Effects of morphology on the radiative properties of internally mixed light absorbing carbon aerosols with different aging status

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Abstract: Light absorbing carbon aerosols play a substantial role in climate change through radiative forcing, which is the dominant absorber of solar radiation. Radiative properties of light absorbing carbon aerosols are strongly dependent on the morphological factors and the mixing mechanism of black carbon with other aerosol components. This study focuses on the morphological effects on the optical properties of internally mixed light absorbing carbon aerosols using the numerically exact superposition T-matrix method. Three types aerosols with different aging status such as freshly emitted BC particles, thinly coated light absorbing carbon aerosols, heavily coated light absorbing carbon aerosols are studied. Our study showed that morphological factors change with the aging of internally mixed light absorbing carbon aerosols to result in a dramatic change in their optical properties. The absorption properties of light absorbing carbon aerosols can be enhanced approximately a factor of 2 at 0.67 um, and these enhancements depend on the morphological factors. A larger shell/core diameter ratio of volume-equivalent shell-core spheres (S/C), which indicates the degree of coating, leads to stronger absorption. The enhancement of absorption properties accompanies a greater enhancement of scattering properties, which is reflected in an increase in single scattering albedo (SSA). The enhancement of single scattering albedo due to the morphological effects can reach a factor of 3.75 at 0.67 μm. The asymmetry parameter has a similar yet smaller enhancement. Moreover, the corresponding optical properties of shell-and-core model determined by using Lorenz -Mie solutions are presented for comparison. We found that the optical properties of internally mixed light absorbing carbon aerosol can differ fundamentally from those calculated for the Mie theory shell-and-core model, particularly for thinly coated light absorbing carbon aerosols. Our studies indicate that the complex morphology of internally mixed light absorbing carbon aerosols must be explicitly considered in climate radiation balance.

OCIS codes: (290.5850) Scattering, particles; (010.1110) Aerosols; (010.1310) Atmospheric scattering; (010.1290) Atmospheric optics.

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1. Introduction

Light absorbing carbon aerosols affects Earth's energy budget by absorbing solar radiation, influencing cloud processes, and altering the melting of snow and ice cover [1, 2]. Due to the black carbon (BC) that dominates absorption of visible solar radiation, light absorbing carbon aerosols could be the one of the most important anthropogenic components of global warming after CO2 in terms of direct forcing [2]. Thus, detailed knowledge of the radiative properties of light absorbing carbon aerosols is needed to clarify the mechanisms of aerosol radiative forcing [3, 4]. Thus far, however, the uncertainties in net climate forcing from light absorbing carbon aerosols are substantial [5, 6].

BC in light absorbing carbon aerosols, which is co-emitted with a variety of other aerosols and aerosol precursor gases, has a complex, fractal-like aggregate structure [7]. Soon after emission, BC mixes with other aerosol components in the atmosphere [8, 9] which can strongly influence the balance of radiative forcing by light absorbing carbon aerosols [10, 11]. The absorption of light absorbing carbon aerosols can be significantly enhanced by internal mixing with other aerosols [12–14]. However, on the basis in situ measurement studies on urban plumes, the radiative absorption enhancement for internal mixed light absorbing carbon aerosols may have been overestimated in models [15–17]. The determination of and accounting for the BC mixing state with other aerosol species is very complex and has not been completely resolved thus far.

The effects of BC morphology on the optical properties of light absorbing carbon aerosols with the external mixing state have been widely studied, and have been determined to have a strong influence on the radiative properties of externally mixed light absorbing carbon aerosols [18–21]. However, by far the most wide-spread method for computing the optical properties of internally mixed light absorbing carbon aerosols is to approximate aerosols by homogeneous spheres, with the assumption that BC is uniformly coated by a concentric shell of weakly absorbent material [22, 23]. The morphology of internally mixed light absorbing carbon aerosols is very complex, depending on the degree of aging, ambient temperature, and relative humidity [24, 25]. Because the morphological effects on the radiative properties of internally mixed light absorbing carbon aerosols have not been completely resolved, additional sophisticated optical models need to be employed for computing the optical properties of such aerosols with different morphological properties.

To investigate and quantify the morphological effects on the radiative properties of internally mixed light absorbing carbon aerosols and their effects on climate, it is essential to adequately model the entire complexity of the interrelationship of optical, physical, and chemical aerosol properties. Remarkable progress in modeling of light scattering by complex fractal aggregate particles has been achieved during the last two decades. The discrete dipole approximation (DDA) method has been used to calculate the optical properties of light absorbing carbon aggregates mixed with other aerosol [7, 25, 26]. In order to improve efficiency of DDA method, simple alternatives models, such as the "core-grey-shell" model [27], are studied to particularly fast compute the optical properties of internally mixed light absorbing carbon aerosols. Moreover, recent theory developments of the superposition T-matrix method enable calculation of the optical properties of internally mixed light absorbing carbon aerosols with various morphologies [28, 29]. The radiative properties of these aerosols can be accurate calculated by using the Multiple Sphere T-Matrix model [30, 31]. The scattering and absorption properties of micrometer-sized water droplets contaminated by black carbon were calculated using extended superposition T-matrix [32].

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The aim of this study is to investigate the radiative properties of internally mixed light absorbing carbon aerosols by comprehensively and quantitatively evaluating the morphological effects on the optical properties of light absorbing carbon aerosols. The findings should improve our understanding of the morphological effects on the radiative properties of internally mixed light absorbing carbon aerosols and their effects on climate.

2. Methodology

2.1 Internally mixed light absorbing carbon aerosol models with various morphological properties

Results of in situ measurements and laboratory studies indicate that freshly emitted BC particles tends to be coated with a thin layer of other aerosol components in the atmosphere through the coagulation and condensation of secondary aerosol compounds [33–35]. With the aging of the light absorbing carbon particles, most BC particles are thickly coated and tend to be compact [36, 37]. The irregular geometry and complex microstructure of light absorbing carbon agglomerates have been suggested to enhance the condensation of organic and inorganic components because of decreased equilibrium vapor pressure from the negative curvature (Kelvin) effect, particularly for larger particles [38, 39].

The aging process of light absorbing carbon aerosols results in a dramatic change in morphological parameters. Freshly emitted BC particles consist of small spherical primary particles combined into branched aggregates (Fig. 1(a)). For thinly coated light absorbing carbon aerosols, the BC particles are thinly coated by other aerosol components, and the morphology of the BC particles is still visible (Fig. 1(b)). For heavily coated light absorbing carbon aerosols, however, the BC particles are embedded into other aerosol components, and the morphology of BC particles is not visible (Fig. 1(c)).

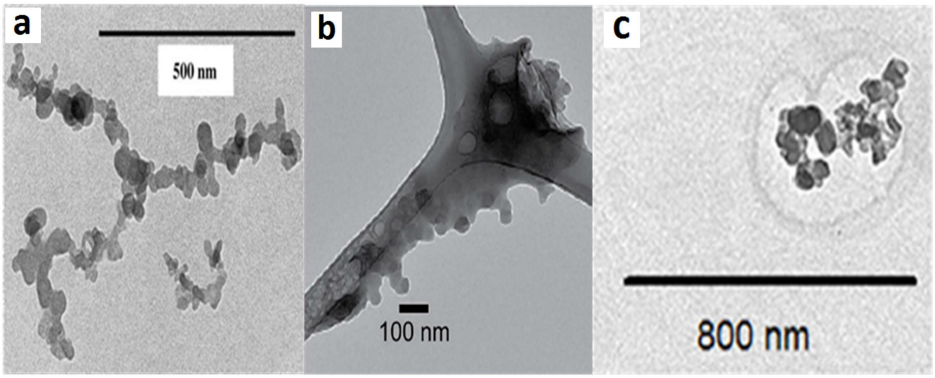

Fig. 1. Transmission electron microscope images of (a) freshly emitted black carbon (BC) particles [40], (b) thinly coated light absorbing carbon aerosols [41], and (c) heavily coated light absorbing carbon aerosols [26].

The morphologies of freshly emitted BC particles, thinly coated light absorbing carbon aerosols, and heavily coated light absorbing carbon aerosols can be modeled by using the parallel diffusion limited aggregation (DLA) algorithm [42]. The construction and morphology of the fractal clusters can be described by the well-known statistical scaling law. In this way, the relationship of the number of the monomer in a cluster N_s and the radius of gyration R_g can be linked by the fractal prefactor k_0 and the fractal dimension D_f. The specific types of clusters were simulated by using the parallel DLA algorithm with input parameters such as N_s, k_0, D_f.

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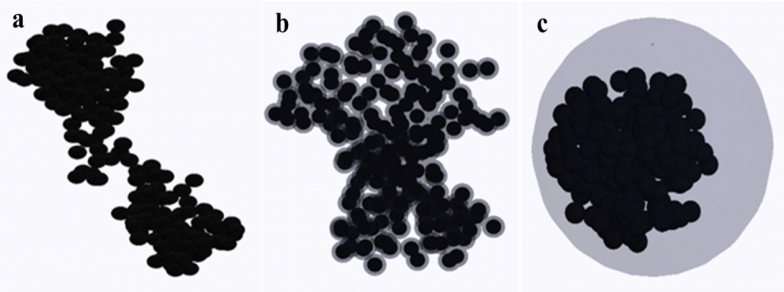

Fig. 2. Schematic diagram showing the transformation of light absorbing carbon aggregates with increasing mass fraction of condensed materials: (a) freshly emitted black carbon (BC) particles, (b) thinly coated light absorbing carbon aerosols, (c) heavily coated light absorbing carbon aerosols.

Compared with freshly BC particles (Fig. 2(a)), each monomer of thinly coated light absorbing carbon aerosols can be assumed as having structures of a concentric core containing BC particles with high absorption and a shell containing weakly absorbing particles, such as organic (Fig. 2(b)) rather than the spherical homogeneous simplification. For heavily coated light absorbing carbon aerosols, BC particles are engulfed in the other components. In such cases, the morphology alters to compact closed structures, and the morphology of other materials coated on the BC particles is simply assumed as homogeneous spheres (Fig. 2(c)). The other material coating the BC particles is assumed to be sulfate in this study. The morphological properties of internally mixed light absorbing carbon aerosols [43–46] are shown in Table 1. The S/C ratio of the soot-sulfate mixtures are defined as the diameter ratio of the volume-equivalent spherical soot core without sulfate shell and the total volume-equivalent sphere with sulfate shell.

Table 1. Morphological Properties of Internally Mixed Light Absorbing Carbon Aerosols

Particle	Morphological parameters	Freshly emitted	Thinly coated	Heavily coated
Light absorbing carbon	k_0	1.2	1.2	1.2
	a	0.02 μm	0.02 μm	0.02 μm
	N_s	200	200	200
	D_f	2.0, 2.2, 2.4, 2.6, 2.8, 3.0	2.0, 2.2, 2.4, 2.6, 2.8, 3.0	2.6, 2.8, 3.0
Sulfate	*(S/C)*	1.0	1.1, 1.3, 1.5, 1.75, 2.0	1.5, 1.75, 2.0

The refractive indices of BC particles are those reported by Change and Charalampopulos [47], whereas the refractive indices of sulfate were obtained from the Optical Properties of Aerosols and Clouds (OPAC) database [48].

2.2 Superposition T-matrix method

The superposition T-matrix approach, which uses the numerically exact solution methods to Maxwell's equations, can be used to calculate the T-matrix descriptions of the light scattering from the cluster with an appropriate superposition technique, and analytically obtain the random-orientation cross sections and scattering matrices of these clusters. Until now, the superposition solution for scattering by a system of optically active spheres is extended to the case where any of the spheres can be located at points that are either internal or external to the other sphere [30].

Using the parallel diffusion limited aggregation (DLA) algorithm and the morphological features in field-emission transmission electron microscope images, the construction and morphology of internally mixed light absorbing carbon aerosols are modeled to initialize the

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superposition T-matrix model, MSTM version 3.0 programs. The absorption, scattering and extinction efficiencies are the output variables of the MSTM program.

In the open-source Fortran coded MSTM version 3.0 program, both exteral and internal mixtures are applicable with the only limitation that the spherical surfaces are nonoverlapping. Each spherical surface will have an associated host sphere, which being the sphere whose refractive index is in contact with the exterior surface. The external medium of these scatters is assumed as vacuum. The random orientation scattering properties are obtained analytically from the superposition T-matrix model method, and can be averaged by multiple calculations for different clusters with the same morphological parameters.

3. Optical properties of internally mixed light absorbing carbon aerosols with various morphological properties

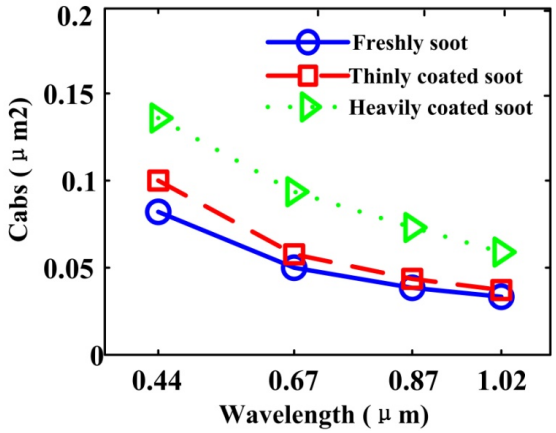

Fig. 3. Absorption cross sections (*Cabs*) of internally mixed light absorbing carbon aerosols with various morphological factors: freshly emitted BC particles (D_f = 2.0, S/C = 1.0), thinly coated light absorbing carbon aerosols (D_f = 2.4, S/C = 1.5), and heavily coated light absorbing carbon aerosols (D_f = 2.8, S/C = 2.0).

The absorption cross sections of light absorbing carbon aerosols represent one of the major factors leading to large uncertainties in the evaluation of light absorbing carbon aerosol climate effects. Figure 3 shows the variation of absorption cross sections of internally mixed light absorbing carbon aerosols with various morphological parameters, including freshly emitted BC particles, thinly coated light absorbing carbon aerosols, and heavily coated light absorbing carbon aerosols. The mass absorption cross sections (MAC) can be computed from the absorption cross sections, which are defined as the absorption cross sections per unit mass of light absorbing carbon.

Estimated of MAC depend on the assumed refractive index and mass density of BC particles. Bond and Bergstrom [49] recommend the values of refractive index at 0.55 μm as following values: $m = 1.75 + 0.63i$, $m = 1.80 + 0.67i$, $m = 1.85 + 0.71i$, $m = 1.90 + 0.75i$, and $m = 1.95 + 0.79i$. The density of real BC particles recommended as 1.7-1.9 g/cm^3 [49]. We assume the refractive index $m = 1.75 + 0.63i$, and the density $\rho_{BC} = 1.8 g/cm^3$. For freshly emitted BC particles, the estimated MAC value (at 0.55 μm) is 5.5 m^2/g. When the refractive index is $m = 1.95 + 0.79i$, the estimated MAC value (at 0.55 μm) is 6.03 m^2/g. Estimated of MAC are in agreement with the range of theoretical modeled values [50, 51] and smaller than the values modeled by Scarnato et al. [52] using a discrete dipole approximation model. Estimated of MAC lies in the range of field measurements of MAC for carbonaceous particles varying from 5 to 25 m^2/g [53, 54], although these values lie below the range

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of $MAC = (7.5 \pm 1.2)\,\mathrm{m}^2/g$ recommended by Bond and Bergstrom [49] based on the publications of measurements.

During the alternation of these morphological parameters, the light absorption can be enhanced approximately a factor of 2, where the enhancement is defined as $Cabs_{coated} / Cabs_{fresh}$, at 0.67 um, and the enhancements depend on the morphological parameters. The coating increases the geometric cross section of the particles, thus intercepting more electromagnetic energy, which, inside the particle, interacts with the highly absorbing carbonaceous material, thus increasing the absorption cross section. For thinly coated light absorbing carbon aerosols, the enhancement is approximately a factor of 1.1 at 0.67 um; that for heavily coated light absorbing carbon aerosols can reach a factor of 2.

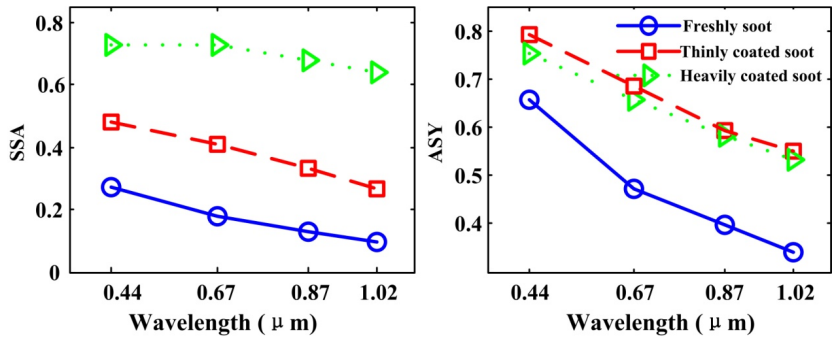

Fig. 4. Single scattering albedo (SSA) and asymmetry parameter (ASY) of internally mixed light absorbing carbon aerosols with various morphological factors: freshly emitted black carbon particles (D_f = 2.0, S/C = 1.0), thinly coated light absorbing carbon aerosols (D_f = 2.4, S/C = 1.5), and heavily coated light absorbing carbon aerosols (D_f = 2.8, S/C = 2.0).

Figure 4 shows the variation of scattering properties including single scattering albedo and asymmetry parameter of internally mixed light absorbing carbon aerosols with various morphological parameters. For freshly emitted BC particles, the single scattering albedo was as low as 0.20 at 0.67um, which is consistent with the measurements for fresh diesel exhaust light absorbing carbon [55], and in agreement with the suggested value of 0.25 ± 0.05 (at 0.55 μm) [49]. The mass extinction cross sections (MEC) can be retrieved from the mass absorption cross sections and single scattering albedo. When the refractive index is $m = 1.95 + 0.79i$ and the density is $\rho_{BC} = 1.8 g/cm^3$, the estimated MEC value (at 0.633 μm) is 7.4 m^2/g, which lie below in the range of $MEC = (8.7 \pm 1.1)\,\mathrm{m}^2/g$ recommended by Mulholland and Croarkin [56] based on the publications of measurements. Possible sources of bias are the choices of refractive index and the density of BC particles.

The scattering properties of coated light absorbing carbon aerosols were significantly higher than those of freshly emitted black carbon particles, even for thinly coated light absorbing carbon aerosols. The single scattering albedo changed from 0.2 for freshly emitted BC particles to 0.4 for thinly coated light absorbing carbon up to 0.75 for heavily coated light absorbing carbon. Moreover, the asymmetry parameter changed from 0.45 for freshly emitted BC particles to 0.7 for heavily coated light absorbing carbon.

During the aging process of light absorbing carbon aerosols, the enhancement of absorption properties accompany an enhancement of scattering properties, which is reflected by an increase in single scattering albedo. The asymmetry parameter also increased during the aging process, which indicates that there is less scattering of incoming radiation back to its source. The enhanced single scattering albedo and asymmetry parameter can lead to a decrease in solar radiation absorption into the atmospheric layer [57–59].

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4. Effects of morphology on the optical properties of internally mixed aerosols

During the aging process of light absorbing carbon aerosols, optical properties such as absorption and scattering are dramatically changed due to the effects of morphology such as formation of coated shell and compaction of BC particles. To quantitatively evaluate the morphological effects on the optical properties of light absorbing carbon aerosols, the shell/core ratio (S/C) was used to represent the states of coating, and fractal dimensions were used to represent the states of compaction of the BC particles.

4.1. Effects of shell/core ratio (S/C)

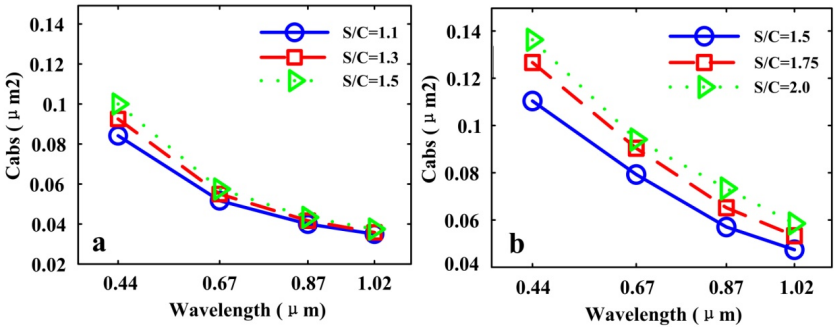

Fig. 5. Absorption cross sections of internally mixed light absorbing carbon aerosols with various shell/core ratio (S/C): (a) thinly coated light absorbing carbon aerosol (D_f = 2.4, S/C = 1.1, 1.3, 1.5), (b) heavily coated light absorbing carbon aerosol (D_f = 2.8, S/C = 1.5, 1.75, 2.0).

The S/C is an important factor in the enhancement of optical properties of internally mixed light absorbing carbon aerosols. Figure 5 shows the variation of absorption cross sections of internally mixed light absorbing carbon aerosols with different shell/core ratio (S/C). During the aging process, a larger S/C leads to stronger absorption. The absorption was enlarged from 0.05 (S/C = 1.1) to 0.1 (S/C = 2.0) at 0.67 μm, which corresponds to an enhancement of a factor of 2. Decreasing the size parameter ($x = \pi D / \lambda$, where D is the particle diameter and λ is the wavelength) resulted in an increase in absorption enhancement. For thinly coated light absorbing carbon, the enhancement of absorption properties due to S/C reached to a factor of 1.25 at 0.44 μm, whereas the enhancement at 1.02 μm can be ignored. For heavily coated light absorbing carbon, the enhancement of absorption properties due to S/C reached a factor of 1.27 at 0.44 μm, whereas the enhancement at 1.02 μm reached a factor of 1.2.

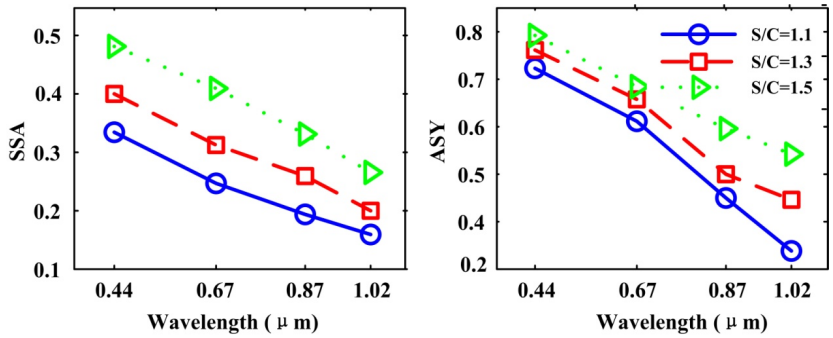

Fig. 6. Single scattering albedo (SSA) and asymmetry parameter (ASY) of thinly coated light absorbing carbon aerosols with various shell/core ratio (S/C). D_f = 2.4, S/C = 1.1, 1.3, 1.5.

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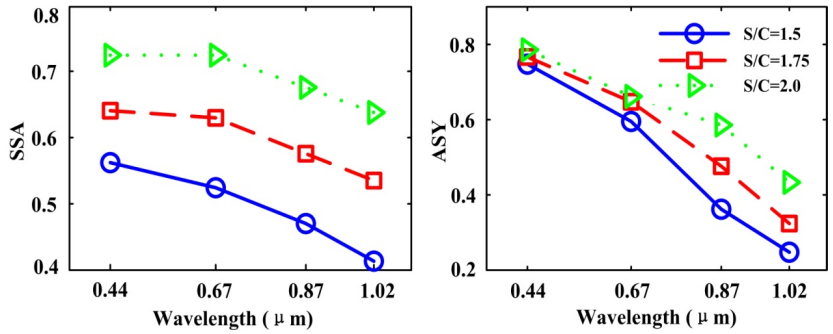

Fig. 7. Single scattering albedo (SSA) and asymmetry parameter (ASY) of heavily coated light absorbing carbon aerosols with various shell/core ratio (S/C). D_f = 2.8, S/C = 1.5, 1.75, 2.0.

Coating BC particles with sulfate resulted in dramatic changes in scattering properties (Figs. 6 and 7). During the aging process, a larger S/C leads to stronger scattering properties (single scattering albedo and asymmetry parameter). The single scattering albedo was enlarged from 0.25 (S/C = 1.1) to 0.72 (S/C = 2.0) at 0.67 μm, which corresponds to an enhancement of a factor of 2.88. The asymmetry parameter was enlarged from 0.6 (S/C = 1.1) to 0.7 (S/C = 2.0) at 0.67 μm, which corresponds to an enhancement of a factor of 1.17. For thinly coated light absorbing carbon, the enhancement of single scattering albedo due to S/C reached a factor of 1.68 at 0.67 μm, whereas for heavily coated light absorbing carbon, the value reached a factor of 1.38 at 0.67 μm. The asymmetry parameter had a similar yet smaller enhancement.

4.2. Effects of fractal dimensions

The effects of D_f on the optical properties of freshly emitted BC particles were studied, and results indicate D_f is a very important morphological parameter for the evaluation of the optical properties of such particles. In this study, therefore, the effects of D_f on the optical properties of only thinly coated light absorbing carbon aerosols and heavily coated light absorbing carbon aerosols were studied.

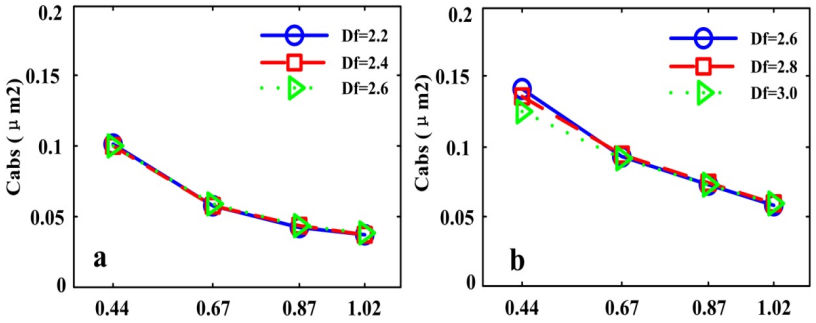

Fig. 8. Absorption cross sections of internally mixed light absorbing carbon aerosols with various fractal dimensions (D_f): (a) thinly coated light absorbing carbon aerosol (D_f = 2.2, 2.4, 2.6, S/C = 1.5), (b) heavily coated light absorbing carbon aerosol (D_f = 2.6, 2.8, 3.0, S/C = 1.75).

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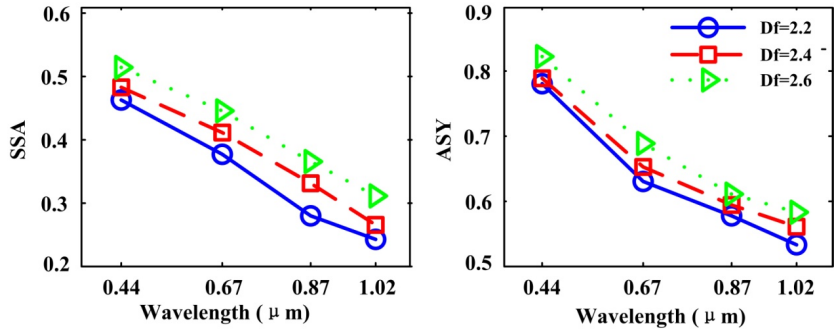

Fig. 9. Single scattering albedo (SSA) and asymmetry parameter (ASY) of thinly coated light absorbing carbon aerosols with various fractal dimensions (D_f = 2.2, 2.4, 2.6, S/C = 1.5)

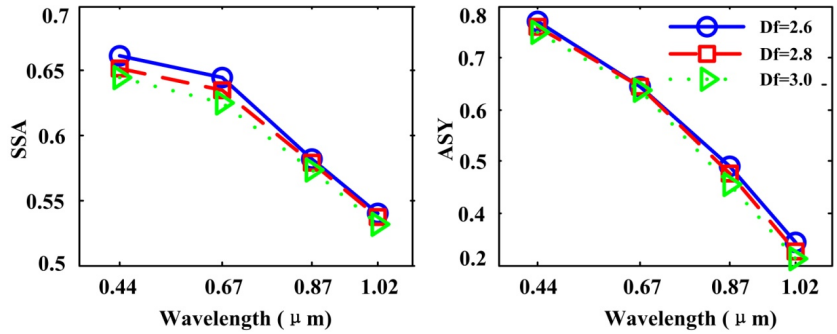

Fig. 10. Single scattering albedo (SSA) and asymmetry parameter (ASY) of heavily coated light absorbing carbon aerosols with various fractal dimensions (D_f = 2.6, 2.8, 3.0, S/C = 1.75)

Figure 8 shows the variation of absorption cross sections of internally mixed light absorbing carbon aerosols (thinly coated and heavily coated) with various fractal dimensions (D_f). Contrary to effects of D_f on the absorption cross sections of freshly emitted BC particles, for thinly coated aerosol, the effects of D_f on the absorption cross sections can be ignored. This general behavior was also found by Wu et al. [31]. Figures 9 and 10 show the effects of fractal dimensions on the scattering properties of internally mixed light absorbing carbon aerosols. For thinly coated light absorbing carbon aerosols, values of single scattering albedo are higher for compact aggregates than for lacy aggregates, due to a stronger scattering interaction and stronger electromagnetic coupling between spherules [60]. The effects of fractal dimensions enhanced various in single scattering albedo and the asymmetry parameter a factor of 1.18 and 1.10 at 0.67 μm. For heavily coated light absorbing carbon aerosols, the effects of fractal dimensions on the scattering properties were small and can be ignored due to the effect of sulfate in the heavy coating.

5. Optical properties of alteration of internally mixed light absorbing carbon aerosols due to assumption of concentric spherical core–shell model

Thus far, the most wide-spread method used for computing the optical properties of internally mixed light absorbing carbon aerosols is the Mie theory with a shell-and-core model, which approximates BC particles by homogeneous spheres with the assumption that BC is uniformly coated by a concentric shell of weakly absorbent material. To quantify the morphological effects on the radiative properties of internally mixed light absorbing carbon aerosols and their effects on climate, the corresponding optical properties of the Mie theory shell-and-core model are presented for comparison.

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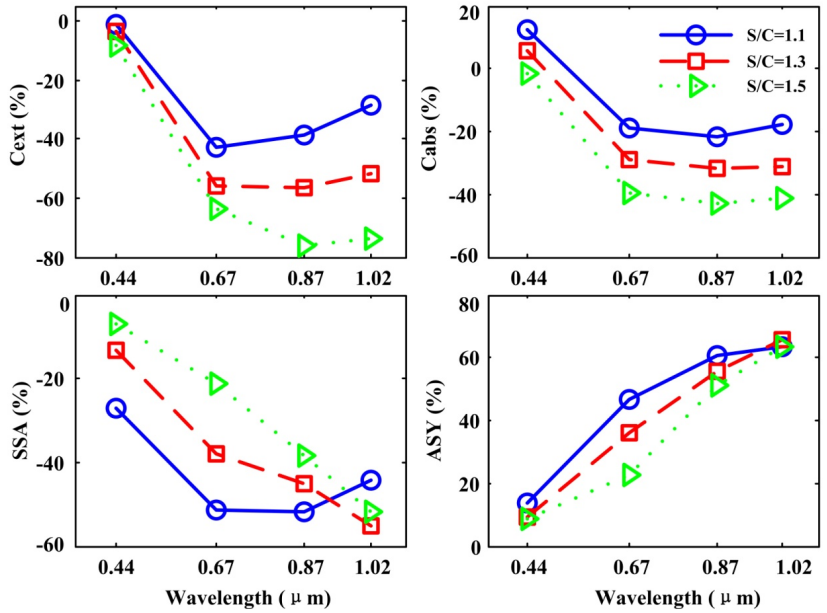

Fig. 11. Ranges for optical properties of thinly coated light absorbing carbon aerosols with alterations due to assumption of the concentric spherical core-shell model (D_f = 2.4, S/C = 1.1, 1.3, 1.5).

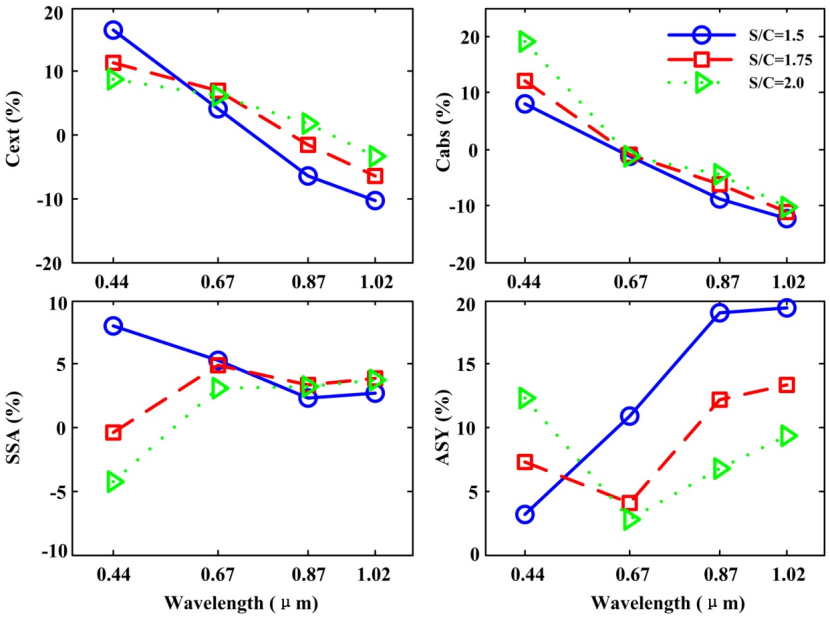

Fig. 12. Ranges for optical properties of heavily coated light absorbing carbon aerosols with alteration due to assumption of the concentric spherical core-shell model (D_f = 2.8, S/C = 1.5, 1.75, 2.0).

Figures 11 and 12 show the ranges for optical properties of internally mixed light absorbing carbon aerosols (thinly coated, and heavily coated) with alteration due to assumption of the concentric spherical core-shell model. For thinly coated light absorbing carbon aerosols, the Mie theory shell-and-core model resulted in a dramatic change in the internally mixed light absorbing carbon aerosol optical properties. In this model, the variation

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of extinction coefficient reached 80%, and the variation of absorption properties reached 40%, where the variation is defined as $(C_{MSTM} - C_{Mei}) / C_{MSTM} * 100\%$. The scattering properties, including single scattering albedo and asymmetry parameter showed similar changes. The variation of single scattering albedo reached 60%, and the variation of asymmetry parameter reached 60%.

For the heavily coated light absorbing carbon aerosols the variation of extinction coefficient was smaller than 20%, and the variation of absorption properties was smaller than 10%. For scattering properties, the variation of single scattering albedo was smaller than 10%, and the variation of asymmetry parameter was smaller than 20%. Compared with thinly coated light absorbing carbon aerosols, the ranges for optical properties of heavily coated light absorbing carbon aerosols with alteration due to the assumption of the concentric spherical core-shell model were weakened.

The optical properties of internally mixed light absorbing carbon aerosol can differ fundamentally from those calculated for Mie theory shell-and-core model, particularly for thinly coated light absorbing carbon aerosol. This indicates that the complex morphology of internally mixed light absorbing carbon aerosols must be explicitly considered in climate radiation balance.

6. Summary and discussion

In this study, morphological effects on the optical properties of internally mixed light absorbing carbon aerosols with different aging status are studied using the numerically exact superposition T-matrix method. The aging process of light absorbing carbon aerosols resulted in dramatic changes in the morphological factors including coating and compaction. During the morphological parameter alteration, the light absorption was enhanced approximately a factor of 2, where the enhancement is defined as $\mathrm{Cabs}_{coated} / \mathrm{Cabs}_{fresh}$, at 0.67 um, and the enhancements depended on the morphological factors. A larger S/C leads to stronger absorption. The absorption was enlarged from 0.05 (S/C = 1.1) to 0.1 (S/C = 2.0) at 0.67 μm, which corresponds to an enhancement of a factor of 2.

The enhancement of absorption properties accompanied greater enhancement of scattering properties larger enhanced, which was reflected in the increase in single scattering albedo. The single scattering albedo changed from 0.2 for freshly emitted BC particles to 0.4 for thinly coated light absorbing carbon up to 0.75 for heavily coated light absorbing carbon. The asymmetry parameter also increased due to the changes in morphological parameters, which indicates that there was less scattering of incoming radiation back to its source. The asymmetry parameter changed from 0.45 for freshly emitted BC particles to 0.7 for heavily coated light absorbing carbon. A larger S/C leads to stronger scattering properties (single scattering albedo and asymmetry parameter). The single scattering albedo was enlarged from 0.25 (S/C = 1.1) to 0.72 (S/C = 2.0) at 0.67 μm, which corresponds to an enhancement of a factor of 2.88. The asymmetry parameter was enlarged from 0.6 (S/C = 1.1) to 0.7 (S/C = 2.0) at 0.67 μm, which corresponds to an enhancement of a factor of 1.17. Such enhancement in single scattering albedo and the asymmetry parameter can lead to a decrease in solar radiation absorption into the atmospheric layer.

Our studies show that the optical properties of internally mixed light absorbing carbon aerosol can differ fundamentally from those calculated for the Mie theory shell-and-core model, particularly for thinly coated light absorbing carbon aerosol. For thinly coated light absorbing carbon aerosols, the Mie theory shell-and-core model resulted in the variation of extinction coefficient reached 80%, the variation of absorption properties reached 40%, the variation of single scattering albedo reached 60%, and the variation of asymmetry parameter reached 60%, where the variation is defined as $(C_{MSTM} - C_{Mei}) / C_{MSTM} * 100\%$. In order to improve the accurate shell-and-core model simulation, some simple alternatives were studied, such as the “core-grey-shell” model, which seems to give highly accurate results.

Our studies indicate that the complex morphologies of internally mixed light absorbing carbon aerosols must be explicitly considered in the climate radiation balance. These findings

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should improve our understanding of morphological effects on the radiative properties of internally mixed light absorbing carbon aerosols and their effects on climate.

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