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Volatility basis-set approach simulation of organic aerosol formation in East Asia

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Organic aerosol (OA) accounts for a significant mass fraction of the submicron aerosols in the atmosphere, and it influences the Earth's climate either directly (by scattering/absorbing of solar radiation) or indirectly (by modifying cloud microphysical properties). Recent studies show that secondary OA accounts for a large fraction of OA globally. However, as secondary OA formation processes are very complicated, estimates of the secondary OA burden in the atmosphere and its impact on climate and human health remain highly uncertain compared with those of other aerosols such as inorganic species.

In this study, OA simulations using the volatility basis-set approach were made for East Asia and its outflow region. Model simulations were evaluated through comparisons with OA measured by aerosol mass spectrometers in and around Tokyo (at Komaba and Kisai in summer 2003 and 2004) and over the outflow region in East Asia (at Fukue and Hedo in spring 2009). The simulations with aging processes of organic vapors reasonably well reproduced mass concentrations, temporal variations, and formation efficiency of observed OA at all sites. As OA mass was severely underestimated in the simulations without the aging processes, the oxidations of organic vapors are essential for reasonable OA simulations over East Asia. By considering the aging processes, simulated OA concentrations considerably increased from 0.24 to 1.28 μg/m^3 in the boundary layer over the whole of East Asia. OA formed from the interaction of anthropogenic and biogenic sources was also enhanced by the aging processes. The fraction of controllable OA was estimated to be 87 % of total OA over the whole of East Asia, showing that most of the OA in our simulations formed anthropogenically (controllable). A large portion of biogenic secondary OA (78 % of biogenic secondary OA) formed through the influence of anthropogenic sources. The high fraction of controllable OA in our simulations is likely because anthropogenic emissions are dominant over East Asia and OA formation is enhanced by anthropogenic sources and their aging processes. Both the amounts (from 0.18 to 1.12 μg/m^3) and the fraction (from 75 % to 87 %) of controllable OA were increased by aging processes of organic vapors over East Asia.

Keywords: aerosol, organic aerosol, regional three-dimensional model, anthropogenic-biogenic interaction, East Asia, volatility basis-set