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A chamber study on effects of aerosol upon ozone production during VOC-NO_x photooxidation

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Tropospheric ozone, a chemically active trace gas and a short-lived climate forcer has increased over the past 30 years throughout the mid-latitudes of the Northern Hemisphere due to emissions of volatile organic compounds (VOCs) and nitrogen oxides (NO_x) from human activities. Previous atmospheric modeling studies have suggested that the increase in surface ozone over East Asia is due to changes in the concentrations of VOCs and nitrogen NO_x, as well as a decrease in the uptake of hydroperoxy (HO_2) radicals due to a decrease in aerosol concentration. However, to the best of our knowledge, there are no chamber studies that have validated the model predictions of the effect of HO_2 uptake on ozone production. To investigate the effect of aerosol on ozone concentration, we conducted chamber experiments of a VOC-NO_x-air irradiation system in the presence of copper(II)-ammonium sulfate aerosol, of which HO_2 uptake coefficient was determined to ~0.5 in a separate experiment. After six hours of irradiation, the total oxidant ($OX = O_3 + NO_2$) concentration decreased with increasing the initial surface concentration of copper-containing aerosol under NO_x-limited conditions. For example, the OX concentration decreased by 4% in the presence of the effective aerosol surface concentration of 8.2×10^8 nm^2 cm^{-3} compared to that measured in the absence of aerosol. In contrast, under VOC-limited conditions, the addition of copper-containing aerosol resulted in only a small decrease in OX. When pure ammonium sulfate particles were used, no or only a small decrease in OX was observed under NO_x-limited conditions. The effect of copper-containing aerosol on ozone concentration was qualitatively explained by the results of a detailed chemical box model that introduced HO_2 uptake. However, the model simulation underestimated the decrease in OX concentration due to aerosols by ~43%. Model results underestimated the decrease in OX concentration probably because the uptake of RO_2 was ignored in model simulations and/or there is an uncertainty of HO_2 uptake coefficient in a time scale of chamber study.