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Morphological effects on the radiative properties of soot aerosols in different internally mixing states with sulfate

J. Dong [a], J.M. Zhao [a], L.H. Liu [a,b,*]

[a] School of Energy Science and Engineering, Harbin Institute of Technology, Harbin 150001, People's Republic of China
[b] Department of Physics, Harbin Institute of Technology, Harbin 150001, People's Republic of China

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ABSTRACT

The radiative properties of soot aerosols largely depend on their mixing state and morphology factors. In this paper, we generated soot aggregates in four mixing states with sulfate, including bare soot, partly coated soot, heavily coated soot and soot with inclusion. The number of monomers and fractal dimension of soot were varied in each mixing state while the radius of monomers was fixed at 0.025 μm. Using the discrete dipole approximation method (DDA), we calculated optical parameters relevant for climate forcing simulation at mid-visible wavelength (0.55 μm). Internal mixing results in enhanced absorption, scattering cross sections as well as the single scattering albedo. The enhancement ratio of the absorption is largest for heavily coated soot, which ranges from 1.5 to 1.65 with a soot volume fraction of 0.15 and is larger for soot with larger fractal dimension. The scattering cross section can be dramatically increased by factors larger than 10 when soot is heavily coated. The increasing of both the scattering cross section and the single scattering albedo is larger for soot aggregates with smaller number of monomers and fractal dimension. The asymmetry parameter is insensitive to the fractal dimension for heavily coated soot and soot with inclusion. Two simplified models including the homogeneous sphere model (HS) and the core shell sphere model (CS) were examined using the DDA results as references. The performance of the HS and CS model largely depends on the morphology factors and the mixing state of soot. For bare and partly coated soot, both the HS and CS model can introduce relative errors as large as several tens percent. For heavily coated soot, the HS model predicts the absorption with relative errors within 10%, while it overestimates the absorption with relative errors no larger than 20% for soot with inclusion. The HS model predicts the single scattering albedo and the asymmetry parameter with relative errors no larger than 10% for heavily coated soot and soot with inclusion, which is much better than the prediction by the CS model.

1. Introduction

Aerosols have important effects on the global climate by altering the radiative properties of the atmosphere [1,2]. Among various aerosols, soot aerosol, also called black carbon or light absorbing carbon, is the dominant absorber of visible solar radiation in the atmosphere [3]. Soot which originates from burning of fossil fuel, biofuel, and biomass, frequently gets internally mixed with weak- or non-absorbing aerosols like sulfates, nitrates, organic matter, sea salt and water after being emitted into atmosphere [3–5]. It has been estimated that soot in mixing state may be the second largest contributor to global

* Corresponding author at: School of Energy Science and Engineering, Harbin Institute of Technology, Harbin 150001, China.
Tel.: +86 451 86402237.
E-mail address: lhliu@hit.edu.cn (L.H. Liu).

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Nomenclature

a	radius of the monomer
C_{abs}	absorption cross section
C_{sca}	scattering cross section
d	dipole spacing
D_f	fractal dimension
f	volume fraction
g	the asymmetry parameter
k_0	fractal prefactor
k	wave number
l	distance
m	refractive index
n	number of aggregate realizations
N_s	number of monomers
p	the potential of an exterior dipole to be added to soot aggregate
R	ratio of integrated parameters of internally mixed soot to those of bare soot
R_g	radius of gyration
X	number of dipoles

Greek symbols

ω	the single scattering albedo
δ	relative error
ε	permittivity
λ	wavelength

warming after carbon dioxide [6,7], whereas others argued that its absorption may be overestimated by climate models [8]. There are still many uncertainties about its effects on the climate. One of the uncertainties concerns the radiative properties of soot aerosols, which is determined by the morphology, the mixing state, and the refractive indices of soot and the coating materials.

Soot aerosols frequently exist in the form of clusters of small, nearly spherical monomers after being emitted. The morphology of soot aggregate can be represented by a fractal cluster described by the following statistical scaling law [9]:

$$N_s = k_0 \left(\frac{R_g}{a}\right)^{D_f} \tag{1}$$

where N_s is the number of monomers in the cluster, D_f is the fractal dimension, k_0 is the fractal prefactor, a is the radius of the monomers, and R_g is the radius of gyration which is a measure of the overall cluster radius and is defined by

$$R_g^2 = \frac{1}{N_s} \sum_{i=1}^{N_s} l_i^2 \tag{2}$$

where l_i is the distance of the ith monomer to the cluster's center of mass. Fractal dimension of freshly emitted soot aggregates is around 1.8 [3], whereas it is found to be larger by use of 3-D electron tomography measurements [10]. With the aging of soot in atmosphere, the lacy soot aggregates may collapse into more compact ones, which can be expressed by the increasing of the fractal dimension. During this process, soot aggregates can frequently get internally mixed with weak- or non-absorbing aerosols. Internally mixed soot aerosols are mainly formed by the condensation of organic matter, sulfate, etc., onto their surfaces [11]. These condensed materials, featuring high viscosity, subsequently fill the voids of the aggregate branches and prevent it from compacting [11]. Therefore, soot aerosols usually keep their fractal structure after they are internally mixed with other materials. Fig. 1 demonstrates electron images of some soot aerosol particles collected by China et al. [12], who classified soot aerosols into four categories: (a) bare soot which is not coated or very thinly coated, (b) partly coated soot where soot voids are filled by coating materials but the soot is not completely engulfed, (c) embedded or heavily coated soot where some monomers are still evidently visible, and (d) soot with inclusions where soot is mixed with but not uniformly coated by other material.

The radiative properties of soot aerosols largely depend on their morphology and mixing state. However, most climate models apply highly simplified models like a homogeneous sphere model or a core shell sphere model to approximate the radiative properties of soot aerosols. The homogeneous sphere model is usually applied with an effective medium theory. Yet the simplified models may introduce significant errors [13] and their usefulness needs to be validated by comparing their radiative properties to those of more realistic ones.

Many researches have been performed to study the radiative properties of soot aerosols considering their fractal morphology and various mixing states. The radiative properties of bare soot with different morphological factors have been investigated extensively [14–18] by the numerically accurate superposition T-matrix method (STM) [19,20] or the generalized Mie-solution method (GMM) [21]. It was shown that the fractal morphology of soot clusters has a profound impact on their radiative properties and that the commonly used homogenous sphere approximation may introduce large errors. Several studies also applied STM or GMM to investigate the radiative properties of internally mixed soot aerosols. Liu et al. [22] studied the effect of water coating on the single scattering properties of soot aggregates. Mishchenko et al. [23] calculated the scattering and absorption properties of micrometer-sized water droplets contaminated by black carbon. Cheng et al. [24] studied the morphological effects on the optical properties of internally mixed light absorbing carbon aerosols with different aging status. The researches above all found an enhancement in the absorption of sunlight as a result of internal mixing and that morphological factors can result in a dramatic change in the optical properties. However, STM and GMM have an obvious limitation that it can only deal with scattering particles composed of spheres and the spheres cannot overlap with each other. The discrete dipole approximation (DDA) method [25,26], on the other hand, is flexible in dealing with scattering particles of arbitrary geometry and composition. By using DDA, Kahnert et al. [27,28] investigated the optical properties