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The heterogeneous chemical kinetics of N₂O₅ on CaCO₃ and other atmospheric mineral dust surrogates

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Abstract. Uptake experiments of N₂O₅ on several mineral dust powder samples were carried out under continuous molecular flow conditions at 298±2 K. At [N₂O₅]₀=(4.0±1.0)×10¹¹ cm⁻³ we have found γ_{ss} values ranging from (3.5±1.1)×10⁻² for CaCO₃ to (0.20±0.05) for Saharan Dust with γ_{ss} decreasing as [N₂O₅]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 HNO₃ upon uptake of N₂O₅ for every investigated sample owing to hydrolysis of N₂O₅ with surface-adsorbed H₂O. Arizona Test Dust and Kaolinite turned out to be the samples that generated the largest amount of gas phase HNO₃ with respect to N₂O₅ taken up. In contrast, the yield of HNO₃ for Saharan Dust and CaCO₃ is lower. On CaCO₃ the disappearance of N₂O₅ was also accompanied by the formation of CO₂. For CaCO₃ sample masses ranging from 0.33 to 2.0 g, the yield of CO₂ was approximately 42–50% with respect to the total number of N₂O₅ molecules taken up. The reaction of N₂O₅ with mineral dust and the subsequent production of gas phase HNO₃ lead to a decrease in [NO_x] which may have a significant effect on global ozone.

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