Roles of heterogeneous reactions in the tropospheric chemistry: a global modeling study

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Heterogeneous reactions in the atmosphere largely affect air quality and climate by changing tropospheric photochemical processes involving gases like O_3, HO_x, NO_x, HNO_3, CO, VOCs, and aerosols. There are many previous studies on the effects of heterogeneous reactions at regional to global scales using direct measurements, remote sensing, and simulation. For the atmospheric chemistry simulation, there is a large uncertainty from the treatment of the heterogeneous reactions in the model due to the still insufficient understanding of the mechanism of heterogeneous reactions.
This study is intended to understand the roles of the heterogeneous reactions in the global tropospheric chemistry using a chemistry-climate model CHASER (MIROCESM) (Sudo et al., 2011) which includes more than 250 chemical reactions in the troposphere and stratosphere with more than 70 chemical species. This model also includes aerosol components (SPRINTARS) for sulfate, nitrate, BC, POA/SOA, dust, and sea-salt; formation of nitrate and SOA, and ageing of BC are calculated tightly linked to the chemistry in CHASER. The present version of CHASER considers heterogeneous reactions of N_2O_5, HO_2, RO_2, and HCHO for surfaces of the individual aerosol components and cloud particles.
The model simulation is validated with the observational data from EANET, EMEP and the ship-based observation. Our performed sensitivity simulations with respect to the heterogeneous reactions show that the impacts of the N_2O_5 heterogeneous reaction may not be as large as previously reported; surface concentrations of NO_x, O_3 and OH are only reduced by 1.6%, 2.2%, 2.6%, respectively, in global mean (in regional, at most, 23%, 5.4%, and 12.8%). Whereas, the HO_2 reaction (to form H_2O_2) can cause a significant effect on distributions of O_3 and related species (NO_x, O_3, OH are changed by 68.7%, -21.5% and -70.1%). In addition to the polluted areas in China and India, we found that in the North Pacific region the heterogeneous reactions can play a critical role in the concentrations of NO_x, O_3, CO, OH, HO_2 (up to 69%, 21%, 7%, 70%, 81%, respectively). The possible large impacts of the heterogeneous reactions are also verified with the ship-based observational data for the North Pacific and Alaska regions.

Keywords: heterogeneous reaction, chemical climate model, North Pacific

