Journal of Quantitative Spectroscopy & Radiative Transfer 187 (2017) 10–19

Contents lists available at ScienceDirect

Journal of Quantitative Spectroscopy & Radiative Transfer

journal homepage: www.elsevier.com/locate/jqsrt

Impact of morphology on the radiative properties of fractal soot aggregates

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ARTICLE INFO

Article history:
Received 13 December 2015
Received in revised form
7 September 2016
Accepted 9 September 2016
Available online 16 September 2016

Keywords:
Overlapping
Aggregate expansion
Fractal soot aggregates
Aerosol optical properties
DDA

ABSTRACT

The impact of morphology on the radiative properties of fractal soot aggregates was investigated using the discrete dipole approximation (DDA). The optical properties of four different types of aggregates of freshly emitted soot with a fractal dimension D_f= 1.65 and a fractal pre-factor k_f= 1.76 were calculated. The four types of aggregates investigated are formed by uniform primary particles in point-touch, by uniform but overlapping primary particles, by uniform but enlarged primary particles in point-touch, and formed by point-touch and polydisperse primary particles. The radiative properties of aggregates consisting of N=20, 56 and 103 primary particles were numerically evaluated for a given refractive index at 0.532 and 1.064 µm. The radiative properties of soot aggregates vary strongly with the volume equivalent radius a_{eff} and wavelength. The accuracy of DDA was evaluated in the first and fourth cases against the generalized multi-sphere Mie (GMM) solution in terms of the vertical–vertical differential scattering cross section (C_{vv}). The model predicted the average relative deviations from the base case to be within 15–25% for C_{vv}, depending on the number of particles for the aggregate. The scattering cross sections are only slightly affected by the overlapping but more significantly influenced by primary particle polydispersity. It was also found that the enlargement of primary particles by 20% has a strong effect on soot aggregate radiative properties.

1. Introduction

When soot particles in the form of fractal-like aggregates are emitted into the atmosphere, they become efficient light absorber and scatterers. Thus, they can influence the environment as a major anthropogenic agent with short-lived climate forcing effects [1]. The chemical composition of soot depends on the type of fuel and mode of combustion. The morphology of soot aerosols consists of the particle mean radius (a), fractal pre-factor (k_f), fractal dimension (D_f), radius of gyration (R_g) and the number of particles (N). The fractal pre-factor and fractal dimension are two important parameters for determining the structure of a fractal aggregate [2–5].The fractal dimension is in the range of 1.6–1.9 for over-ventilated or oxygen-rich combustion, and in the range of 2.1–2.3 for non-flaming or fuel-rich combustion [6]. Freshly generated soot aggregates are generally open structured with $D_f < 2$; aged soot aggregates are internally mixed with $D_f > 2$, as defined by Bond and Bergstrom [2]. In other words, when the fractal dimension increases, the aggregates change from chain-like (or lacy) to more compact structures. Another important parameter that affects the radiative properties of soot aggregates is the refractive index. Bond and Bergstrom investigated the wavelength-dependent refractive indices and radii of the primary particles [2]. They evaluated a refractive index range from 1.75+0.435i to 2+i at the

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http://dx.doi.org/10.1016/j.jqsrt.2016.09.005
0022-4073/

visible spectrum; the particle radii were identified to be between 10–25 nm.

Several methods are widely used for the evaluation of the radiative properties of fractal soot aggregates, such as the Rayleigh–Debye–Gans (RDG) approximation, the generalized multi-particle Mie (GMM) method, the discrete dipole approximation (DDA) and the *T*-Matrix method. The RDG approximation [7,8] is commonly applied and is used to assess the angular distributions of scattered light and mass scattering, mass extinction and mass absorption coefficients for fractal aggregates. The *T*-matrix method with Waterman's null-field method [9] was applied for homogenous axisymmetric and non-axisymmetric shapes. In addition, the superposition *T*-matrix method has been applied to study the radiative properties of various aggregate particles [10]. The superposition *T*-matrix method solves Maxwell's equations to calculate the total field scattered by soot aggregates which is expressed as a superposition of individual fields scattered from each sphere. The GMM method is based on the framework of Mie theory for a particle, and additional theorems for spherical vector wave functions [11,12]. According to Xu and Khlebtsov [13], the theoretical frameworks for the *T*-matrix and GMM methods are very similar; however, the latter has some advantages over the former.

Soot particles generated in hydrocarbon flames were analyzed in an experimental study using the fast Fourier transform (FFT) inversion technique by Di Stasio [14]. In that study, the author focused on the depolarization ratio, scattering angles and intensity of light scattering for small aggregates. The *T*-matrix method was used for calculation of the radiative properties of soot and soot-containing aggregate particles with refractive indices of $m=1.75+0.435i$ and $m=2+i$ at wavelength $\lambda=628$ nm in [3–5]. The superposition *T*-matrix method was applied to aggregates based on particle radius ($a=25$ nm) and refractive index ($m=1.77+0.63i$) by Kahnert [15], who concluded that the interaction among particles enhances the absorption cross sections. He also found that the absorption properties of soot aggregates with particle radii smaller than approximately 40 nm are not sensitive to variation of the radius of the particles. In addition, Kahnert [15] demonstrated that the fractal dimension (D_f) determines how much mass in the aggregate interacts with the electromagnetic field.

Liu and Snelling [16] used the RDG approximation to analyze the radiative properties of soot aggregates, and compared the results with the GMM method. The absorption properties of soot aggregates with 30 nm diameter ($D_f=1.78$, $k_f=2.3$ and $m=1.60+i0.6$) were investigated at 532 and 1064 nm. The effects of the aggregate absorption model on soot temperature in laser-induced incandescence (LII) modeling were investigated by Liu and Smallwood [17]. For this purpose, the absorption cross sections were analyzed using three aggregate absorption models; the RDG approximation, the electrostatics approximation (ESA) and the GMM method. They concluded that the RDG approximation underestimated the aggregate absorption cross sections, and that the ESA was somewhat better than the RDG approximation. In addition, the effects of aggregate morphology on the absorption ability of soot aggregates were studied using the GMM method in their study. Polarization curves were experimentally obtained for four soot aggregates and compared with black carbon samples at $\lambda=0.632$ µm by Francis et al. [18]. Two of their four soot aggregates were produced from combustion in over-ventilated conditions; and the other two were in under-ventilated conditions. According to the results from their measurements, polarization was found to increase with the diameter of the soot aggregates. In addition, they confirmed that soot aggregates with high flow ventilation rates of 450 m^3/h have higher polarization values than those with low flow ventilation rates of 100 and 50 m^3/h.

A recent experimental study determined the radiative properties of uncoated and coated soot aggregates generated in a Santoro-style diffusion burner [19]. In that study, Bueno et al. performed size-dependent absorption measurements using a laser at 0.405 µm and compared the results to those from the Lorenz–Mie theory of scattering and absorption. The absorption cross section was found to be dependent on the aggregated morphology, although the optical properties of the fresh soot aggregates varied with time. Soot aggregates with no coating and five different lightly coated aggregates were analyzed using the DDA [20]. It was found that the absorption and scattering properties of aggregates increase with increasing coating thickness. Absorption cross sections of soot aggregates in the infrared region were studied by Prasanna et al. [21], in which the DDA, multi-sphere *T*-matrix (MSTM) and electrostatic approaches were used to examine the effects of various parameters on the absorption of soot. They investigated the effects of different fractal parameters on absorption cross sections at different refractive indices by assuming monodisperse spheres with point contact. Additionally, a model for the absorption cross section, which includes all independent variables, was defined. The MSTM method was found to be suitable for such calculations. Li et al. [22] applied the DDA to single-smoke aggregates in the diameter range of 0.1–0.4 µm, and calculated the light transmitted flux from the radiative transfer equation. They investigated the effects of smoke diameter, smoke density and incident wavelength on radiative heat transfer in a smoke layer, concluding that the smoke diameter is the dominant parameter compared to wavelength, density and transmitted flux. Scarnato et al. studied the optical properties of uncoated and coated black carbon soot aggregates with $m=1.95-i0.79$ at a wavelength range of 0.2–1.0 µm using the DDA [23]. In their study, five aggregates are formed by primary particles of 40 nm diameter and another one by primary particles of 30 nm diameter. They tested two kinds of coated aggregate. The first were black carbon aggregates with partial surface contact on NaCl; the other were black carbon aggregates encapsulated in NaCl. The authors presented their numerical results in terms of the mass absorption coefficient and the single scattering albedo for all the aggregates. All the studies discussed above were conducted for soot aggregates without any overlapping between primary particles.

The depolarization ratios and the optical cross sections of soot aerosols produced from methane, ethylene and