Modeling secondary organic aerosol formation through cloud processing of organic compounds

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Abstract

Interest in the potential formation of secondary organic aerosol (SOA) through reactions of organic compounds in condensed aqueous phases is growing. In this study, the potential formation of SOA from irreversible aqueous-phase reactions of organic species in clouds was investigated. A new proposed aqueous-phase chemistry mechanism (AqChem) is coupled with the existing gas-phase Caltech Atmospheric Chemistry Mechanism (CACM) and the Model to Predict the Multiphase Partitioning of Organics (MPMPO) that simulate SOA formation. AqChem treats irreversible organic reactions that lead mainly to the formation of carboxylic acids, which are usually less volatile than the corresponding aldehydic compounds. Zero-dimensional model simulations were performed for tropospheric conditions with clouds present for three consecutive hours per day. Zero-dimensional model simulations show that 48-h average SOA formation is increased by 27% for a rural scenario with strong monoterpene emissions and 7% for an urban scenario with strong emissions of aromatic compounds, respectively, when irreversible organic reactions in clouds are considered. AqChem was also incorporated into the Community Multiscale Air Quality Model (CMAQ) version 4.4 with CACM/MPMPO and applied to a previously studied photochemical episode (3–4 August 2004) focusing on the eastern United States. The CMAQ study indicates that the maximum contribution of SOA formation from irreversible reactions of organics in clouds is 0.28 μg m^{-3} for 24-h average concentrations and 0.60 μg m^{-3} for one-hour average concentrations at certain locations. On average, domain-wide surface SOA predictions for the episode are increased by 9% when irreversible, in-cloud processing of organics is considered. Because aldehydes of carbon number greater than four are assumed to convert fully to the corresponding carboxylic acids upon reaction with OH in cloud droplets and this assumption may overestimate carboxylic acid formation from this reaction route, the present study provides an upper bound estimate of SOA formation via this pathway.

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