact sizes of BC aggregates can be known based on these coated BC sizes and shell/core ratios. The bulk radiative properties calculated above include the scattering cross section (C_{sca}), absorption cross section (C_{abs}), single scattering albedo (SSA), and asymmetry factor (g), and these ensemble averaged properties are integrated over sizes computed for the same values of F and D_p/D_c.

The radiative properties of partially coated BC aggregates is investigated at an incident wavelength of 550 nm, and related refractive indices of BC and organics are assumed as $1.85 - 0.71i$ [38] and $1.55 - 0i$ [50], respectively. The shell/core ratio D_p/D_c of coated BC is assumed to be in the range of 1.1–2.7 on the basis of the SP2 measurements in London [33] and Beijing [34]. Note that D_c is the volume equivalent spherical core diameter of BC while D_p is the volume equivalent spherical diameter of coated BC (i.e., the whole particle). With the inhomogeneous coated BC model defined, which depicts quite realistic geometries, it is possible to systematically study the radiative properties of partially coated BC aerosols affected by their microphysics with more details.

3. Results and discussion

3.1. Effect of coating structure on radiative properties of partially coated BC

Fig. 2 illustrates radiative properties of partially coated BC aggregates at different BC coating volume fractions as a function of shell/core ratio. Five BC coating volume fractions (i.e., $F = 0.01$, 0.25, 0.5, 0.75, 0.99) and two BC fractal dimensions (i.e., $D_f \approx 1.8$, 2.8) are considered. The radiative properties (including C_{sca}, C_{abs}, SSA and g) are averaged over an ensemble of BC-organics internal-mixed particles with the aforementioned size distributions (i.e., $r_g = 0.075 \mu m$ and $\sigma_g = 1.59$). As shown in Fig. 2a–e, scattering cross sections of partially coated BC are sensitive to shell/core ratio, BC coating volume fraction, and BC geometry, and the shell/core ratio shows a more substantial impact in determining coated BC scattering. It is evident that, the C_{sca} becomes much stronger with the increase of shell/core ratio. For a BC D_f of ~2.8, as D_p/D_c increases from 1.1 to 2.7, the C_{sca} of partially coated BC increases from ~0.05 µm^2 to over 0.06 µm^2. One can see that, with a fixed BC D_f, the C_{sca} generally decreases slightly, as F increase from 0.01 to 0.99. In addition to D_p/D_c and F, the C_{sca} is also sensitive to BC geometry, and the sensitivity of the C_{sca} to BC geometry becomes stronger as D_p/D_c and F become smaller. When BC aggregates become compact, i.e., D_f increasing, for a fixed D_p/D_c and F, the C_{sca} of partially coated BC becomes large, and this is consistent with the bare BC results of Liu et al. [51].

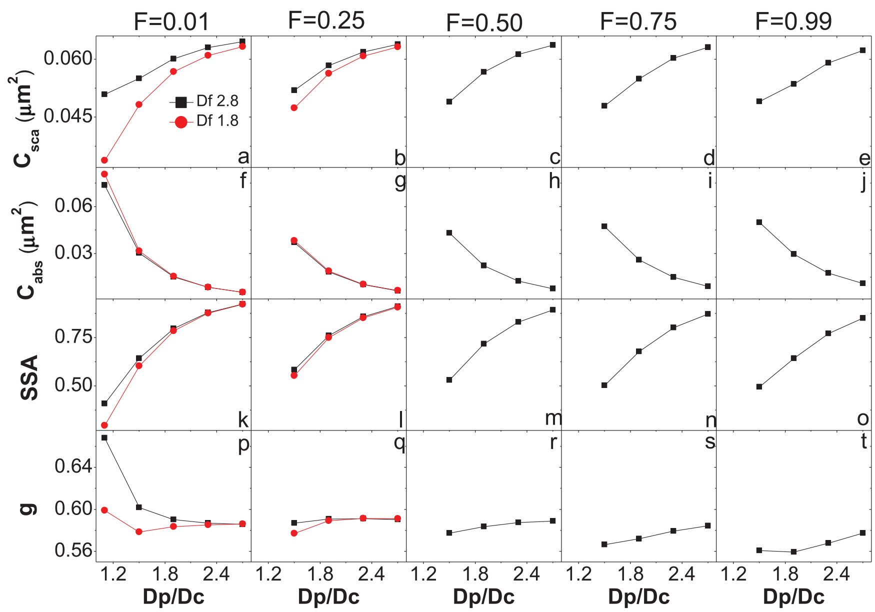

Fig. 2. Scattering cross section (C_{sca}), absorption cross section (C_{abs}), single scattering albedo (*SSA*) and asymmetry factor (*g*) of BC aggregates partially coated with organics as a function of shell/core ratio (D_p/D_c, spherical volume-equivalent particle diameter/BC core diameter). The BC coating volume fractions (*F*) of 0.01, 0.25, 0.5, 0.75 and 0.99 are considered (from left to right). Black squares and red circles indicate BC fractal dimensions of ~2.8 and ~1.8, respectively. Note that BC fractal dimension of ~1.8 is used only for *F* values of 0.01 and 0.25.

The absorption cross sections of BC particles partially coated by organics are illustrated in Fig. 2f–j. Absorption cross section of coated BC is sensitive to shell/core ratio, and the C_{abs} increases significantly with the decrease of D_p/D_c, indicating that increasing BC fraction or decreasing coating fraction can obviously enhance particle absorption as D_p is fixed. Compared to shell/core ratio, the C_{abs} of partially coated BC exhibits less sensitive to BC coating volume fraction, and the C_{abs} generally show a slight increase as *F* augments. Furthermore, Similar C_{abs} results (differences within 10%) are found between lacy and compact coated BC aggregates, although the lacy aggregates are more absorptive than the compact ones at fixed D_p/D_c and *F*. This may be due to that the magnetic field cannot penetrate deeply into compact BC aggregates, and therefore some BC monomers inside the aggregates do not contribute to the absorption [23].

With the changes on the scattering and absorption cross sections discussed, the effects of coating microphysics on the *SSA* can be easily understood, which is illustrated in Fig. 2k–o. The *SSA* is quite sensitive to D_p/D_c, and it should be expected that, for fixed *F* and BC geometry, the *SSA* increases with D_p/D_c increasing. For partially coated BC with $F = 0.5$, the *SSA* can increase in the ranges of 0.53–0.89, when D_p/D_c augments from 1.5 to 2.7. As *F* increases, the *SSA* becomes more sensitive to D_p/D_c, i.e., showing larger variation. Meanwhile, with the augment of *F*, the *SSA* decreases for fixed D_p/D_c and BC D_f, and this may be due to that the coated BC aggregates show a much stronger absorption enhancement than scattering. For partially coated BC aggregates with different BC geometries, the compact aggregates have larger *SSA* than the lacy ones, probably in association with that the absorption of coated BC decreases more than the scattering with BC becoming compact.

Fig. 2p–t shows the results of the asymmetry factor *g* of partially coated BC, which is another key aerosol parameter for radiative forcing simulation. The effect of coating microphysics on the asymmetry factor appears more complicated, and this may be associated with the significant variations of particle geometry at different coating state. The *g* seems more sensitive to D_p/D_c compared to *F* and BC D_f. For coated BC aggregates with different BC D_f, the compact aggregates generally have larger *g* than the lacy ones. Generally, radiative properties of BC aggregates partially coated by non-absorbing organics are sensitive to BC coating volume fraction, particle geometry, and shell/core ratio, wherein the shell/core ratio seems to play a more substantial role in determining the radiative properties. For a fixed size distribution, it is feasible to simplify the partially coated BC aggregate as a model with the only detailed parameter of shell/core ratio, unless BC coating volume fraction is too small and BC is thinly coated.

3.2. Effect of particle size distribution on radiative properties of partially coated BC

Fig. 3 shows radiative properties of BC aggregates (BC D_f of ~2.8) with a BC coating volume fraction of 0.5 as functions of particle size distribution and shell/core ratio. The lognormal size distributions are assumed for partially coated BC particles with r_g (*y* axis) ranging from 0.025 to 0.15 μm, and σ_g fixed as the aforementioned 1.59. Fig. 3 clearly illustrates that radiative properties of partially coated BC are quite sensitive to particle size distribution. For a fixed D_p/D_c, the C_{sca} and C_{abs} increase as r_g increases, i.e., particles becoming larger. With increased r_g, partially coted BC aggregates show a much stronger scattering enhancement than absorption, and thus, the *SSA* can increases from approximately 0.3 to 0.9 depending on D_p/D_c. Meanwhile, the asymmetry factor *g* increases with r_g increasing, mainly due to the augment of particle volumes. It is interesting to see that, the C_{sca} and *g* of partially coated BC aggregates become more sensitive to particle size distribution than D_p/D_c, although their C_{sca} and *SSA* are highly sensitive to both size distribution and D_p/D_c.'Considering the importance of *SSA* on the determination of aerosol radiative effects, the shell/core ratio and size distribution of coated BC particles should be well estimated.

The preceding analysis demonstrates that particle microphysics exert an important impact on radiative properties of partially coated BC particles. Therefore, to produce reliable estimations of BC radiative forcing from aerosol-climate models, the parameterization of realistic BC coating microphysics appears to be a must, which, nevertheless, could be a challenging task in view of more observations still being needed. However, based on our sensitivity study, BC coating volume fraction is a relatively insensitive

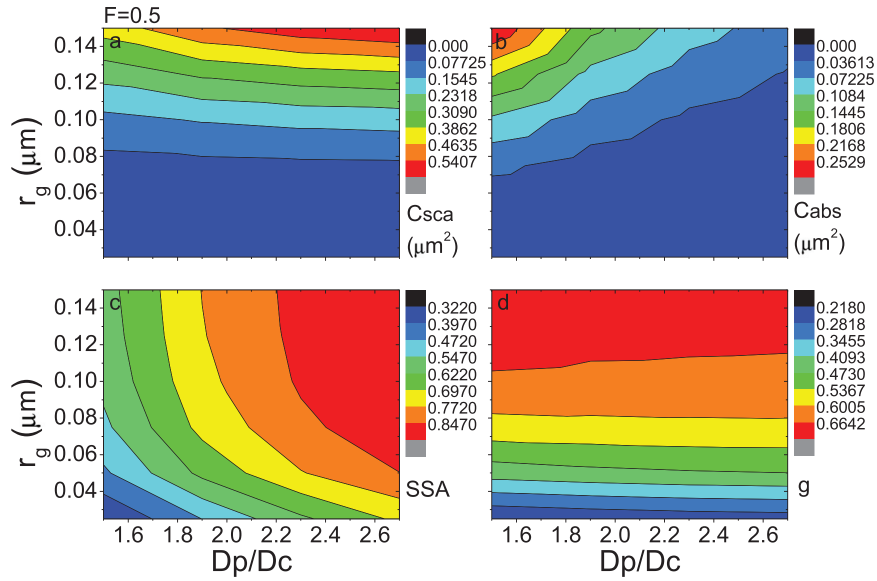

Fig. 3. Scattering cross section (C_{sca}), absorption cross section (C_{abs}), single scattering albedo (*SSA*) and asymmetry factor (*g*) of coated BC aggregates (BC fractal dimension of ~2.8) with different shell/core ratio and particle size distribution. A BC coating volume fraction of 0.5 is considered, and the geometric standard deviations (σ_g) for applied lognormal distribution is 1.59.

parameter, and BC fractal dimension is sensitive to partially coated BC radiative properties only if BC is thinly coated and BC coating volume fraction is too small. Meanwhile, particle size distribution and shell/core ratio affect radiative properties of partially coated BC significantly, wherein particle scattering and asymmetry parameter are more sensitive to size distribution.

3.3. Absorption and scattering enhancements of BC aggregates due to coating

It is reported that BC absorption and scattering can be enhanced due to coating [e.g., 52], and this subsection will discuss absorption and scattering enhancements of partially coated BC affected by their microphysics. The absorption enhancement E_{abs} and scattering enhancement E_{sca} of BC aggregates due to coating are given in forms of:

$$E_{abs} = \frac{\langle C_{abs}\rangle_{particle}}{\langle C_{abs}\rangle_{bareBC}}, \tag{9}$$

$$E_{sca} = \frac{\langle C_{sca}\rangle_{particle}}{\langle C_{sca}\rangle_{bareBC}}, \tag{10}$$

where $\langle C_{abs}\rangle_{particle}$ and $\langle C_{sca}\rangle_{particle}$ are the bulk absorption and scattering cross sections of BC aggregates coated by organics, whilst $\langle C_{abs}\rangle_{bareBC}$ and $\langle C_{sca}\rangle_{bareBC}$ are the bulk absorption and scattering cross sections of the same bare BC aggregates (without coating), respectively.

Fig. 4 shows absorption enhancements of partially coated BC aggregates (BC D_f of ~2.8) as functions of BC coating volume fraction and shell/core ratio. A fixed aforementioned lognormal size distribution is assumed for partially coated BC particles with r_g of 0.075 μm and σ_g of 1.59. Fig. 4 clearly depicts that the E_{abs} of BC aggregates due to partially coated by organics is quite sensitive to BC coating volume fraction and shell/core ratio, and can vary by a factor of 2. For BC with a small *F* close to 0, the E_{abs} of coated BC is around 1 and shows weak variation on D_p/D_c. With *F* becoming larger, the E_{abs} becomes larger and more sensitive to shell/core ratio, i.e., showing larger variation. As *F* is almost 1, the E_{abs} of coated BC aggregates reaches a value close to ~2.0. Fig. 5 illustrates absorption enhancements of partially coated BC aggregates with different particle size distribution and BC coating volume fraction. A

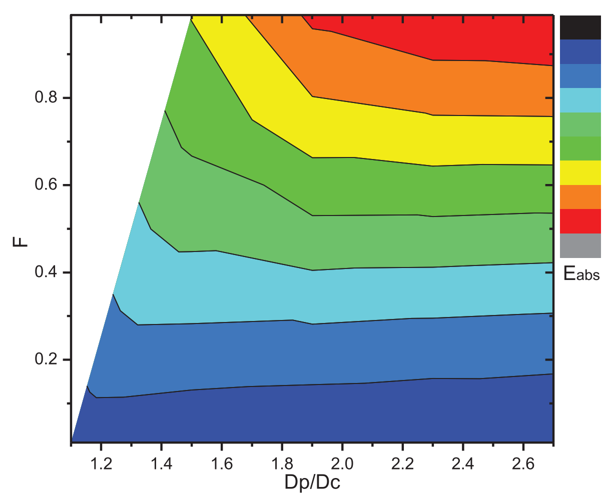

Fig. 4. Absorption enhancement (E_{abs}) of partially coated BC aggregates (BC fractal dimension of ~2.8) with different shell/core ratio (D_p/D_c) and BC coating volume fraction (*F*).

fixed BC D_f of ~2.8 and D_p/D_c of 2.7 are considered. Obviously, the E_{abs} of coated BC is sensitive to particle size distribution, and has much wider ranges for larger *F*. Overall, the absorption enhancement of BC partially coated by organics is sensitive to BC coating volume fraction, shell/core ratio, and size distribution, and the sensitivity of the absorption enhancement to particle size distribution and shell/core ratio becomes stronger as BC coating volume fraction becomes larger. For more about absorption enhancement, one can see Zhang et al. [53]. Meanwhile, BC coating volume fraction appears to be responsible for the absorption enhancement, since the absorption enhancement of coated BC can increase from ~1.0 to a value close to 2.0 with BC coating volume fraction increasing from almost 0.0 to nearly 1.0.

The scattering enhancements of BC aggregates (BC D_f of ~2.8) due to partially coated by organics as functions of BC coating volume fraction and shell/core ratio are shown in Fig. 6. The aforementioned fixed lognormal size distribution (i.e., $r_g = 0.075\mu m$ and $\sigma_g = 1.59$) is assumed for partially coated BC aggregates. As shown

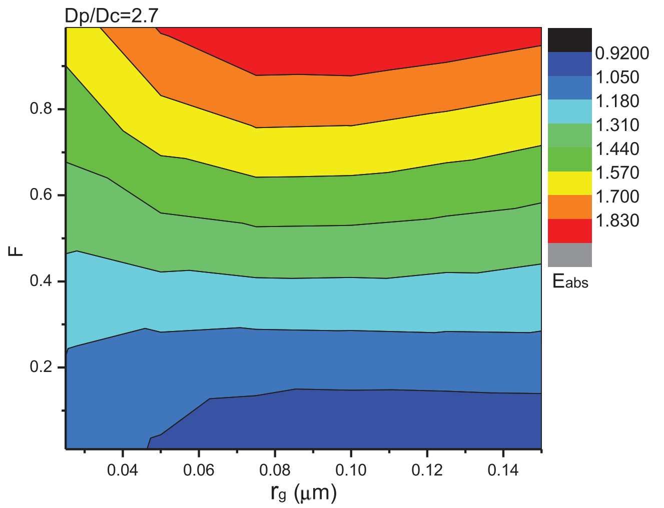

Fig. 5. Absorption enhancement (E_{abs}) of partially coated BC aggregates with different particle size distribution and BC coating volume fraction (F). The BC fractal dimension of ~2.8 and shell/core ratio of 2.7 are considered, while the geometric standard deviation (σ_g) for applied lognormal distribution is 1.59.

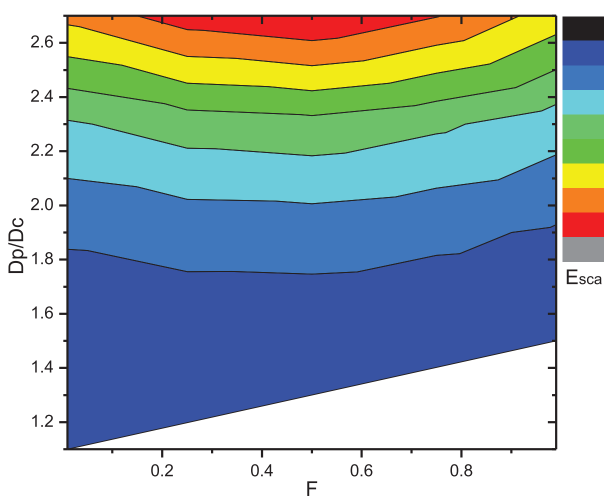

Fig. 6. Scattering enhancement (E_{sca}) of partially coated BC aggregates (BC fractal dimension of ~2.8) with different BC coating volume fraction (F) and shell/core ratio (D_p/D_c).

in Fig. 6, partially coated BC aggregates with larger D_p/D_c have larger E_{sca} for the same F, and the E_{sca} becomes more sensitive to F with increased D_p/D_c. For a thin coating with small D_p/D_c, the E_{sca} is less than 10, whereas it varies from 54 to 72 for a heavy coating with D_p/D_c reaching 2.7. This indicates that reduced BC content or increased coating fraction fortify BC scattering enhancement. Note that for a fixed D_p/D_c, the E_{sca} has a maximum with moderate F, and that the E_{sca} of coated BC with large F decreases with the increase of F may be due to the shielding effect. Fig. 7 shows scattering enhancements of partially coated BC aggregates with different particle size distribution and shell/core ratio. A BC coating volume fraction of 0.5 and BC D_f of ~2.8 are considered. The E_{sca} of partially coated BC is dependent on particle size distribution and shell/core ratio, and the dependence of the E_{sca} on shell/core ratio is larger than size distribution. The E_{sca} of partially coated BC with a fixed D_p/D_c decreases as r_g increases, i.e., particle becoming larger. For partially coated BC with a D_p/D_c of 1.5, the E_{sca} is in a range between 3 and 5, and shows weak variation on particle size distribution. Nevertheless, the variation of E_{sca} becomes stronger as the coating becomes heavier with a D_p/D_c of 2.7, and its values vary from 48 to 115. To summarize, scattering enhancement of BC due to partially coated by organics is sensitive to shell/core ratio, BC coating volume fraction and size distribution, and the sensitivities of scattering enhancement to particle size distribution and BC coating volume fraction become stronger as shell/core ratio becomes larger. Furthermore, the dependence of scattering enhancement of partially coated BC on shell/core ratio is larger than size distribution and BC coating volume fraction.

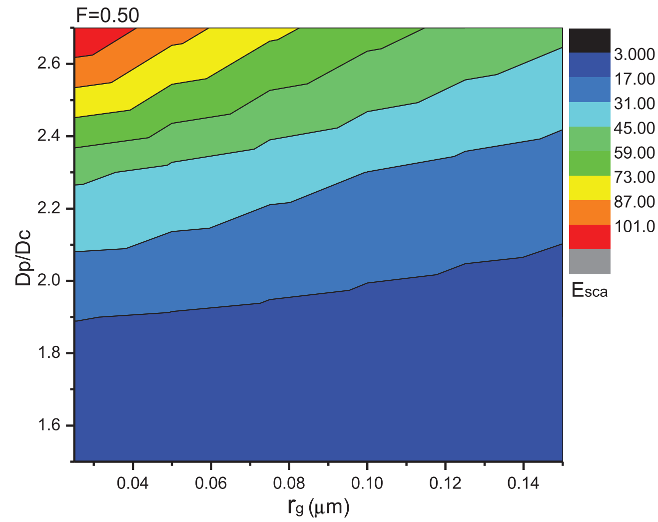

Fig. 7. Scattering enhancement (E_{sca}) of partially coated BC aggregates with different particle size distribution and shell/core ratio (D_p/D_c). The BC fractal dimension of ~2.8 and BC coating volume fraction (F) of 0.5 are considered, while the geometric standard deviation (σ_g) for applied lognormal distribution is 1.59.

4. Conclusions

The study develops a simple inhomogeneous model to study radiative properties of aged BC particles calculated by the famous MSTM, and reveals the effects of BC coating volume fraction, shell/core ratio, BC geometry and size distribution on BC radiative properties and BC absorption and scattering enhancements due to partial coating of organics. Our results indicate that radiative properties of BC aggregates partially coated by organics are sensitive to BC coating volume fraction, BC geometry, shell/core ratio, and size distribution, whereas their sensitivities are quite distinct. Some BC radiative properties are significantly impacted by some microphysical parameters, whose errors should be considered after using simplified model and parameters, whereas many insensitive BC parameters under different coating status are existent at a specified range. The BC coating volume fraction is a relatively insensitive parameter, and BC fractal dimension is sensitive to partially coated BC radiative properties only if BC is thinly coated and BC coating volume fraction is too small. Meanwhile, particle size distribution and shell/core ratio affect radiative properties of partially coated BC significantly, and of the two, particle scattering and asymmetry parameter are more sensitive to size distribution.

Our study also indicates that BC absorption and scattering can be significantly enhanced after partially coated by organics. The absorption enhancement of partially coated BC particles varies from ~1.0 to ~2.0, while their scattering enhancement can reach a value up to ~100. The BC coating volume fraction seems to be responsible for BC absorption enhancement, and with BC coating volume fraction becoming larger, the absorption enhancement becomes more sensitive to particle size distribution and shell/core ratio. However, the dependence of BC scattering enhancement on shell/core ratio is larger than size distribution and BC coating volume fraction, and the dependences of scattering enhancement to size distribution and BC coating volume fraction becomes stronger as shell/core ratio becomes larger.

The aim of this study is to investigate the sensitivities of particle microphysical parameters on radiative properties of partially coated BC aggregates, and it may be helpful for the simplifications of particle model and its parameter selection. With significant impacts on BC radiative properties, it becomes urgent to consider the coating for BC measurements and radiative simulations. The efficient model developed in our study may be employed for further simulation applications as partial coating is a more common mixing state for aged BC aerosols [18].

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