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The effect of black carbon on scattering and absorption of solar radiation by cloud droplets

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Abstract

Scattering and absorption characteristics of water cloud droplets containing black carbon (BC) inclusions are calculated at a visible wavelength of 0.55 μm by a combination of ray-tracing and Monte Carlo techniques. In addition, Lorenz–Mie calculations are performed assuming that the same amount of BC particles are mixed with water droplets externally. The results show that it is unlikely under normal conditions that BC aerosols can modify scattering and absorption properties of cloud droplets in any significant way except for geographical locations very close to major sources of BC. The differences in the single-scattering co-albedo and asymmetry parameter between BC-fraction-equivalent internal and external mixtures are negligibly small for normal black carbon loadings, which makes possible the use of the much simpler external mixing model in radiative transfer modeling irrespective of the actual form of mixing. For a fixed amount of BC internally mixed with cloud droplets, the absorption is maximal when the effective radius of the BC inclusions is about 0.05–0.06 μm. Published by Elsevier Science Ltd.

Keywords: Black carbon; Cloud water droplets; Internal and external mixing; Single-scattering co-albedo; Asymmetry parameter

1. Introduction

Black carbon (BC) has long been recognized as an important atmospheric pollutant [1]. It plays a significant role in the absorption of solar radiation by atmospheric aerosols and

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possibly also by clouds. 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 effect of BC impurities on the absorption of solar radiation by cloud water droplets was considered by Danielson et al. [4] using an idealized model with an inner sphere of an absorbing aerosol particle surrounded by a concentric shell of pure water. Chýlek et al. [2] calculated the spectral cloud albedo using an effective medium approximation, which substitutes a heterogeneous internal water–carbon mixture by a fictitious homogeneous material characterized by an effective refractive index. However, the applicability of various effective medium approximations to water droplets containing relatively large soot inclusions remains somewhat uncertain and requires further theoretical and experimental research [5]. Recently, the exact solution for electromagnetic scattering by a host sphere containing one or several non-concentric spherical inclusions has become available (e.g., [6–10]). However, the practical implementations of this solution are still limited in terms of the maximal size parameter of the host and the number and size of inclusions and become very time-consuming when applied to realistic cloud water droplets with multiple randomly positioned inclusions. Therefore, in this paper we address the problem of scattering and absorption of solar radiation by cloud droplets containing BC inclusions using the ray-tracing/Monte Carlo approach developed by Macke et al. [11–13]. This approximate technique assumes that the size of the host particle is much larger than the wavelength of the incident radiation and that the inclusions are randomly and sparsely distributed, and we expect that these conditions are adequately satisfied by an average cloud water droplet (radius $\sim 10\ \mu m$) at visible wavelengths. The results thus obtained are compared with those calculated with the standard Lorenz–Mie formulation and assuming that the same amount of BC particles are mixed with water droplets externally. This comparison is used to derive conclusions about the specific effects of internal mixing on radiative properties of cloud droplets contaminated with soot.

2. Optical properties of black carbon

An important consequence of the presence of BC in the atmosphere is increased absorption of solar radiation [14]. The magnitude of absorption depends on the BC refractive index (especially its imaginary part) and the size, shape, and porosity of BC particles. It also depends on whether the BC particles are mixed with cloud droplets internally or externally and is a function of the average size of the cloud droplets and the exact location of BC inclusions within the droplets. The external and internal mixture models are shown schematically in Fig. 1. Since there is no obvious reason to assume that BC inclusions should have a preferential location inside water droplets, the scattering and absorption properties of contaminated water droplets should be averaged over a random distribution of BC particle locations.

The imaginary part of the BC complex refractive index depends on the original composition of the material that was burned and on the burning process itself. Consequently, there are no universal well-defined optical constants of BC [15]. Following the suggestion by d'Almeida et al. [16], we have adopted for this study the refractive index $1.75 + 0.44i$. This refractive index was also used in recent publications by Chýlek et al. [14,15].

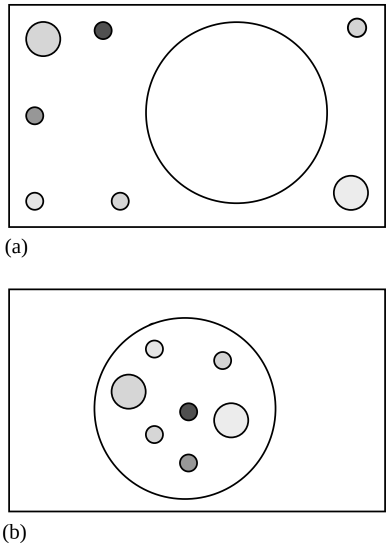

Fig. 1. External (a) and internal (b) mixing of large cloud droplets and smaller aerosol particles.

Table 1
Black carbon concentration in cloud water

Reference	Mean BC (μg kg^{-1})	Range of BC (μg kg^{-1})	Location	Degree of internal mixing (%)
Twohy et al. [29]		23–79	Southern California	
Chýlek et al. [23]	40	10–61	Nova Scotia (Canada)	9
Kou [30]	16	8–41	Nova Scotia (Canada)	6
Bahrmann and Saxena [31]	74.2	20.7–196.9	North Carolina	13
Hallberg et al. [34]				6

3. Observations

In order to evaluate the effect of BC on the radiation balance of the Earth's atmosphere, one needs precise information on the global distribution of BC [17,18]. Although there have been several measurements of the BC concentration in cloud water droplets, the observational data are incomplete, and no clear global or regional picture can be deduced. Rather than rely on a definite set of local observations, Chýlek et al. [15] estimated the lower and upper bounds on the black carbon mixing ratio (by mass) in cloud water for stratus-type clouds to be 2.4×10^{-9} and 8×10^{-6}. A summary of BC concentration measurements is given in Table 1.

4. Ray-tracing/Monte Carlo model

The ray-tracing/Monte Carlo technique is a simple and efficient hybrid method combining ray optics and Monte Carlo radiative transfer concepts. This method permits the treatment of light scattering and absorption by arbitrarily shaped host particles containing small, randomly positioned spherical and nonspherical inclusions and is valid for host particles that are large compared to the wavelength of the incident radiation. The ray-tracing program takes care of individual reflection and refraction events at the outer boundary of the host particle, while the Monte Carlo routine simulates the process of multiple internal scattering by the inclusions. A detailed description of this model is provided in [11–13]. The respective computer code is publicly available at (http://www.ifm.uni-kiel.de/me/research/Projekte/RemSens/SourceCodes/source.html).

5. Model computations

Since BC is found in cloud droplets in measurable amounts, it has been of interest from the climatic standpoint to determine whether the typical BC concentrations could cause significant excess absorption of solar radiation relative to absorption by cloud droplets alone. This absorption would be caused by both increased single-scattering co-albedo $1-\varpi$ and increased asymmetry parameter g of the cloud droplet/BC mixture, where ϖ is the single-scattering albedo. To find the upper bound on the absorption effect of BC, we consider the maximal plausible BC mixing ratio in cloud water 8×10^{-6} (by mass) as estimated by Chýlek et al. [15]. The specific density of BC is largely unknown and depends on the actual burning process. In this paper, we have adopted a low BC specific density of $\rho = 1\ \mathrm{g\,cm^{-3}}$ consistent with our desire to estimate an upper limit on the BC absorption effect. The refractive index of water at a visible wavelength of 0.55 μm and the water specific density are taken to be $1.33+2\times10^{-9}\mathrm{i}$ and $1\ \mathrm{g\,cm^{-3}}$, respectively. Below we will consider separately the cases of internal and external mixing of BC and cloud water.

5.1. Internal mixing

First, we consider a spherical 10 μm-radius water droplet containing a 8×10^{-6} fraction (by mass) of polydisperse, randomly distributed, spherical BC particles. The size distribution of the small BC inclusions was determined on only a very few occasions (e.g., [19–22]). Most of BC particles are in the submicron size range with a typical mode radius between 0.03 and 0.06 μm [23]. Instead of specifying a fixed effective radius r_{eff} of the BC inclusions, we vary it from 0.01 to 0.22 μm, with $r_{\mathrm{eff}} = 0.22$ μm corresponding to one inclusion per host particle under the condition that such a composite particle contains the 8×10^{-6} BC fraction (by mass), to see how the effective radius of these inclusions affects the total scattering and absorption properties of cloud droplets. We assume that the size distribution of the BC particles is given by the standard gamma distribution [24]

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

where $a = r_{\mathrm{eff}}$ and $b = v_{\mathrm{eff}}$. In this study, the effective variance v_{eff} is fixed at 0.1, representing a moderately wide size distribution. The model variable values used in this study are summarized in Table 2. The scattering and absorption properties of BC-contaminated cloud droplets at the wave-

Table 2
Model parameter values used in this study[a]

Mixing ratio by mass	ρ (g cm^{-3})		Refractive index		$r_{\rm eff}$ (μm)		$v_{\rm eff}$		Relative BC refractive index[b]
	BC	W	BC	W	BC	W	BC	W	
8×10^{-6}	1	1	$1.75+0.44$i	$1.33+2\times10^{-9}$i	0.01–0.22	10	0.1	—	$1.316+0.331$i

[a]BC=black carbon; W=water.
[b]The BC refractive index is divided by that of water at 0.55 μm.

length $\lambda=0.55$ μm (corresponding to the maximum in the spectral distribution of the solar radiation) have been calculated by a combination of ray-tracing and Monte Carlo techniques as mentioned above. The single-scattering properties of BC particles have been computed assuming the spherical particle shape and using the Lorenz–Mie code described by Mishchenko et al. [25] and available at (http://www.giss.nasa.gov/∼crmim).

5.2. External mixing

BC particles not only can act as cloud condensation nuclei and be found inside cloud droplets but can also exist outside the droplet as interstitial aerosols. The optical properties of externally mixed cloud droplets and BC aerosols can be well represented by the traditional Lorenz–Mie theory provided that the cloud and aerosol particles are widely separated [26]. We have performed the Lorenz–Mie computation assuming the same mass fraction of BC. Assuming independent scattering, the total single-scattering albedo ϖ and asymmetry parameter g of the mixture are given by [27]

$$\varpi=\frac{C_{\rm sca,W}+NC_{\rm sca,BC}}{C_{\rm ext,W}+NC_{\rm ext,BC}}, \tag{2}$$

$$g=\frac{g_{\rm W}C_{\rm sca,W}+Ng_{\rm BC}C_{\rm sca,BC}}{C_{\rm sca,W}+NC_{\rm sca,BC}}, \tag{3}$$

where $C_{\rm sca}$ and $C_{\rm ext}$ are the scattering and extinction cross sections per particle and N is the number of BC particles per cloud droplet assuming that the BC fraction by mass is fixed at 8×10^{-6}. The subscripts W and BC correspond to water droplets and BC aerosols, respectively.

5.3. Numerical results

Fig. 2 shows the single-scattering co-albedo $1-\varpi$ and asymmetry parameter g of water droplets internally and externally mixed with BC aerosols at the wavelength 0.55 μm as a function of the BC particle effective radius $r_{\rm eff}$ computed for the BC mass fraction 8×10^{-6}. It is clear that internal mixing enhances absorption compared to external mixing, which has already been pointed out in previous studies (see, e.g., [2,29,28]). The absorption is maximized at $r_{\rm eff}\approx0.05$ μm for internal mixing ($1-\varpi\approx4.4\times10^{-4}$) and at $r_{\rm eff}\approx0.08$ μm for external mixing $1-\varpi\approx4.4\times10^{-4}$. Since

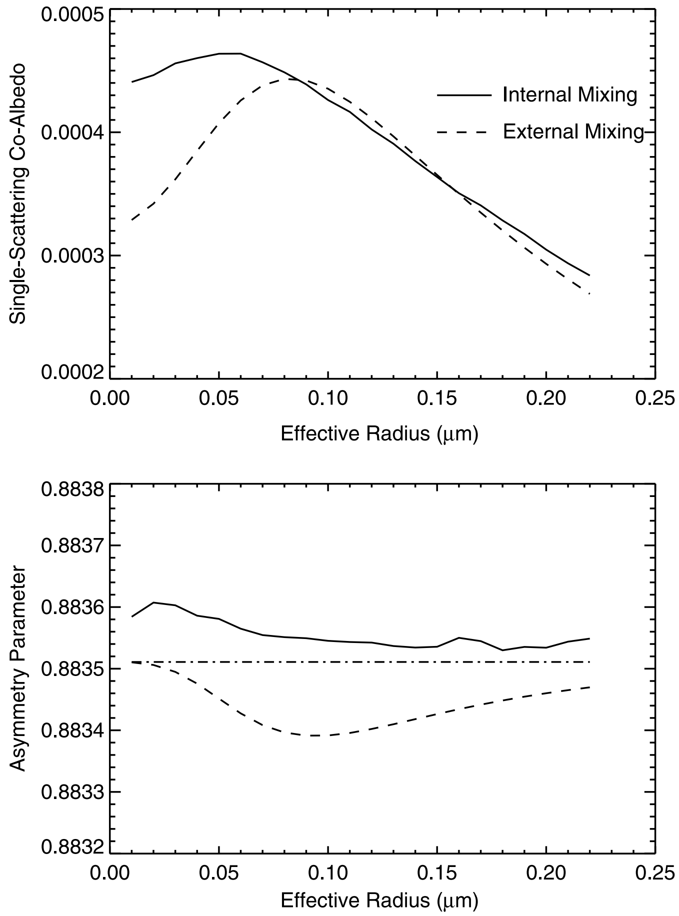

Fig. 2. Single-scattering co-albedo $1 - \varpi$ and asymmetry parameter g for mixtures of cloud droplets and BC particles versus BC particle effective radius at a wavelength of 0.55 μm. The effective variance of the BC particle size distribution is fixed at 0.1, and the BC mass fraction is fixed at 8×10^{-6}. The solid curves show the results for the internal mixture, whereas the dashed curves represents the external mixture. The dash–dotted curve depicts the asymmetry parameter of pure 10 μm-radius water droplets.

the embedded BC particles decrease the ray-tracing part of the total phase function of heterogeneous droplets and thus increase the fractional contribution of the diffraction part, the total asymmetry parameter of BC-contaminated water droplets increases relative to that of pure water droplets [11].

Although we have adopted the upper limit on the BC mass fraction equal to 8×10^{-6} [15], the measured BC fractions in water cloud droplets are usually two orders of magnitude smaller [23, 29–31]. To demonstrate the effect of varying BC amount, Fig. 3 depicts the single-scattering co-albedo and asymmetry parameter as a function of the BC mass fraction at the same wavelength 0.55 μm, with r_{eff} and v_{eff} of BC particles fixed at 0.05 μm and 0.1, respectively. We have chosen the value $r_{\text{eff}} = 0.05$ μm because it maximizes the absorption effect of internally mixed BC particles and because this value appears to be quite realistic according to the measurement results reported in [23,32,33].

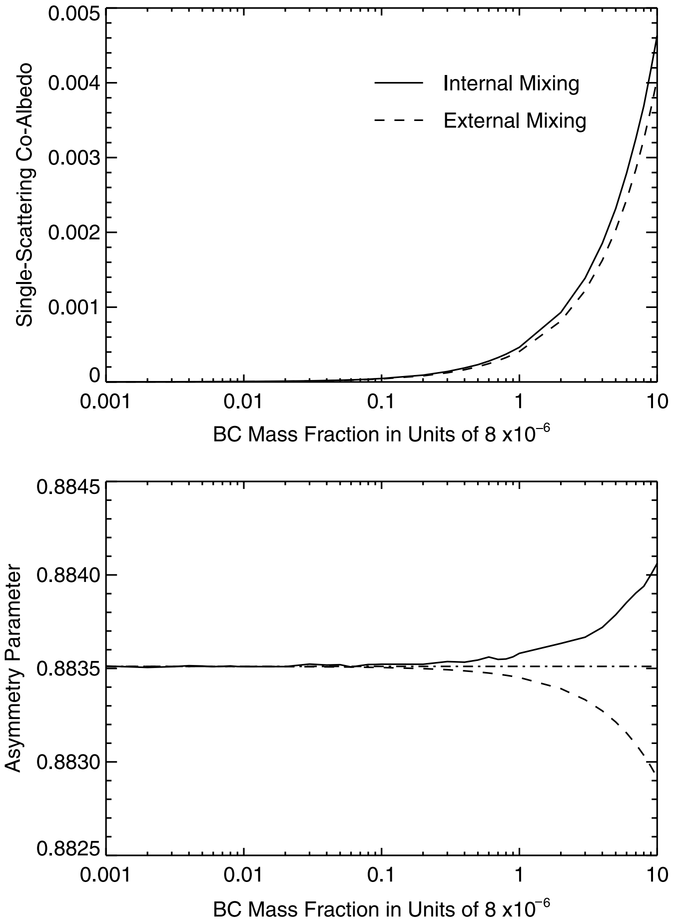

Fig. 3. Single-scattering co-albedo $1-\varpi$ and asymmetry parameter g for mixtures of cloud droplets and BC particles versus BC mass fraction. The BC particle effective radius is 0.05 μm and the effective variance is 0.1. The solid curves show the results for the internal mixture, whereas the dashed curves represent the external mixture. The dash–dotted curve depicts the asymmetry parameter of pure 10 μm-radius water droplets.

Fig. 3 shows that absorption increases approximately linearly with increasing BC mass fraction. However, it is also obvious that the traditionally measured amounts of BC cannot cause significant indirect forcing by strongly increasing cloud absorption.

The absolute difference in the single-scattering co-albedo and asymmetry parameter results between the cases of internal and external mixing is negligible when the BC mass fraction is less than 8×10^{-7}. The latter value is still an order of magnitude larger than those measured for the majority of water clouds [23,29–31]. Fig. 4 shows the relative external/internal differences (in percent) in $1-\varpi$ and g as a function of the BC amount at the wavelength 0.55 μm. The values of r_{eff} and v_{eff} of the BC particles are 0.05 μm and 0.1, respectively. The relative differences in the asymmetry parameter are very small, less than 0.13% in absolute value even when the BC mass fraction is as high as 8×10^{-5}. The relative differences in the single-scattering co-albedo are about -13% when the BC mass fraction is greater than 2.4×10^{-7} and reach -28% when the BC fraction is 2.4×10^{-8}–8×10^{-8}

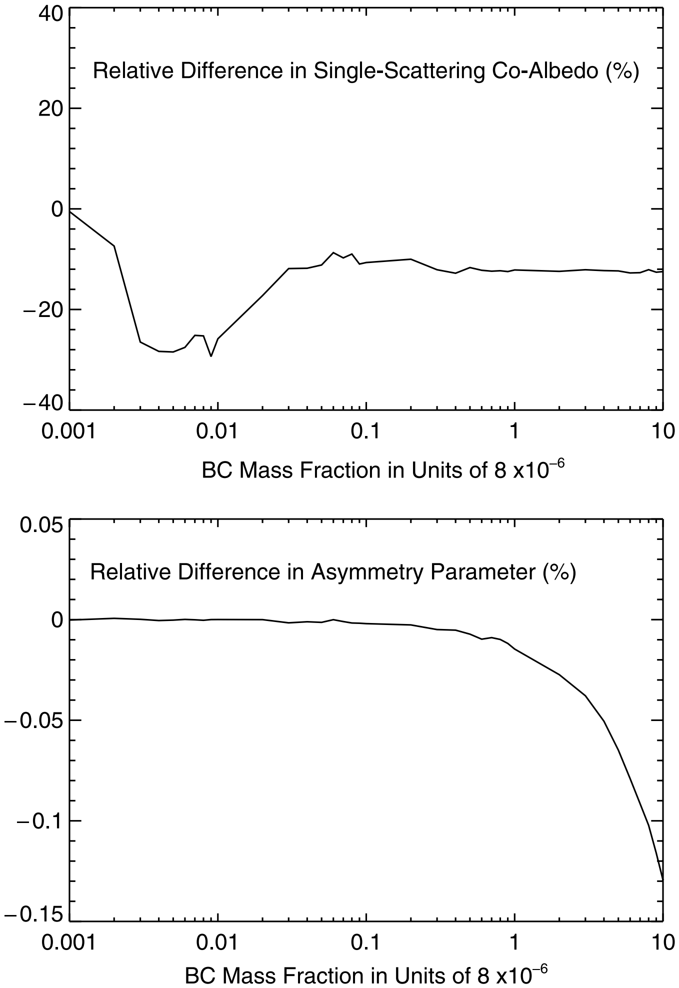

Fig. 4. Relative differences (in %) between the single-scattering co-albedo and asymmetry parameter for external and internal mixtures of cloud droplets and BC particles versus the BC mass fraction. The BC particle effective radius and effective variance are 0.05 μm and 0.1, respectively.

(thus representing a very clean atmosphere). Taking into account that the majority of BC particles remain outside cloud droplets [23,29–31,34] and that the differences in $1-\varpi$ and g between the internal and external mixtures are very small, we conclude that irrespective of the actual form of mixing, one can always use the much simpler external mixing scheme in radiative transfer modeling with great confidence.

6. Discussion

Despite the use of a different approach to compute the optical properties of BC-contaminated cloud droplets, our conclusions are in remarkable agreement with those derived by Chýlek et al. [2]

and Twohy et al. [29]. Chýlek et al. [2] found that a mass fraction of internally mixed graphitic carbon of about 7×10^{-6} is required to increase the single-scattering co-albedo of droplets forming thick stratus clouds from 10^{-7} (pure water) to 10^{-3}. Twohy et al. [29] concluded that BC mass concentrations in excess of 20, 000 μg per 1 kg of cloud water are necessary to reduce the albedo of a cloud with an optical thickness of 30 by 0.03. The observed BC mass concentrations [23,29–31] are usually too low to reduce the cloud albedo in any significant way.

There are a number of remaining uncertainties about the BC optical constants and their variability with type of BC. As a result, the measured refractive indices vary appreciably. The absorption by a small black carbon particle can be shape dependent and may be enhanced by porosity. We have assumed in this study that BC particles are randomly distributed inside water droplets, whereas a preferential location of BC impurities may cause an enhanced absorption. Absorption also depends on the (variable) size of cloud droplets. Although disregarding these uncertainties may result in a biased quantitative estimate of the effect of BC particles on cloud droplet optical properties, it is unlikely to affect our conclusions in a significant way because the observed BC concentrations are so small.

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