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Parameterization of N_2O_5 reaction probabilities on the surface of particles containing ammonium, sulfate, and nitrate

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Abstract. A parameterization was developed for the heterogeneous reaction probability (γ) of N_2O_5 as a function of temperature, relative humidity (RH), particle composition, and phase state, for use in advanced air quality models. The reaction probabilities on aqueous NH_4HSO_4, $(NH_4)_2SO_4$, and NH_4NO_3 were modeled statistically using data and uncertainty values compiled from seven different laboratory studies. A separate regression model was fit to laboratory data for dry NH_4HSO_4 and $(NH_4)_2SO_4$ particles, yielding lower γ values than the corresponding aqueous parameterizations. The regression equations reproduced 80% of the laboratory data within a factor of two and 63% within a factor of 1.5. A fixed value was selected for γ on ice-containing particles based on a review of the literature. The combined parameterization was applied under atmospheric conditions representative of the eastern United States using 3-dimensional fields of temperature, RH, sulfate, nitrate, and ammonium. The resulting spatial distributions of γ were contrasted with three other parameterizations that have been applied in air quality models in the past and with atmospheric observational determinations of γ. Our equations lay the foundation for future research that will parameterize the suppression of γ when inorganic ammoniated particles are mixed or coated with organic material. Our analyses draw attention to a major uncertainty in the available laboratory data at high RH and highlight a critical need for future laboratory measurements of γ at low temperature and high RH to improve model simulations of N_2O_5 hydrolysis during wintertime conditions.

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