Atmospheric black carbon radiative forcing driven by coating morphology

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Upon release in the atmosphere, the morphology and surface properties of soot promote the adsorption of local vapour phase molecules which initiates condensation. Coated soot particles contribute significantly to radiative forcing, due to the so-called lensing effect and to their extensive atmospheric lifetime [1]. However, the knowledge of the coating morphology is still limited, and non-uniform coating – which has been observed repeatedly via microscopy [2] – is generally omitted in models. Similarly, there is a need to understand the mechanisms of coating morphology evolution from local particle sources up to the high troposphere. Multiple sources of uncertainty related to the coating are commonly identified in the literature when modelling atmospheric soot ageing, especially coating morphology, kinetics, and associated chemical composition.

In this work, the compact aerosol aggregate model (CA^2M) [3] model is extended to represent a liquid coating on the surface of soot agglomerates and study the optical properties during encapsulation by a non-uniform coating (NUC). Typical atmospheric agglomerates morphologies are studied, i.e., open or compact structures including non-idealities, with coatings composed of organic carbon and sulfates. To simulate the impact of atmospheric soot ageing, both the physical and optical particle properties are calculated by coupling CA^2M to a discrete dipole approximation model [4]. In addition, several properties inherent to the estimation of atmospheric lifetime such as volume, mass, effective density, and equivalent diameters are obtained simultaneously. Finally, a new parametrisation in terms of mass absorption cross-section is proposed and used in a global circulation model (ECHAM6.3) and the aerosol microphysics module (HAM6).

Our results in terms of absorption enhancement by NUC-BC (E_{abs} = 1.35 at m_{rt}) agree with experimental observations from traffic and urban emissions [5], whereas using a core-shell model leads to significant overestimations (E_{abs} = 1.75 at m_{rt}). The transition (m_{rt}~14) is defined as the mass ratio ([coating+particle]/particle) required for the combined droplet to become spherical. Beyond this transition the droplet grows radially up to mass ratios of 50, as typically observed in the upper troposphere. The direct radiative forcing (DRF) induced by NUC-BC is about 0.18 W/m² for initial DLCA soot agglomerates which is 25 % less than the DRF induced by core-shell particles. The maximum DRF corresponds to regions in the China and Indo-Gangetic plains and to a lesser extent in the Arabic peninsula, Central Africa, and South America. Finally, we will shed light into how the BC dry deposition is affected by the present non-uniform coating and how it compares with simplified morphologies.

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