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Modelling the optical and radiative properties of freshly emitted light absorbing carbon within an atmospheric chemical transport model

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Abstract. Light absorbing carbon (LAC) aerosols have a complex, fractal-like aggregate structure. Their optical and radiative properties are notoriously difficult to model, and approximate methods may introduce large errors both in the interpretation of aerosol remote sensing observations, and in quantifying the direct radiative forcing effect of LAC. In this paper a numerically exact method for solving Maxwell's equations is employed for computing the optical properties of freshly emitted, externally mixed LAC aggregates. The computations are performed at wavelengths of 440 nm and 870 nm, and they cover the entire size range relevant for modelling these kinds of aerosols. The method for solving the electromagnetic scattering and absorption problem for aggregates proves to be sufficiently stable and fast to make accurate multiple-band computations of LAC optical properties feasible. The results from the electromagnetic computations are processed such that they can readily be integrated into a chemical transport model (CTM), which is a prerequisite for constructing robust observation operators for chemical data assimilation of aerosol optical observations. A case study is performed, in which results obtained with the coupled optics/CTM model are employed as input to detailed radiative transfer computations at a polluted European location. It is found that the still popular homogeneous sphere approximation significantly underestimates the radiative forcing at top of atmosphere as compared to the results obtained with the aggregate model. Notably, the LAC forcing effect predicted with the aggregate model is less than that one obtains by assuming a prescribed mass absorption cross section for LAC.

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