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On the radiative properties of soot aggregates part 1: Necking and overlapping

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ABSTRACT

There is a strong interest in accurately modelling the radiative properties of soot aggregates (also known as black carbon particles) emitted from combustion systems and fires to gain improved understanding of the role of black carbon to global warming. This study conducted a systematic investigation of the effects of overlapping and necking between neighbouring primary particles on the radiative properties of soot aggregates using the discrete dipole approximation. The degrees of overlapping and necking are quantified by the overlapping and necking parameters. Realistic soot aggregates were generated numerically by constructing overlapping and necking to fractal aggregates formed by point-touch primary particles simulated using a diffusion-limited cluster aggregation algorithm. Radiative properties (differential scattering, absorption, total scattering, specific extinction, asymmetry factor and single scattering albedo) were calculated using the experimentally measured soot refractive index over the spectral range of 266–1064 nm for 9 combinations of the overlapping and necking parameters. Overlapping and necking affect significantly the absorption and scattering properties of soot aggregates, especially in the near UV spectrum due to the enhanced multiple scattering effects within an aggregate. By using correctly modified aggregate properties (fractal dimension, prefactor, primary particle radius, and the number of primary particle) and by accounting for the effects of multiple scattering, the simple Rayleigh–Debye–Gans theory for fractal aggregates can reproduce reasonably accurate radiative properties of realistic soot aggregates.

1. Introduction

Soot emitted from various combustion systems and fires into the atmosphere have been identified as the second largest contributor to global warming just after CO_2 [1,2] and found harmful to human health [3]. Once emitted into the atmosphere, soot particles undergo the so-called ageing processes during which soot aerosols are mixed with organic aerosols and often become coated or partially coated by organic matters (OM) and water [4,5]. The radiative properties of soot are highly required in many branches of science and technology, such as optically based diagnostics of soot properties (concentration, particle size), remote sensing, radiative heat transfer in combustion systems, and solar radiation. However, the radiative properties of soot depend on the refractive index of the bulk material, its morphology, and the degree of coating and the mixing state with the coating materials. It has been established through thermophoretic sampling followed by transmission electron microscopy (TEM)

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image analysis that combustion-generated soot can be described reasonably well as fractal aggregates formed by nearly uniform spherical particles [6,7]. The structure of fractal aggregates can be described by the following statistical scaling law [8]:

$$N_p = k_f \left(\frac{2R_g}{D_p} \right)^{D_f} \tag{1}$$

where N_p is the number of primary spheres in the aggregate, R_g is the gyration radius, k_f and D_f are the fractal prefactor and fractal dimension, respectively, and D_p is the primary particle diameter.

The commonly used approach in climate models for calculation of radiative properties is the Mie theory by choosing an equivalent particle diameter and an effective optical index in the case of coated soot, e.g., [9]. However, such approach has been demonstrated to cause significant error due to the rather complex fractal geometry of such particles that cannot be represented by a sphere [10–13].

In parallel, significant research efforts have been devoted to generate fractal aggregates that closely mimic the geometry of soot observed from transmission electron microscopy (TEM) images and to develop numerical methods for calculating the radiative properties of such aggregates. The numerical approaches for generating fractal aggregates can be classified into mimicking methods and tunable algorithms. The diffusion limited cluster–cluster aggregation (DLCA) method [14–17] belongs to the former and has often been used to simulate combustion generated soot, since it produces the observed fractal dimension of about 1.7–1.8. In tunable algorithms, the fractal dimension and prefactor are prescribed. The algorithm developed by Filippov et al. [18] is a typical tunable one. A numerical method for fractal aggregate generation returns the spatial coordinators of each constituent primary particle of an aggregate and the resultant aggregates are formed by point-touch primary particles of known diameter. By specifying the refractive index of each constituent primary particle the radiative properties of such numerically generated fractal aggregates can be accurately calculated by either the generalized Mie method (GMM) [19,20], the T-matrix (TM) method [21], or the discrete dipole approximation DDA [22–24].

Although very accurate numerical results of radiative properties of fractal aggregates formed by point-touch primary particles can be obtained by GMM, TM, and DDA, their main drawbacks are the requirement of large computational resources and very long computing time. Consequently, these methods cannot be coupled to climate models for calculations of radiative properties in the foreseeable future. Climate models require some simple model that is very computationally efficient and reasonably accurate. For the radiative properties of fractal aggregates formed by monodisperse primary particles the simple Rayleigh–Debye–Gans theory for fractal aggregates (RDG-FA) has been developed [25–28] and shown to be fairly accurate through numerical evaluation [25,29] and experimental validation [12,30]. A complete review of this theory has been made by Sorensen [31]. More recently, an effort has been made to incorporate the multiple scattering effects into the RDG-FA theory by introducing correction factors obtained from more accurate numerical results using DDA [32,33]. This is a very promising approach to retain the simplicity and efficiency of the RDG-FA formalism for climate modelling and in the meantime to achieve similar accuracy of the time-consuming DDA calculations. Such extended RDG-FA is also of great interest in various applications of optically based diagnostics to soot, such as static light scattering (SLS) [26,34], laser-induced incandescence (LII) [35,36], or in the experimental determination of the refractive index of soot [37–41].

Studies conducted so far on the radiative properties of numerically generated fractal aggregates have been almost exclusively focused on soot aggregates formed by monodisperse point-touch primary particles. However, the morphology of real soot aggregates revealed by TEM images is much more complex, e.g., [6,7,13], and involves more realistic aspects that are missing in DLCA aggregates, such as polydispersity of primary particles and overlapping and necking between neighbouring primary particles. Several studies made an attempt to account for these realistic aspects of the morphology of real soot. The effects of primary particle polydispersity on soot aggregate radiative properties were

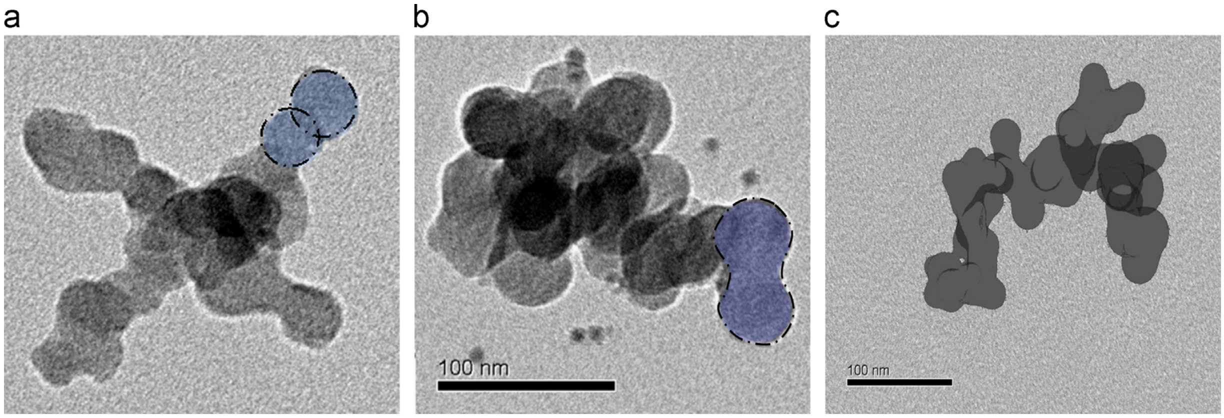

Fig. 1. TEM images of combustion generated soot aggregates, (a) and (b), and the virtual image of a numerically generated soot aggregate. An example of primary particle overlapping and necking is marked in (a) and (b), respectively. The numerical aggregate is generated with primary particle overlapping and necking. (a) Overlapping $C_{ov}=0.15$ (from the CAST soot generator [49]), (b) necking $\alpha=0.5$ (from an ethylene diffusion flame [34]), (c) virtual soot aggregate ($C_{ov}=0.15$, $\alpha=0.5$).