Heterogeneous Uptake of Gaseous N_2O_5 by Sulfate Aerosols

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Abstract

The heterogeneous uptake of gaseous N_2O_5 by ammonium sulfate [$(NH_4)_2SO_4$], ammonium bisulfate [NH_4HSO_4], and sulfuric acid [H_2SO_4] aerosols as a function of relative humidity has been investigated at room temperature and atmospheric pressure. Ammonium-containing aerosols were generated by a constant output atomizer and conditioned by passing through a diffusion dryer. Sulfuric acid aerosols were produced by the homogeneous reaction of SO_3 and H_2O in a borosilicate vessel. Addition of a dry or wet N_2 flow controlled the relative humidity (RH) of these aerosol flows. Using a chemical ionization mass spectrometer (CIMS) for N_2O_5 concentration monitoring and a scanning mobility particle spectrometer (SMPS) for aerosol characterization, reaction probabilities (γ) in the range of 0.001 to 0.1 for the uptake of N_2O_5 were determined as a function of RH. The results are expressed as follows: $\gamma[(NH_4)_2SO_4] = 2.79 \times 10^{-4} + 1.30 \times 10^{-4} \times (RH) - 3.43 \times 10^{-6} \times (RH)^2 + 7.52 \times 10^{-8} \times (RH)^3$, $\gamma[NH_4HSO_4] = 2.07 \times 10^{-3} - 1.48 \times 10^{-4} \times (RH) + 8.26 \times 10^{-6} \times (RH)^2$, and $\gamma[H_2SO_4] = 0.052 - 2.79 \times 10^{-4} \times (RH)$. We suggest that the water content and phase in the ammonium-containing aerosols control the reactivity of N_2O_5 while liquid-phase ionic reactions primarily dominate the uptake in sulfuric acid aerosols.