Light absorption properties of coated black carbon aggregates with increasing fractal dimension

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Soot Aggregates (SAs) in the atmosphere significantly influence the earth's radiation balance, visibility, and public health. They are formed from high-temperature, incomplete combustion of fossil and biomass fuels via diffusion-limited cluster aggregation (DLCA) of spherical monomers. SAs can contain a significant amount of surface coatings of organic compounds which may alter their native fractal morphology through capillary and surface tension forces. Depending on the strength of these restructuring forces, the morphologies of SAs can be parameterized with increasing fractal dimension (D) with values ranging from $D = 1.8$ to 3.0. We used three aggregation mechanisms– DLCA, Percolation, and Face-centered cubic stacking – to generate aggregates with $D = 1.8$, 2.5, and 3, respectively. This range of D closely mimics the different morphologies of real-world SAs: bare ($D = 1.8$); partially collapsed ($D = 2.5$), and fully collapsed ($D = 3$). Next, we coated these numerical aggregates with non-refractory materials using a custom-made algorithm and calculated their numerically-exact optical properties using the discrete dipole approximation (DDA) algorithm. In many climate models, SAs are approximated by an equivalent-mass core-shell spherical model due to the ease of calculating optical properties using Lorentz–Mie theory. Keeping this in mind, we computed the optical properties of core-shell spheres equivalent in mass to our coated SAs. Comparisons of the core–shell spheres with the coated aggregates showed that the mass absorption cross-sections (MAC) were significantly underestimated in the core-shell approximation with increasing particle size. Due to their porous nature, the monomers of the aggregates are entirely illuminated by the incident light while the optical skin depths of the equivalent core-shell spheres prevent the black carbon core from participating in light absorption.

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