A modeling study of scattering and absorption properties of tar-ball aggregates

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Atmospheric tar balls (TBs) form an important class of atmospheric brown carbon (BrC) particulates. The morphology of the individual TBs is typically described as amorphous and nearly spherical. However, several studies reported observations of TBs aggregated with other aerosols or agglomerations consisting of up to tens of individual TBs. We use the superposition *T*-matrix method to compute the scattering matrix elements and optical cross sections for a variety of TB aggregates each of which is composed of a number of monomers whose sizes follow a log normal distribution. The results for a TB aggregate can differ fundamentally from those calculated for two simplified models commonly used in climate modeling, viz., the external mixture of TBs and the respective volume-equivalent sphere model. Clustering of individual TBs into an aggregate can either enhance or weaken absorption depending on the wavelength, the monomer size, and how absorptive the BrC material is. In the case of strongly absorptive BrC, aggregation results in enhanced absorption only at 1064 nm, while at 355 and 532 nm TB aggregates become less effective absorbers relative to the corresponding external mixtures. The effect of aggregation is always to increase the single-scattering albedo and asymmetry parameter, sometimes more than tenfold. The significant scattering-matrix differences between a TB aggregate, the "equivalent" external mixture, and the volume-equivalent sphere model demonstrate the failure of the conventional Lorenz–Mie theory to represent the scattering properties of morphologically complex BrC aerosols. We show that TB aggregates can help explain exceptionally strong and spectrally dependent lidar depolarization ratios reported in several recent studies.

Preferred mode of presentation: Oral