

## Optical and CCN properties of black carbon aerosols calculated using a mixing state resolved aerosol model

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Black carbon (BC) aerosols efficiently absorb solar radiation in the atmosphere and have been recognized as one of the most important aerosols for climate forcing. The magnitude of the radiative effects of BC depends on mixing state, which is the degree to which BC particles are coated with other aerosol compounds. Because BC is initially emitted into the atmosphere as bare, hydrophobic particles, the coating of BC through condensation of water-soluble compounds, such as ammonium sulfate, increases the efficiencies of both light absorption and wet removal of BC. In this study, a two-dimensional aerosol representation, which enables the resolution of the BC mixing state, was incorporated into an aerosol model module, Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (MADRID), and a new box model, MADRID-BC, was developed. An accurate mass transfer algorithm was also implemented into this model to partition condensable compounds among coexisting BC and BC-free particles, which do not contain BC, with different particle diameters.

The Pacific Exploration of Asian Continental Emission phase C (PEACE-C) aircraft observations conducted around Japan in March 2004 show that the mass fraction of thickly coated BC particles increased during horizontal transport from the Nagoya urban area within the planetary boundary layer (PBL) over the ocean. The MADRID-BC box model generally reproduces this feature well when observed bulk aerosol concentrations are used as constraints, suggesting that the evolution of the BC mixing states in this particular case can be interpreted primarily as a condensational growth process. Furthermore, the coating thickness of individual BC particles is predicted for the first time by model calculations.

Based on the predicted BC mixing states, aerosol optical properties are calculated using Mie theory by adopting a core-shell treatment. The BC coatings enhance light absorption by 38 and 58% in air leaving the source region and after a half day's transport, respectively. When one assumes all aerosol compounds are internally mixed with BC, the absorption coefficient and single scattering albedo are significantly overestimated and underestimated, respectively. These results show that it is essential to take BC-free aerosols into account in order to predict BC mixing states and aerosol optical properties in aerosol model calculations.

Cloud condensation nuclei (CCN) properties are also estimated based on the predicted BC mixing states using Kohler theory. When particles in air parcels leaving the source region and after experiencing a half day's transport are examined, 55 and 83% of the BC mass, respectively, are found to be contained within particles, with CCN activities for supersaturation of 0.05%. These results indicate the importance of the uplifting of BC particles near source regions for efficient long-range transport through the free troposphere.