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ESTIMATION OF BLACK CARBON EFFECT ON LIGHT SCATTERING AND ABSORPTION BY CLOUD WATER DROPLETS

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1. INTRODUCTION

Black carbon (BC) has long been recognized as an important atmospheric pollutant (Penner et al., 1996). It plays a significant role in the absorption of solar radiation by atmospheric aerosols and possibly also by clouds. Enhanced absorption by black carbon particles imbedded in water droplets could potentially reduce the cloud albedo (Chýlek et al., 1984), thereby causing a significant indirect forcing of climate.

The effect of BC impurities on the absorption of solar radiation by cloud water droplets was considered by Chýlek et al. (1984) using an effective medium approximation. 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. Recently, the exact solution for electromagnetic scattering by a host sphere containing one or several non-concentric spherical inclusions has become available (e.g., Fuller et al., 1994). 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. Therefore, in this study 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. (2000). 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. OBSERVATIONS

In order to evaluate the effect of black carbon on the radiation balance of the Earth's atmosphere, one needs global information on the distribution of BC throughout the atmosphere. 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. (1996) 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.

3. MODEL COMPUTATIONS

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. (1996). Below we will consider separately the cases of internal and external mixing of BC particles and cloud water droplet.

3.1 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 with effective radius r_{eff} varying from 0.01 to 0.22μm. We assume the size distribution of the BC particles is given by the standard gamma distribution. The scattering and absorption properties of BC-contaminated cloud droplets at the wavelength λ = 0.55 μm 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.

3.2 External Mixing

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. We have performed the Lorenz-Mie computation assuming the same mass fraction of BC.

3.3 Numerical Results

Fig. 1 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_{eff} computed for the BC mass fraction 8×10^{-6}. The absorption is maximized at $r_{\mathrm{eff}} \approx$ 0.05μm for internal mixing and at $r_{\mathrm{eff}} \approx$ 0.08 μm for external mixing. The total asymmetry parameter of BC-contaminated water droplets increases relative to that of pure water droplets. To demonstrate the effect of varying BC amount, Fig. 2 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. It is 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. Taking into account that the majority of BC particles remain outside cloud droplets 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.

4. DISCUSSION

There are a number of 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. In this study, we have assumed that BC particles are randomly distributed inside water droplets. However, an enhanced absorption may be caused by a preferential location of BC impurities. Absorption also depends on the (variable) size of cloud droplets. Disregarding these uncertainties may result in a biased estimate of the effect of BC particles on cloud droplet optical properties. However, the observed BC concentrations appear to be far too small to cause a significant indirect climate forcing by substantially increasing cloud absorption.

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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. (1989)		23–79	Southern California	
Chylek et al. (1996)	40	10–61	Nova Scotia (Canada)	9
Kou (1996)	16	8–41	Nova Scotia (Canada)	6
Bahrmann and Saxena (1998)	74.2	20.7–196.9	North Carolina	13
Hallberg et al. (1992)				6

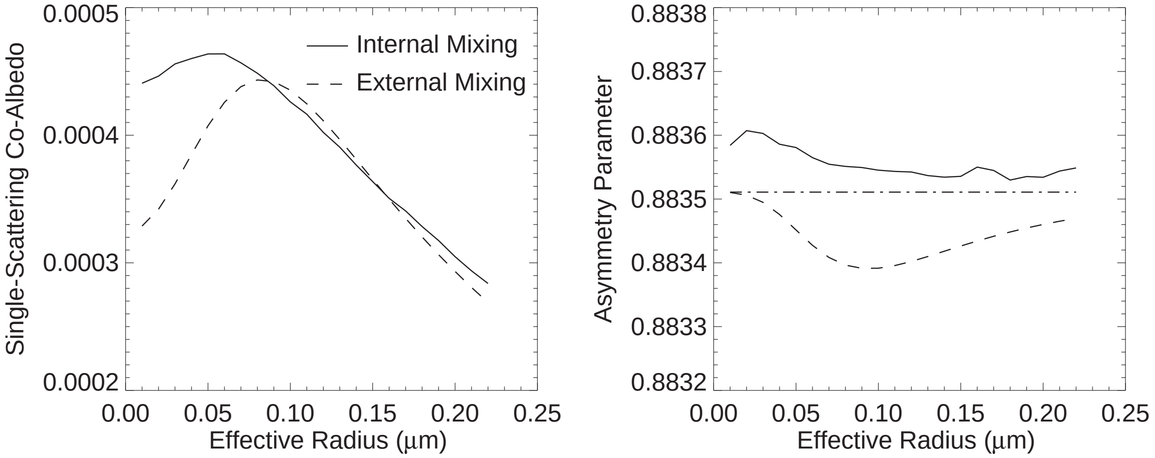

Fig. 1. Single-scattering co-albedo $1-\varpi$ and asymmetry parameter g for mixtures of cloud droplets and BC particles versus BC particle effective radius at a wavelength of 0.55μm. The effective variance of the BC particle size distribution is fixed at 0.1. The dash-dotted curve depicts the asymmetry parameter of pure 10-μm-radius water droplets.

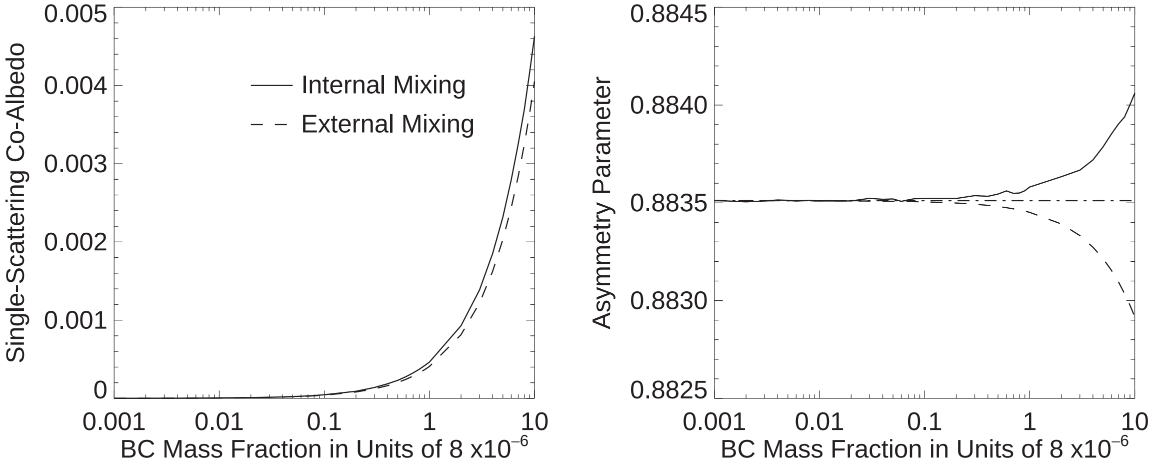

Fig. 2. Single-scattering co-albedo $1-\varpi$ and asymmetry parameter g for mixtures of cloud droplets and BC particles versus BC mass fraction. The BC particle effective radius is 0.05μm and the effective variance is 0.1. The dash-dotted curve depicts the asymmetry parameter of pure 10-μm-radius water droplets.