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Single scattering properties of semi-embedded soot morphologies with intersecting and non-intersecting surfaces of absorbing spheres and non-absorbing host

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ABSTRACT

The optical properties of light absorbing soot aerosols generally change through interactions with weakly absorbing particles, resulting in complex mixing states, and have been highlighted as a major uncertainty in assessing their radiative forcing and climatic impact. The single scattering properties of soot aggregates partially embedded in the host sulfate particle (semi-embedded soot-containing mixtures) are investigated for two kinds of morphologies with intersecting and non-intersecting surfaces. The surfaces cannot be overlapped in the non-intersecting surface morphology, while the intersecting surface morphology is unconstrained. Based on the modified diffusion limited aggregation (DLA) algorithm, the models with non-intersecting surfaces are simulated and applied for the single scattering calculations of semi-embedded soot-containing mixtures using the superposition T-matrix (STM) method. For comparison, the models with intersecting surfaces are simulated with the same morphological parameters, but some soot monomers are intersected by the host sphere. Due to the limitation of current STM method, the optical properties of these models with intersecting surfaces are calculated using the discrete dipole approximation (DDA) method. The soot volume fractions outside sulfate host ($F_{s,out}$) are introduced and applied to characterize the mixing states of the soot-containing aerosols. These simulations show that the absorption cross-sections of those internally, deeply, half and slightly embedded mixed soot particles ($F_{s,out}=0.0$, 0.2, 0.5, 0.8) are $\sim$105%, $\sim$65%, $\sim$43% and $\sim$14% larger than the semi-external mixtures ($F_{s,out}=1.0$), respectively. The results also indicate that the differences of extinction cross-sections, single scattering albedo (SSA) and asymmetry parameter (ASY) between simulations with intersecting and non-intersecting surfaces are small ($<1\%$) for semi-embedded soot-containing mixtures with the same morphological parameters. Within the range of visible and near-infrared wavelengths, the relative deviations of absorption cross-sections between these different morphologies are also small ($<\sim5\%$). Therefore, based on these simulations, the single scattering properties of semi-embedded soot-containing mixtures are rarely influenced by the morphological differences between the absorbing spheres intersecting and non-intersecting the non-absorbing host, which can nearly be ignored in the single scattering calculations.

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1. Introduction

Soot, which is a major aerosol constituent, resulting from fossil fuel, biofuel, and biomass combustion, is frequently found to be mixed with other weak or non-absorbing materials, such as sulfate, organics, dust, and sea salt [1–5]. The mixing state of soot in atmospheric aerosols approaches an internal mixture that leads to a much higher positive forcing [6–8]. The radiative forcing contribution of these light absorbing aerosols is still quite uncertain in climate forcing assessments because of the incomplete understanding of the radiative properties of soot and soot-containing particles with complex morphologies and chemical compositions.

Previous laboratory experiments show that the embedding of soot aggregates in weakly absorbing particles leads to the enhancement of light absorption and scattering. This amplification of the absorption cross-section for soot internally mixed (or coated) with sulfate or organics was measured as ~150–200% [9–12]. Soot coated with a concentric soluble shell yields radiative forcing estimates that are ~50% higher than those obtained with an external mixture model and ~40% lower than those with a homogeneous internal mixture model [13]. The radiative forcing at the top of the atmosphere was simulated as two times higher if fresh black carbon (BC) is modeled as an aggregate instead of a homogeneous sphere [14]. The radiative forcing is also ~20% less when modeling internally mixed BC particles as embedded lacy aggregates than with a simple core–shell shape, which is the shape assumed in many climate models [15]. These studies indicate that it is necessary to account for the optical effects of morphological differences and mixing states of soot in climate modeling [16].

The effects of aggregation and heterogeneity on light scattering and absorption have been analyzed for these types of morphologically complex aerosols using various methods in previous studies [17,18]. The optical properties of soot-containing aerosols in climate models are commonly obtained based on the morphological simplification of homogenous spheres using the Lorenz–Mie–Debye theory [19] or the assumption of homogeneous aggregates calculated using the Rayleigh–Debye–Gans (RDG) approximation with effective medium theory [20]. However, large discrepancies have been measured and simulated between the heterogeneous aggregates and the equivalent homogeneous sphere approximations due to the morphologies, components, and multiple scattering [21–25]. The radiative properties of soot aggregates are mainly influenced by complex morphological and chemical parameters, such as fractal structure, monomer character, mixing state, and refractive index. Until now, several methods were used to calculate effectively the optical properties of heterogeneous aggregated soot aerosols, including the generalized multi-particle Mie (GMM) method [26,27], the superposition T-matrix (STM) method [28–30], the geometric-optics surface-wave approach [31,32], and the discrete dipole approximation (DDA) [33–35].

For aggregated heterogeneous soot-containing mixtures, STM and DDA can be considered as the most popular methods with open source codes [36]. In Multiple Sphere T-Matrix (MSTM) version 3.0, the superposition T-matrix approach can be applied to arbitrary configurations of spheres located internally or externally to other spheres, the only restriction is that the surfaces of the simulated spheres do not overlap [37]. Wu et al. compared the optical effects of coated soot aggregates using the superposition T-matrix method, which showed the amplification of the absorption cross-section for thicker weakly absorbing coating [38]. Prasanna indicated that the MSTM method can be considered the best-suited method for one-point contact spherical monomers, based on comparisons between MSTM, DDA, and electrostatic results [39]. According to the DDA model, Soewono and Rogak indicated that the discrepancy between the scattering properties predicted and the equivalent core–shell Mie model become larger as the coating thickness of the soot monomer increases [40]. Scarnato et al. showed that the optical properties of internally mixed soot particles composed of soot and sodium chloride are consistent with reported experimental results [41]. Kahnert et al. indicated that the DDA method is sufficiently accurate for the optical calculations of external mixtures of soot aggregates and sulfate particle compared to the STM method with small enough dipole space and adequate discrete orientation angles, and then the optical properties of the semi-embedded mixing states for sulfate particle and soot aggregates are simulated using the DDA method [42]. Kahnert et al. also provided an important overview over model geometries for computing light scattering by small particles in atmospheric optics. Various morphological particle properties are discussed, such as overall non-sphericity, pristine shapes, aggregation, and different forms of inhomogeneity (e.g., porous and compact inhomogeneous morphologies), as well as encapsulated aggregates [43].

In this paper, we use a universal term “soot-containing mixtures” for indicating all kinds of mixtures of light absorbing soot aggregate and other weakly absorbing particle, such as semi-external, semi-embedded and internal soot-containing mixtures. Based on the modified diffusion limited aggregation (DLA) algorithm [44] and the constraint that all the surfaces cannot be overlapped, the morphologies with non-intersecting surfaces are first introduced and applied to the single scattering calculations of semi-embedded soot-containing mixtures using the STM method. For comparison, the models with intersecting surfaces of soot monomers and sulfate host are also simulated using the same morphological parameters. Due to the scattering calculation of models with intersecting surfaces beyond the capability of the current STM method, their optical properties are calculated using the DDA method. The quantitative differences of single scattering properties between the semi-embedded soot morphologies with intersecting and non-intersecting surfaces are further compared and discussed.

In Section 2, the theory and method for calculating the optical properties of aggregated heterogeneous soot-containing mixtures are introduced. In Section 3, random-orientation averaging results are presented for comparison and discussion. Conclusions are in Section 4.