Modeling Black Carbon Effect on Radiative Properties of Clouds in a Global Climate Model

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Introduction

Carbonaceous aerosols can act as cloud condensation nuclei and thereby change the optical properties of clouds. They are composed of two components, called black carbon (BC) and organic carbon (OC). OC, like sulfate aerosol, is mainly scattering. BC may be distinguished by its resistance to chemical and thermal attack and by its ability to strongly absorb solar radiation; specific absorption coefficients are estimated in the range of 3 m²g to 20 m²g (Liousse et al. 1993). This ability lowers the single-scattering albedo of aerosols, therefore reducing the amount of solar radiation reflected by the aerosols. The presence of BC in cloud may also reduce the cloud albedo. One aspect that is important in the assessment of indirect forcing by carbonaceous aerosols is to include the effect of BC on absorption by clouds. We have modeled this effect in the following manner.

For a collection of cloud droplets with randomly positioned carbon inclusions, the single-scattering albedo of the cloud drops can be estimated using an effective medium approximation (Chylek and Videen 1998). The dielectric constant of the effective medium can be approximated with the Maxwell Garnett mixing rule (Chylek et al. 1988; Bohren and Huffman 1983),

$$m^{2} = m_{0}^{2} \frac{m_{a}^{2} + 2m_{0}^{2} + 2v_{a}(m_{a}^{2} - m_{0}^{2})}{m_{a}^{2} + 2m_{0}^{2} - v_{a}(m_{a}^{2} - m_{0}^{2})}$$

where m = n + ik (k > 0) is the refractive index for the droplet/BC mixture, m_o is the (complex) refractive index of water, m_a is the refractive index of BC, and v_a is the volume fraction of BC within the drop. Chylek et al. (1996) demonstrated that if BC is randomly distributed within the drop, the effective medium approximation is reasonable.

For small volume fractions of BC, using the explicit complex square root to get the components of m from the dielectric constant m^2 can result in unacceptable truncation. We therefore used the iteration counted by j and defined by

$$n_{j} = \sqrt{Re(m^{2}) + k_{j-1}^{2}}$$

 $k_{j} = Im(m^{2}) / (2n_{j})$

until we obtained convergence for k.

Given the refractive index for the droplet with embedded BC particles, we can approximate the single-scattering albedo of a cloud drop using geometric optics. The absorption coefficient of the drop may be approximated by

$$C_{abs} = \frac{4}{3}\pi a^{3} \frac{\alpha}{n} \left[n^{3} - (n^{2} - 1)^{3/2} \right]$$

where $\alpha = 4\pi k/\lambda$, and λ is the wavelength of light (Bohren and Huffman 1983). Since the extinction coefficient is $C_{ext} = 2\pi a^2$, the single-scattering albedo may be approximated by $1 - [C_{abs}/C_{ext}]$. In practice, this approximation only holds if $2a\alpha \ll 1$, which fails to be true for typical BC inclusions in water drops ($\nu_a \approx 10^{-7}$) in the wavelength region $\lambda > 2 \,\mu$ m. Fortunately, in this region absorption by water dominates that by BC, such that a smooth fit between the albedo predicted above and that from pure water provides a sufficiently accurate approximation. By comparison with the single-scattering albedo tables of Stephens (1979), we developed an empirical approximation,

$$C_{abs} = \frac{\frac{4}{3}\pi a^{3} \alpha n^{2} \left[1 - (1 - n^{-2})^{3/2}\right]}{1 + 1.6a\alpha}$$

In the limit of $2a\alpha >> 1$, this expression reduces to a single-scattering albedo given by

$$w_0 = 1 - \frac{5}{12} n^2 \left[1 - (1 - n^{-2})^{3/2} \right]$$

These optical properties were averaged over each wavelength band in the radiation module. Variations of the single-scattering albedo and co-albedo with BC volume fraction as well as wavelength for different sizes of cloud drops are shown in Figure 1.

These approximations have been incorporated into a fully coupled global climate/chemistry model in evaluation of the impact of absorbing particles on aerosol indirect forcing. This model is the revised version of Chuang et al. (1997). The major improvement is the usage of a higher spectral resolution in the solar radiation package. This new version now can calculate the following aerosol compounds: sulfate, dust, sea salt, and carbonaceous aerosols (biomass and fossil fuel organic and black carbon).



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Figure 1. Variations of the single-scattering albedo (left panel) and co-albedo (right panel) of cloud with BC volume fraction and wavelength.

1.0 The (um)

10.0

Figures 2a and 2b present the simulated global distributions of annual average radiative forcing by anthropogenic carbonaceous aerosols through the change of cloud albedo with (Figure 2b) and without (Figure 2a) the BC absorbing effect on clouds. The global average indirect forcing is about the same magnitude for these two simulations (-1.44 W m⁻² with the BC effect versus -1.50 W m⁻² without the BC effect). However, significant differences are observed over regions in Europe, Africa, and East China (see Figure 3). Therefore, simulation without the BC absorbing effect on clouds could overestimate the local indirect forcing due to anthropogenic carbonaceous aerosols by up to 40%. Further investigation on the corresponding changes in cloud liquid water content and cloud lifetime is needed in order to better quantify the effects of aerosols on the radiation budget.



Figure 2. Simulated annual average radiative forcing (W m⁻²) due to the effect of anthropogenic carbonaceous aerosols on cloud albedo. Upper panel: without the BC absorbing effect on clouds; lower panel: with the BC absorbing effect on clouds.



Figure 3. Percentage increase in the magnitude of carbonaceous aerosol indirect forcing without the BC absorbing effect on clouds.

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