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The heterogeneous chemical kinetics of N_2O_5 on $CaCO_3$ and other atmospheric mineral dust surrogates

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Abstract. Uptake experiments of N_2O_5 on several mineral dust powder samples were carried out under continuous molecular flow conditions at 298±2 K. At $[N_2O_5]_0=(4.0\pm1.0)\times10^{11}$ cm^{-3} we have found γ_{ss} values ranging from $(3.5\pm1.1)\times10^{-2}$ for $CaCO_3$ to (0.20±0.05) for Saharan Dust with γ_{ss} decreasing as $[N_2O_5]_0$ increased. The uptake coefficients reported in this work are to be regarded as upper limiting values owing to the fact that they are based on the geometric (projected) surface area of the mineral dust sample. We have observed delayed production of HNO_3 upon uptake of N_2O_5 for every investigated sample owing to hydrolysis of N_2O_5 with surface-adsorbed H_2O. Arizona Test Dust and Kaolinite turned out to be the samples that generated the largest amount of gas phase HNO_3 with respect to N_2O_5 taken up. In contrast, the yield of HNO_3 for Saharan Dust and $CaCO_3$ is lower. On $CaCO_3$ the disappearance of N_2O_5 was also accompanied by the formation of CO_2. For $CaCO_3$ sample masses ranging from 0.33 to 2.0 g, the yield of CO_2 was approximately 42–50% with respect to the total number of N_2O_5 molecules taken up. The reaction of N_2O_5 with mineral dust and the subsequent production of gas phase HNO_3 lead to a decrease in $[NO_x]$ which may have a significant effect on global ozone.

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