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Radiative properties of light absorbing carbon mixed with soluble compounds: Comparison of different morphological assumptions

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We consider internally mixed aerosols composed of light absorbing carbon (LAC) and soluble compounds. The aerosol morphology is represented by fractal aggregates of LAC encapsulated into spherical shells of weakly absorbing, soluble material. Optical properties are computed by the discrete dipole method for UV, visible, and IR wavelengths, and for different LAC volume fractions. The results of these detailed reference computations are compared to morphologically simpler model particles, namely, external mixtures of LAC and soluble spheres, internally, homogeneously mixed spheres with an effective refractive index, and inhomogeneous internal mixtures with a spherical LAC core concentrically covered with a spherical soluble shell. Different effective medium theories are tested for homogeneous internal mixtures. The results indicate that the external mixture model gives the poorest agreement with the coated aggregate model. Somewhat surprisingly, the concentric core-shell model performs considerably poorer than the homogeneous internal mixture model in conjunction with Maxwell-Garnett effective medium theory. However, the homogeneous internal mixture model tends to overestimate the absorption cross section, and it underestimates the single-scattering albedo.

Physically, these results can be interpreted as follows. The electromagnetic field cannot penetrate deeply into a compact LAC sphere. Thus this model particle excludes much of the LAC mass from interacting with the incident field, hence it underpredicts the absorption cross section. This problem affects both the external mixture and the core-shell model. The former suffers from an additional shortcoming. A coating of soluble material on an LAC aggregate increases the particle's physical cross section. As a result, this increases the scattering cross section. Additionally, the coating tends to focus more electromagnetic energy onto the LAC core, thus increasing the absorption cross section of the particle as compared to a bare LAC aerosol. These effects are not captured by the external mixture model, but they are included in the core-shell model. These are the reasons why both the external mixture and the core shell model underestimate the absorption cross section, but the core-shell model still gives more accurate results. The homogeneous internal mixture model, on the other hand, distributes the carbon mass evenly throughout the soluble particle. In a realistic aggregate, at least some of the monomers are shielded by other monomers from interacting with the external field. Thus the homogeneous internal mixture model tends to overestimate the absorption cross section.

Based on these results, we propose an improved model geometry. We consider a core-shell-shell model, i.e. a particle consisting of a spherical soluble core, a concentric spherical inner shell composed of LAC, and a concentric spherical outer shell of soluble material. With this model geometry we aim at mimicking the amount of LAC mass that is exposed to or shielded from the external field in a realistic coated aggregate. We demonstrate that for a suitable choice of the core radius we can reproduce the optical cross sections, the single scattering albedo, and the asymmetry parameter of the coated aggregates for all wavelengths and volume fractions considered in this study. Thus this model is particularly suited for assessing the short-wave climate forcing effect of internally mixed LAC aerosols.