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Development and validation of a size and mixing state resolved three-dimensional model

MATSUI, Hitoshi[1*], KOIKE Makoto[1], KONDO Yutaka[1], MOTEKI Nobuhiro[1]

[1]Graduate School of Science, University of Tokyo

The mixing state of black carbon (BC) aerosols is one of the most important issues for estimating radiative and climatic impact of aerosols. In this study, we developed a size and BC mixing state resolved three-dimensional model based on the WRF-chem model (MS-resolved WRF-chem). We adopted two-dimensional aerosol bin representation: one dimension is aerosol dry diameter (12 bins between 40 nm and 10 um) and another dimension is BC mass fraction to total aerosol mass concentrations in dry condition (10 bins). The detailed aerosol microphysical processes such as condensation/evaporation and coagulation are simulated with 120 aerosol bins.

The MS-resolved WRF-chem model was applied to East Asia in March and April 2009 when the A-FORCE aircraft campaign was conducted over the Yellow Sea and the East China Sea. Model calculations were compared with observed BC mixing state obtained by a single-particle soot photometer (SP2). Model calculations generally reproduced the main features of observed BC mixing state (e.g., BC-free to total aerosol number concentration ratio, diversity of shell (total particle dry diameter) to BC core diameter ratios (SC ratio), mean SC ratio, and their temporal variations and diameter dependence).

We evaluated the impact of microphysical processes (condensation and coagulation) on the BC mixing state of atmospheric particles. Sensitivity simulations suggest that BC aging processes can be classified into two regimes. Condensation processes are dominant for the growth of thinly coated BC particles (higher BC mass fractions), while coagulation processes are necessary to produce thickly coated BC particles (lower BC mass fractions). Although the impact of coagulation processes on total aerosol mass concentrations are very limited, they would be important in terms of the optical and radiative properties and the lifetime of BC.

We also conducted sensitivity simulations focused on mixing state of emissions. We compared two simulations: 1) the base case simulation in which all the BC emissions were assumed to be externally-mixed particles and 2) the sensitivity simulation in which about 50% of BC emissions were assumed to be internally-mixed particles based on the measurements at an urban site in Tokyo. The results suggest that the mixing state in emissions is an important factor to understand the BC mixing state of atmospheric particles more quantitatively.

Keywords: Aerosol, Black carbon, Mixing state, Regional three-dimensional model, Bin model, East Asia