Journal of Quantitative Spectroscopy & Radiative Transfer 205 (2018) 184–195

Contents lists available at ScienceDirect

Journal of Quantitative Spectroscopy & Radiative Transfer

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

A model study of aggregates composed of spherical soot monomers with an acentric carbon shell

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ARTICLE INFO

Article history:
Received 12 May 2017
Revised 26 October 2017
Accepted 28 October 2017
Available online 28 October 2017

Keywords:
Coated soot particles
Acentric core–shell monomers model
Optical properties

ABSTRACT

Influences of morphology on the optical properties of soot particles have gained increasing attentions. However, studies on the effect of the way primary particles are coated on the optical properties is few. Aimed to understand how the primary particles are coated affect the optical properties of soot particles, the coated soot particle was simulated using the acentric core–shell monomers model (ACM), which was generated by randomly moving the cores of concentric core–shell monomers (CCM) model. Single scattering properties of the CCM model with identical fractal parameters were calculated 50 times at first to evaluate the optical diversities of different realizations of fractal aggregates with identical parameters. The results show that optical diversities of different realizations for fractal aggregates with identical parameters cannot be eliminated by averaging over ten random realizations. To preserve the fractal characteristics, 10 realizations of each model were generated based on the identical 10 parent fractal aggregates, and then the results were averaged over each 10 realizations, respectively. The single scattering properties of all models were calculated using the numerically exact multiple-sphere T-matrix (MSTM) method. It is found that the single scattering properties of randomly coated soot particles calculated using the ACM model are extremely close to those using CCM model and homogeneous aggregate (HA) model using Maxwell–Garnett effective medium theory. Our results are different from previous studies. The reason may be that the differences in previous studies were caused by fractal characteristics but not models. Our findings indicate that how the individual primary particles are coated has little effect on the single scattering properties of soot particles with acentric core–shell monomers. This work provides a suggestion for scattering model simplification and model selection.

1. Introduction

Understanding of optical properties of soot particles is extremely important for atmospheric remote sensing, climate change studies and fire detection, etc. However, optical properties of soot particles are still uncertain due to the incomplete understanding of the optical properties of soot aerosols with complex morphologies and chemical compositions [1].

Freshly emitted soot particles generally present fractal structure characteristics, but trend to be coated with non-absorbing aerosols under the influences of the atmospheric aging, which leads to more complex morphologies. Therefore, the research on random non-absorbing coatings is very important for the understanding of scattering properties of soot particles. Soot particles are classified into four categories by China et al. [2]: (1) embedding; (2) partly coating; (3) bare; and (4) internal mixing. This work mainly targets on the partly coated soot particles which is an important component of soot aerosols. The single core–shell (SC) model using the core–mantle Mie theory was commonly applied in climate model to simplify the calculations [3–5]. But the calculation results can lead to large deviation compared with measurements and more realistic models [6–15]. Although many studies have concerned on the optical properties of soot particles with mixing states [7,8,10,16], the effect of the way soot particles are coated on the optical properties is still unclear and few studies investigate how the primary particles are coated influences the optical properties of soot particles.

Soot particles tend to be randomly coated with non-absorbing materials in atmospheric aging. Dong et al. [17] simulated the random coatings by adding dipoles. In that work, whether a discrete dipole is added was dominated by the sum of the inverse square of the distance between the point and every center of monomer sphere. A morphology of coatings submerging the soot monomers can be simulated in this way. However, soot particles may be randomly coated with a thin organic materials layer in some cases. Another alternative model is also commonly applied for calculating

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https://doi.org/10.1016/j.jqsrt.2017.10.024

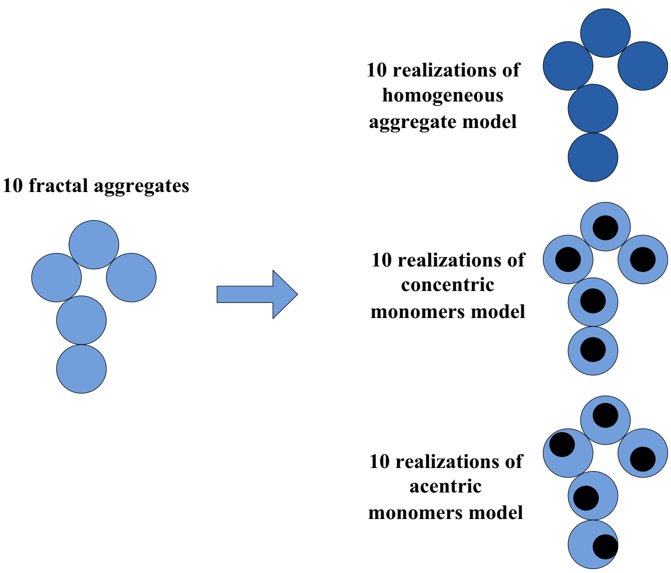

Fig. 1 . The way to preserve fractal nature of different models.

the optical properties of soot particles, which assumes that soot monomers are contacted in point with each other, and every soot monomer is coated by non-absorbing materials with constant thickness. This leads to the intersecting of the coatings and uncertainty of soot volume fractions [18–20]. Wu et al. [18] added dipoles at edges of the whole structure one by one. In that work, a realistic morphology of thinly coated soot particles was simulated. However, the coatings are completely random and not easy to be controlled, so it is hard to understand how primary particles are coated influences the optical properties of soot particles using such model. Another important problem is that the former three models are generally calculated with the discrete dipole approximation (DDA) method [21], and in order to gain accurate calculation results, the dipole number should be considerably large, which leads to relatively higher computational time cost. In order to reduce computation cost, another model was developed, which is composed of concentric core–shell monomers which are contacted in point with each other [20,22,23]. Their optical properties were calculated using the numerically exact multiple-sphere T-matrix (MSTM) method [24,25], which demands for less computational time cost than DDA method [26]. However, the individual primary particles are randomly coated in reality, namely are not the concentric core–shell appearance. Although in reality the soot particles have complex morphology, the model with real shape is very computationally expensive, so it is reasonable to conduct some simplifications. The closed-shell morphology reflect the real morphology to a great extent. It can represents the process of accumulation of a refractory material around individual soot monomers, and is an example of where coating material not only covers the outer layers of soot aggregates but also fills the internal voids among primary spherules. In addition, it is more likely to be put into practice than other models due to the high computation efficiency, so we conduct a detailed study on this morphology. To reflect the randomness of coatings, we proposed the acentric core–shell monomers model, which generated by randomly moving the cores of concentric core–shell monomers model. The aim of this paper is to investigate how the primary particles are coated affects the optical properties. The way primary particles are coated was simulated by changing the movement patterns of soot cores.

The single scattering properties calculated using ACM model were compared with those using SC model, HA model and CCM model. Previous studies [18,23] concluded that the results calculated using HA model, CCM model and more realistic model were different. However, the differences can be also caused by fractal nature but not models. In order to eliminate optical diversities caused by fractal characteristics, the structures of all models were generated based on 10 identical parent fractal aggregates in this study, so all models shared the same structures. In this work, the soot monomer was fixed to be 0.02 um, so the total monomer radius varies with the soot fractions. We generated soot particles with different soot volume fractions by scaling the 10 aggregates generated at first to maintain the fractal characteristics identical.

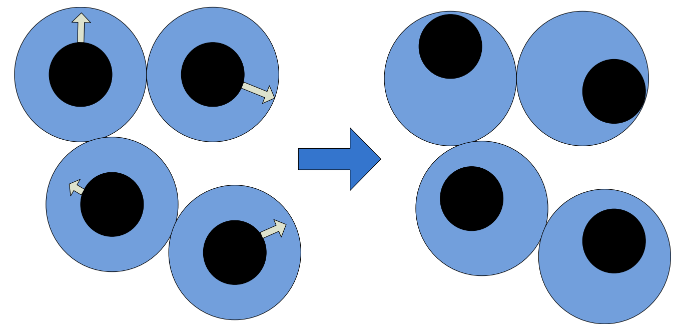

Fig. 2. The generation of acentric core–shell monomers model.

The generation of models for randomly coated soot particles, the theory and method for calculating the optical properties of the coated soot particle are introduced in Section 2. The random-orientation averaged results of different models are discussed in Section 3. Conclusions are given in Section 4.

2. Models for soot particles coated with random non-absorbing aerosols

In this work, soot particles were simulated using the ACM model. The SC model, HA model, and CCM model were used for comparison. The volumes are same for all models, which is the sums of soot monomers and coatings. The monomer radius of HA model, CCM model and ACM are identical. Differently, the HA model is composed of homogeneous monomers with the single refractive index, while CCM model and ACM model are constructed as two spheres with different refractive indices for the highly absorbing core and the weakly absorbing shell. The difference between the CCM model and the ACM model is that the positions of core centers of acentric core–shell monomers model are random.

2.1. Generations of models for soot particles coated with random non-absorbing aerosols

Diffusion limited aggregation (DLA) algorithm [27] is often applied to generate fractal structures. In this work, the fractal aggregates were generated by a tunable DLA code developed by Wozniak et al. [28]. It preserves fractal parameters at each step of the aggregation compared to the ordinary DLA code. This allows to avoid generation of multi-fractal aggregates [29]. The construction of the fractal structure satisfies the well-known fractal laws [3]:

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

$${R_g}^2 = \frac{1}{N}\sum_{i=1}^{N_s} {l_i}^2 \quad (2)$$

where N_s is the number of the monomers in the cluster, R is the mean radius of the monomers, k_0 is the fractal prefactor, D_f is the