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Performance of the discrete dipole approximation for optical properties of black carbon aggregates

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ABSTRACT

The optical properties of black carbon (BC) are fundamental for radiative transfer and remote sensing. BC geometry is successfully represented by an idealized model named "fractal aggregate", and numerous methods are available and widely used to simulate the corresponding optical properties. This study systematically evaluates the performance of the discrete dipole approximation (DDA) for optical simulations of BC aggregates. The Multiple Sphere T-Matrix (MSTM) results are used as references for accuracy evaluation. The differences between the DDA and MSTM can be controlled to be less than 3% by using dipole size much smaller than the monomer size, and the DDA efficiency is sensitive to aggregate structures, e.g. lacy or compact. We find that shape representation for small-sized monomers during DDA discretization leads significant errors, i.e., up to 10%, and relatively large refractive index of BC also affects the DDA accuracy. However, the MSTM treats the BC monomers as perfect spheres without overlapping, and the imperfect structure that is implicitly introduced in the DDA simulations due to the spatial discretization may be a better representation of realistic BC particles. Moreover, the accuracy and efficiency of the DDA can be improved by defining dipoles on the particle boundary to have refractive indices given by the effective medium approximation (EMA). This leads to the adequate shape representation even using larger dipole sizes, and results in the DDA accuracy comparable to that of the reference MSTM solution.

1. Introduction

Black carbon (BC), produced by incomplete combustion, is one of the most significant contributors to global warming due to its strong absorption of solar radiation [1]. It plays an important role in the radiative forcing at regional and global scales, and may enhance the occurrence of extreme haze pollution episodes by inducing heating in the planetary boundary layer [2]. Thus, the optical properties of BC aerosol are fundamental for a better understanding of its radiative effects and remote sensing studies.

BC particles normally exist as aggregates of spherical monomers [3,4], and fractal geometry is used for describing BC complex morphologies [5–7]. Mathematically, the parameters that describe fractal aggregates are related by [5]:

$$N = k_f \left(\frac{R_g}{a}\right)^{D_f}. \quad (1)$$

Here, N is the number of monomers in an aggregate, and a is the monomer radius. The gyration radius R_g quantifies the spreads of the monomers. The fractal prefactor k_f and fractal dimension D_f are two important parameters describing the compactness of an aggregate. Relatively compact aggregates with a fixed N and a have smaller R_g, so require a larger D_f or k_f to satisfy the relationship in Eq. (1).

With geometries based on the fractal aggregate rigorously defined, accurate simulations of BC optical properties become possible. Numerous numerical models are developed and applied to account for the optical properties of BC aggregates [8–10]. Among the existing methods, the Multiple Sphere T-Matrix method (MSTM) [8,11] and the Generalized Multi-particle Mie (GMM) method [12,13] are specially designed for scattering properties of multiple spheres. Both the MSTM and GMM can only consider aggre-

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Table 1
Comparison of the previous studies using the DDA to calculate optical properties of BC aggregates.

Reference	Refractive index[a]	Size			dpl[c]	RE[d]
		N	Monomer size[b]	Coating size[b]		
Kahnert et al. [22]	1.76 + 0.63i	4–1600	0.15	1.0–6.0	~ 40	<3%
Prasanna et al. [23]	2.5 + 1.5i	2	0.01	0	3000–6000	3%–8%
Prasanna et al. [23]	2.2 + 1.3i	20–100	0.024	0	~1000	~20%
Wu et al. [24]	1.76 + 0.57i	200	0.17	~2.8	~ 150–800	8%–1%
Moteki [25]	1.75 + 0.44i	1	0.05–0.5	0	Not given	<20%
Liu et al. [26]	1.73 + 0.59i	200	0.17	1.0	80	~3%
Doner et al. [27]	1.75 + 0.63i	200	0.12–0.29	0	110–255	8%
Tazaki & Tanaka [28]	3.4 + 0.05i	64	0.0006–0.6	0	~50–50,000	33%–1%

[a] Refractive index of black carbon aerosols when coating material is also present.
[b] The size is given as the size parameter.
[c] Number of dipoles per wavelength (see text).
[d] The relative errors of the integral scattering properties (extinction or absorption) by comparing with MSTM.

gates of perfect spheres without overlapping. Meanwhile, more general methods that are applicable for particles with arbitrary geometries can also calculate BC optical properties, such as the discrete dipole approximation (DDA) [14,15] and finite difference time domain method (FDTD) [16]. The geometric-optics method has also been extended and improved to account for optical properties of aggregates by considering surface wave [17,18]. A well-chosen method may not only save significant computational resources but also provide more robust and accurate results, whereas an inappropriate method may draw misleading or wrong conclusions. Therefore, a complete and rigorous understanding on the performance of different numerical methods themselves is needed.

Among those numerical models, the DDA solves the electromagnetic integral equation in the frequency domain, and has been extensively applied to investigate aggregate optical properties, especially those with irregular or inhomogeneous geometries [19–21]. In the framework of the DDA, a particle is discretized into small sub-volumes (namely dipoles), and interactions derived from the aforementioned integral equation can be approximated by those among point dipoles. Thus, the DDA is fully flexible on particle geometries. Considering that the MSTM or GMM can only be applied to fractal aggregates of perfect spheres without overlapping [11–12], the DDA is widely used to investigate the effects of more realistic monomer irregularity, e.g., overlapping, necking, and coating, on BC optical properties [19–28]. During the application of the DDA, those studies either briefly evaluate the DDA for BC aggregate applications [22–28], or directly carry out the DDA simulations for their particular applications without validation [29–31].

Table 1 summarizes some of the previous studies that include discussions on the DDA performance [22–28]. Most of these studies use the DDA for their particular applications related to aggregate optical properties, and the DDA is evaluated only briefly. As another rigorous solution of the Maxwell equations, the MSTM solves equations in the multiple spherical boundary domain, so it is often used as the reference method for the evaluations of other methods. Table 1 lists some key parameters used for the DDA, i.e., refractive index, particle sizes, and spatial resolution used, and the last column is the relative errors (REs) of the DDA results (mostly for the cross sections), which range between over 30% to almost zero. The parameter "dpl", namely dipoles per lambda (wavelength), is widely used in the DDA literature to describe the discretization level. While certain rules-of-thumb exist for satisfactory accuracy, e.g. dpl $\approx 10|m|$, where m is the complex refractive index, they are largely misleading when the particle or some of its structural elements is much smaller than the wavelength [20,32]. This is illustrated by the values of dpl in Table 1. The large variation is not so much due to inherent factors affecting accuracy, but rather due to the varying sizes of the monomer and of the whole aggregate. Still, we continue to use dpl or derived parameters in this paper due to their convenience for labeling different discretization. Overall, there are two factors complicating the application of the DDA for calculating BC aggregate optical properties. First, the performance of the DDA method is challenged if the real part of refractive index becomes relatively large [33]. Second, BC monomers have small size, so the choice of discretization (dpl) becomes critical, especially if both numerical accuracy and efficiency are considered.

Considering the significant uncertainties and demands for the DDA, this study presents a complete and systematic investigation on the performance of the DDA for the aggregate optical simulations, and develops a treatment based on the effective medium approximation to improve the DDA accuracy and efficiency. The paper is organized as follows. Section 2 introduces the DDA method and simulation parameters. The performance of the DDA is discussed in Sections 3, and a treatment based on the effective medium approximation is presented in Section 4 to improve the DDA performance. Section 5 concludes this study.

2. Method

As one of the most widely used numerical solution of light scattering by nonspherical particles, the features of the DDA has been extensively studied for general applications, whereas, typically, the particles are larger than the incident wavelengths. However, this study focuses on a special case of particles with relatively small overall sizes but complex geometries. The ADDA implementation (v1.3) developed by Yurkin and Hoekstra [34–37] is used in this study; it is parallelized with the MPI to be run on computer clusters. Due to the similar default settings and accuracy to another DDA code DDSCAT [36], our results can be understood for the DDA method in general. Some important parameters related to the DDA simulations are introduced in this section.

The DDA discretizes particles into small dipoles, and, for a given particle, its efficiency and accuracy are mostly determined by the dpl used for the simulation. The dipoles should be much smaller than both the incident wavelength and the particle length scale to better represent its geometry. For BC aggregates, typical monomer diameters range between 10 and 100 nm, and we are interested in their optical properties in ultraviolet, visible, or shortwave infrared wavelengths, i.e., in the order of a few hundred nanometers. Thus, the rule-of-thumb dpl value of 10|m| is definitely not sufficient; it should be large enough to adequately represent these small monomers. An alternative parameter describing the dipole size is its size parameter kd, where $k = 2\pi/\lambda$ and d are wavenumber and dipole size, respectively. The two are related as $kd = 2\pi/dpl$.

We use the default DDA formulation in ADDA [37], namely interaction of point dipoles and the lattice dispersion relation