2.111 Direct measurement of heterogeneous reactive uptake of N2O5 on ambient aerosols in the polluted environment in China.

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Abstract:

Dinitrogen pentoxide (N_2O_5) is a reactive intermediate in the atmospheric oxidation of nitrogen oxides (NO_x), and its heterogeneous reaction on ambient aerosols play key roles in the atmospheric cycling of NO_x and photochemistry. Recent studies have revealed some knowledge gaps in the N_2O_5 reactivity on aerosol surfaces, which cannot be well characterized by parameterizations derived previously in lab studies based on aerosol compositions. To better understand the reactive uptake of N_2O_5 on the complex ambient aerosols, an in-situ experimental approach for direct N_2O_5 reactivity measurement was developed. The method utilizes an aerosol flow tube reactor coupled with an iterative chemical box model and can be used for the polluted environment with high and variable ambient precursors. Laboratory tests and model simulations have been performed to characterize the system, and the results demonstrated the applicability of this method under conditions of high NO_2/O_3 and fresh NO emission. This in-situ flow tube system was further deployed in the field to measure the N_2O_5 uptake coefficient in a suburban site in South China. During the observation period, ambient NO_2 and O_3 levels were in the range of 3~34 ppbv and 3~83 ppbv, respectively. The measured $\gamma(N_2O_5)$ ranged from 0.002 to 0.042. $\gamma(N_2O_5)$ showed positive dependence on aerosol water content and was suppressed by nitrate. Although these features generally follow the relationship suggested by lab studies, significant discrepancies exist between field-measured $\gamma(N_2O_5)$ and estimated values from different parameterizations. The results indicate that the complicated effects of some physicochemical parameters on the heterogeneous reactivity are still not well characterized, and further improvement of the parameterization is needed to account for the uptake variability in high-humidity and polluted conditions.