Multiple scattering of light by water cloud droplets with external and internal mixing of black carbon aerosols*

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(Received 2 August 2011; revised manuscript received 14 October 2011)

The mixture of water cloud droplets with black carbon impurities is modeled by external and internal mixing models. The internal mixing model is modeled with a two-layered sphere (water cloud droplets containing black carbon (BC) inclusions), and the single scattering and absorption characteristics are calculated at the visible wavelength of 0.55 μm by using the Lorenz–Mie theory. The external mixing model is developed assuming that the same amount of BC particles are mixed with the water droplets externally. The multiple scattering characteristics are computed by using the Monte Carlo method. The results show that when the size of the BC aerosol is small, the reflection intensity of the internal mixing model is bigger than that of the external mixing model. However, if the size of the BC aerosol is big, the absorption of the internal mixing model will be larger than that of the external mixing model.

Keywords: aerosols multiple scattering, Monte Carlo method, phase function

PACS: 42.25.Fx, 42.25.Bs **DOI:** 10.1088/1674-1056/21/5/054204

1. Introduction

Black carbon (BC) has long been recognized as an important atmospheric pollutant.[1] The enhanced absorption by BC particles imbedded in water droplets could potentially reduce the cloud albedo,[2] thereby causing a significant indirect forcing of climate.[3] The scattering and the radiative properties of an internal cloud droplet mixture can differ from those of a composition-equivalent external mixture. The potential differences may influence the results of remote sensing studies of tropospheric aerosols and calculations of the direct aerosol forcing of the climate.[4,5]

The effect of BC impurities on the absorption of solar radiation by cloud water droplets was considered by Danielson *et al.*[6] using an idealized model. Liu *et al.*[7] calculated the single scattering differences between the water cloud droplets with BC-fraction-equivalent internal and external mixtures. Mishchenko *et al.*[8] calculated the optical cross sections and the elements of the Stokes scattering matrices of semi-external two-component mixtures of different types of aerosols.

In this paper, we compute the multiple scattering characteristics of water cloud droplets containing BC inclusions, assuming that the inclusions are placed centrally in the outer droplet (Fig. 1(a)), and compare them with those of the same amount of BC particles mixed with water droplets externally (Fig. 1(b)).

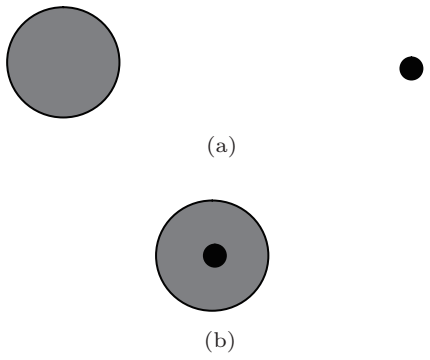


Fig. 1. (a) External and (b) internal particle mixtures.

2. Single scattering

In our computations, we assume that the shapes of a black carbon particle and a cloud droplet are spherical, and the internal mixing model is a concentric sphere, the external mixture is modeled by cloud droplets and black carbon aerosols separated by dis-

*Project supported by the Natural Science Foundation of Shandong Province, China (Grant No. ZR2009AQ013).

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tances much greater than their sizes and scattering light independently of each other. Important single scattering characteristics of discrete random media are the ensemble averaged scattering and extinction cross sections ($C_{\rm scat}$ and $C_{\rm ext}$) and the normalized scattering phase function $P(\theta)$, where θ is the scattering angle. Additional useful quantities include the ensemble averaged absorption cross section, the single scattering albedo ω_0, and the asymmetry parameter g defined as[9]

$$g = \frac{1}{2}\int_0^{\pi} \mathrm{d}\theta \sin\theta P(\theta)\cos\theta. \tag{1}$$

Due to the variability of physical properties of clouds in both space and time domains, the size of a cloud particle is polydisperse. Thus we can consider the radius of droplet r as a random value, which is characterized by distribution function $n(r)$.

The computations are performed at a visible wavelength of $\lambda = 0.55$ μm assuming that each aerosol model is represented by a gamma distribution of the following type:[10]

$$n(r) = \text{const} \times r^{(1-3b)/b} \exp\left(\frac{-r}{ab}\right). \tag{2}$$

Hansen and Travis[11] found that the effective radius and variance

$$r_{\rm ef} = \frac{\int_0^{\infty} r\pi r^2 n(r)\mathrm{d}r}{\int_0^{\infty} \pi r^2 n(r)\mathrm{d}r}, \tag{3}$$

$$v_{\rm ef} = \frac{\int_0^{\infty} (r - r_{\rm ef})^2 \pi r^2 n(r)\mathrm{d}r}{r_{\rm ef}^2 \int_0^{\infty} \pi r^2 n(r)\mathrm{d}r} \tag{4}$$

are important parameters for any particle-size distribution, and they also found that the size distributions for different clouds with the same values of $r_{\rm ef}$ and $v_{\rm ef}$ will have similar scattering properties. For the gamma distribution, $a = r_{\rm ef}$, $b = v_{\rm ef}$.

In our computations, we have used two types of aerosol particle mixtures. Mixture 1 is composed of concentric sphere particles with an effective shell (water cloud droplet) radius of 6 μm, and the effective radius of the core component (black carbon) is set at the following four representative values: 0.1 μm, 0.5 μm, 1.0 μm, and 3.0 μm. Mixture 2 consists of equal numbers of water cloud droplets and black carbon particles in the form of an external mixture, and the effective radii are the same as those of mixture 1. We use the following values of the relative refractive indexes for the two different aerosol species: 1.33 for water cloud droplet, and $1.75 + 0.44$i for black carbon.[12] The effective variance for all aerosol types is fixed at 1/9, thereby representing a moderately wide size distribution. Although solid aerosol particles should be presumed to have nonspherical shapes, our main interest here is in evaluating the potential effects of different type mixtures on light scattering. Therefore, for the sake of simplicity, we assume that the black carbon aerosol species consist of spherical particles.

We calculate the single scattering characteristics of concentric sphere clusters and compare the results with those of pure water cloud droplets and black carbon particles with the same size distributions. Figure 2(a) gives the phase functions of concentric spheres with different effective core sizes. We can see that the relative phase function differences are rather small. A notable exception is the case with the effective core radius of 3 μm, which represents a relatively larger

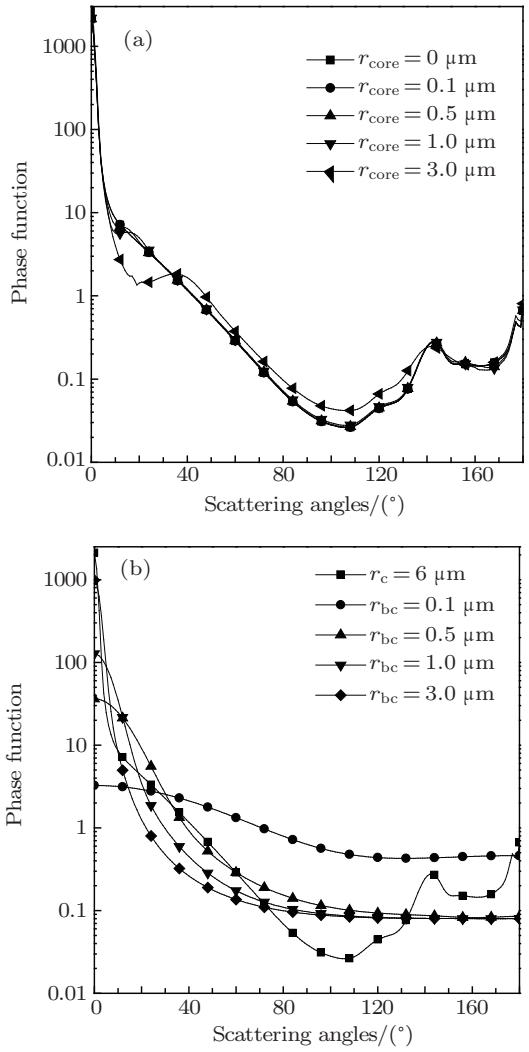


Fig. 2. Phase functions for water cloud droplets, black carbon aerosols, and the concentric spheres with water droplet as the shell and BC impurities as the core. Panel (a) shows the results with different effective core sizes, and panel (b) shows the results with an effective core radius of 6 μm.

core. Figure 2(b) gives the phase functions of water droplets with an effective radius of 6 μm. The results are compared with the phase function of BC aerosols. We can see that the differences are large, especially at the exact forward-scattering direction where the interference effects result in a significant enhancement of the aggregate phase functions.

Tables 1 and 2 give the scattering and extinction cross sections, the single scattering albedos, and the asymmetry parameters for pure water cloud droplets, BC aerosols, and internal mixtures with an effective radius of water cloud droplet equaling to 6 μm. From the tables, we can see that the single scattering albedo decreases with the increase of the core size for the internal mixtures. However, for the BC aerosols, the law is exactly the opposite. The extinction cross sections of the internal mixtures are the same as those of the pure water cloud droplets whatever the size of the core, but the scattering cross section decreases with the increase of the size of the core.

Table 1. Optical characteristics of black carbon inner mixtures.

Effect radius of core/μm	0.1	0.5	1.0	3.0
$C_{\rm ext}$	167.38	167.38	167.38	167.38
$C_{\rm sca}$	167.35	166.19	163.03	132.95
g	0.851	0.8497	0.8469	0.827
ω_0	0.99979	0.9929	0.9740	0.7943

Table 2. Optical characteristics of black carbon aerosols and water cloud droplets.

Type	Cloud droplet	BC	BC	BC	BC
Effect radius/μm	6	0.1	0.5	1.0	3.0
$C_{\rm ext}$	167.38	0.0318	1.433	5.24	43.19
$C_{\rm sca}$	167.4	0.0109	0.687	2.677	23.82
g	0.851	0.373	0.83	0.88	0.90
ω_0	1	0.343	0.479	0.511	0.552

3. Multiple scattering simulations using the Monte Carlo method

The multiple scatterings of light by a cluster of water cloud droplets containing black carbon impurities and the same amount of BC particles mixing with water droplets externally are calculated by using the Monte Carlo techniques.[13] The cloud layer is assumed to be a vertically homogeneous plane-parallel layer.

After an incident photon is launched into the medium, it travels a free path distance l given by

$$l = -\langle l\rangle \log[R(0,1)], \tag{5}$$

where $\langle l\rangle$ is the mean free path length between two subsequent scattering events, and $R(0,1)$ is a uniformly distributed random number within the interval of (0,1). The number density of inclusion n is described by the mean free path length $\langle l\rangle$ or by the volume extinction coefficient $\beta_x = 1/\langle l\rangle$. For an ensemble of N particles per volume with a standard gamma function distribution of particle size, β_x is given by

$$\beta_x = N\int_{r_1}^{r_2} \beta_x(r) n(r)\,{\rm d}r, \tag{6}$$

where $n(r)$ is the normalized particle size distribution function. In this paper, we choose $n(r)$ to be a uniform distribution function.

For a cluster of water cloud droplets containing BC inclusions, if the photon has not reached one of the boundaries of the medium, its previous direction is changed along scattering angle α and azimuth angle ϕ according to

$$\int_0^{\alpha} P(\theta)\sin\theta\,{\rm d}\theta = R(0,1)\int_0^{\pi} P(\theta)\sin\theta\,{\rm d}\theta, \tag{7}$$

$$\phi = R(0,2\pi), \tag{8}$$

where $P(\theta)$ denotes the scattering phase function of the medium. The processes stated in Eqs. (5) and (7) repeat until the photon enters one of the boundaries of the particle layer or the photon energy falls below 10^{-5} of the incident energy. However, there exists a difference in the direction changing between BC-fraction-equivalent internal and external mixtures, which is the scattering angle α. When the photon enters the external mixture layer, it can be scattered to the water cloud droplets, and it can also be scattered to the black carbon particles. Because the numbers of the two types of particles are the same, the probabilities are 50% each. If a photon is scattered by the black carbon particles, its previous direction will be changed along scattering angle α according to Eq. (7), and the phase function of Eq. (7) will be the phase function of the black carbon particles. If it is scattered by the water cloud droplet, the phase function of Eq. (7) will be the phase function of the water cloud droplets.

We compute the light intensities diffusely reflected by the particle layers consisting of the two types of particles respectively, i.e., water droplets internally and externally mixed with the BC aerosols.

It is convenient to define reflection and transmission functions as[14]

$$I_{\rm r}(0,\mu,\phi) = \mu_0 R(\tau,\mu,\mu_0,\phi-\phi_0)F, \tag{9}$$

where $I_{\rm r}(0,\mu,\phi)$ is the reflected light intensity, $R(\tau,\mu,\mu_0,\phi-\phi_0)$ is called the reflection function, τ is the optical thickness, μ_0 is the incidence angle, μ is the observation angle, ϕ–ϕ' is the difference between the incidence and the observation azimuthal angles, and the incident solar flux through the upper boundary is $\pi F\mu_0$. After the reflection function is obtained, the plane albedo can be obtained as

$$A_{\rm p} = \frac{1}{\pi}\int_0^1 \mu {\rm d}\mu \int R(\mu,\mu_0,\phi){\rm d}\phi. \tag{10}$$

Figure 3 shows the reflection functions of water droplets internally and externally mixed with BC aerosols at the wavelength of 0.55 μm. The incident light is normal to the layer, i.e., $\mu_0 = 0°$. The results are azimuth-independent. In our computations, the optical thickness is $\tau = 8$. It is clear that the external mixing enhances the absorption compared to the internal mixing when the effective radius of the BC aerosol is smaller than 3 μm, and the reflection function is larger for the external mixing in cases where the effective radius of the BC aerosol is bigger. This is because the contribution of the smaller BC inclusion to the total scattered signal for the internal mixing is small.

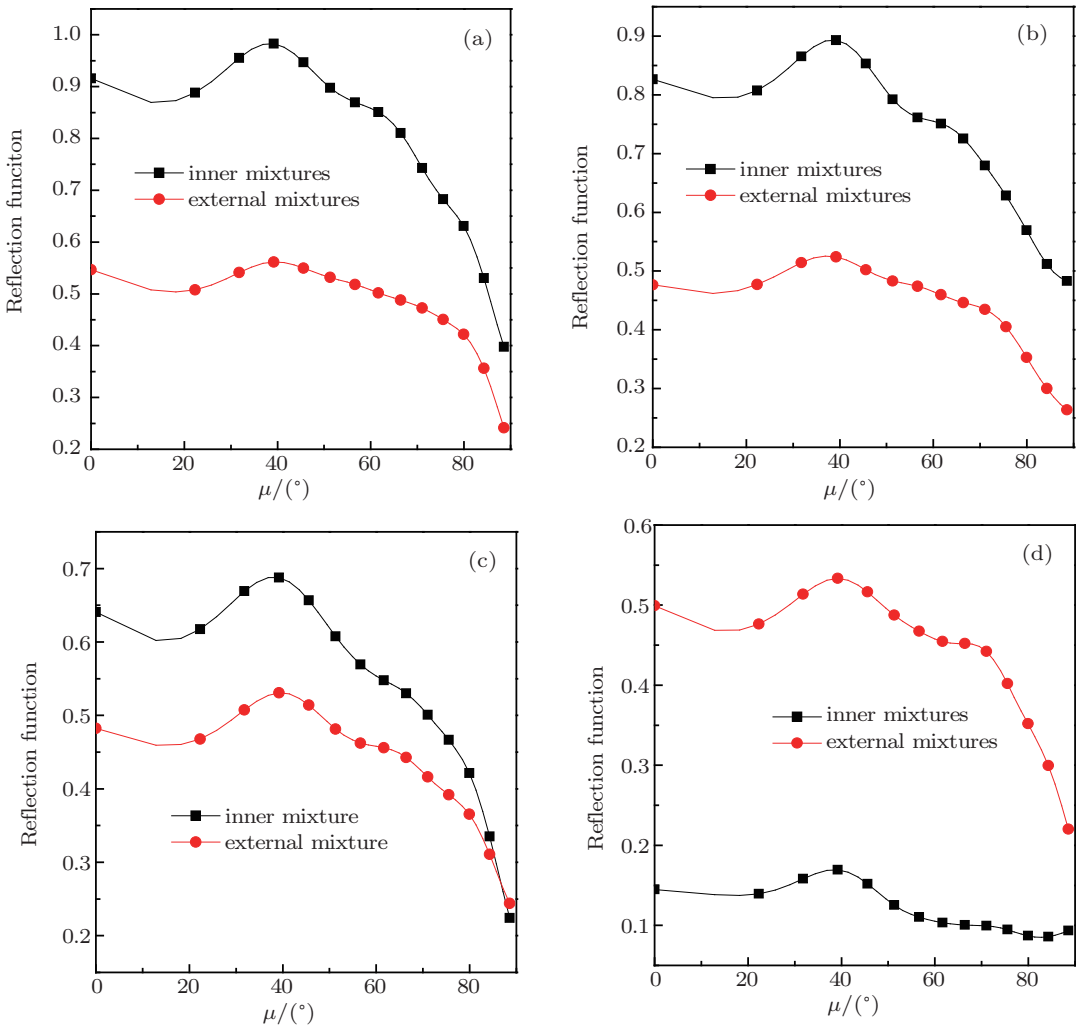


Fig. 3. (colour online) Reflection functions for two types of mixtures of water cloud droplets and BC aerosols. The radii of BC particles in panels (a)–(d) are 0.1 μm, 0.5 μm, 1.0 μm, and 3.0 μm, respectively.

Figure 4 depicts the plane albedo as a function of the cosine of the illumination zenith angle, the optical thickness is $\tau = 8$, and the effective radius of the water cloud droplet equals 6 μm. From the results, we can see that the differences of the plane albedo for external mixtures are small, and the plane albedo slightly in-

creases with the increase of the size of the BC aerosol. However, the plane albedo of the external mixture decreases with the increase of the size of the BC aerosol. As shown in Fig. 4, when the effective radius of the BC aerosol is smaller than 3.0 μm, the plane albedo of the internal mixing model is bigger than that of the external mixing model, when the effective radius of the BC aerosol is bigger than 3.0 μm, the plane albedo of the internal mixing model is smaller than that of the external mixing model. We can conclude that the size of the BC aerosol will affect the absorption and the scattering characteristics of the mixtures.

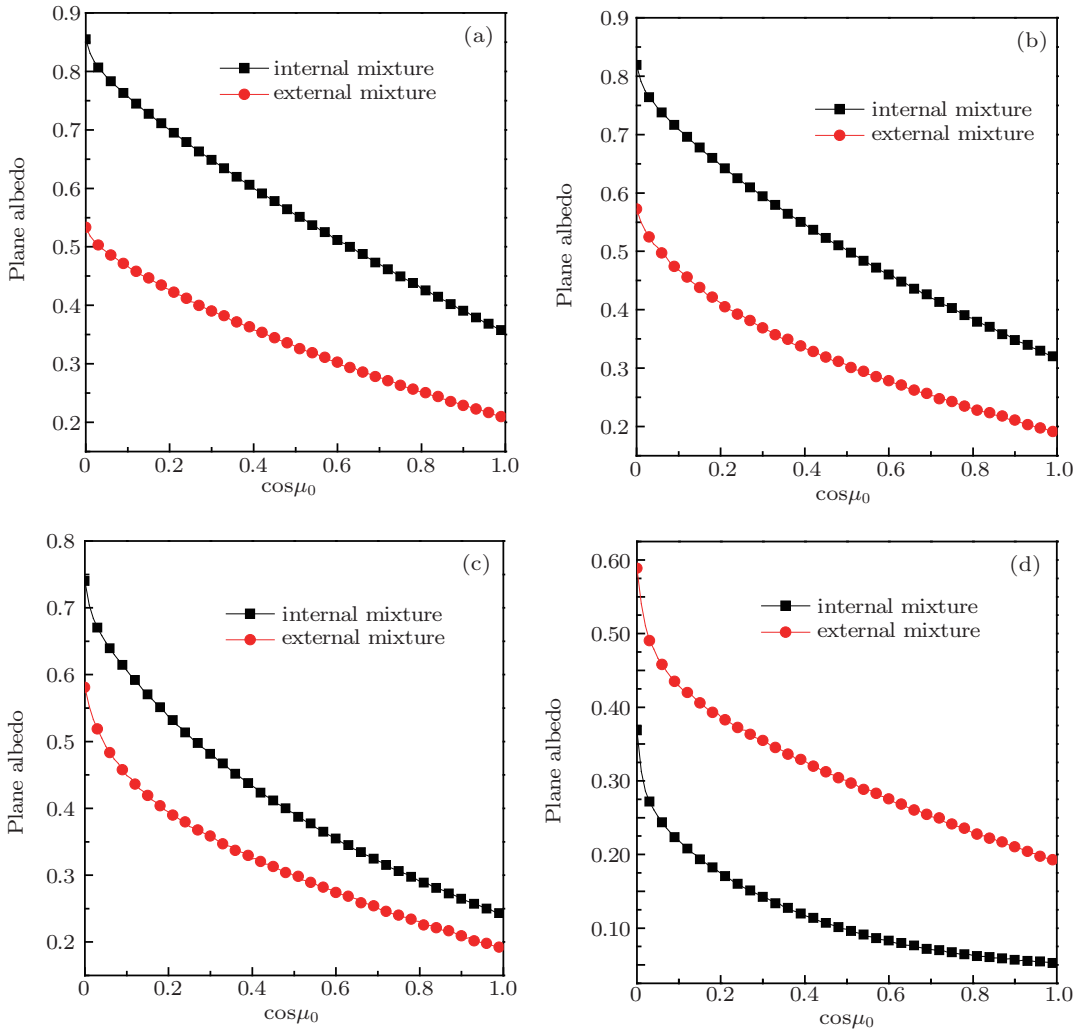


Fig. 4. (colour online) Plane albedos for two types of mixtures of water cloud droplets and BC aerosols. The radii of BC particles in panels (a)–(d) are 0.1 μm, 0.5 μm, 1.0 μm, and 3.0 μm, respectively.

4. Conclusion

In this paper, we study the multiple scattering characteristics of unpolarized light by internal and external BC aerosol mixtures. From the results, we conclude that the size of the BC aerosol will greatly affect the absorption of the mixture. When the size of the BC aerosol is small, the absorption of the external mixture will be larger than that of the internal mixture. If the size of the BC aerosol is big, the conclusion is the opposite. So, if the BC aerosols are mixed with cloud or fog, the sizes of the BC aerosols and the types of the mixtures will be important influencing factors for the absorption, and the mixture model should be chosen properly in remote sensing studies and atmospheric radiation balance computations.

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