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Homogeneous nucleation rates of nitric acid dihydrate (NAD) at simulated stratospheric conditions – Part II: Modelling

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Abstract. Activation energies ΔG_{act} for the nucleation of nitric acid dihydrate (NAD) in supercooled binary HNO_3/H_2O solution droplets were calculated from volume-based nucleation rate measurements using the AIDA (Aerosol, Interactions, and Dynamics in the Atmosphere) aerosol chamber of Forschungszentrum Karlsruhe. The experimental conditions covered temperatures T between 192 and 197 K, NAD saturation ratios S_{NAD} between 7 and 10, and nitric acid molar fractions of the nucleating sub-micron sized droplets between 0.26 and 0.28. Based on classical nucleation theory, a new parameterisation for $\Delta G_{act}=A\times(T \ln S_{NAD})^{-2}+B$ is fitted to the experimental data with $A=2.5\times10^6$ kcal K^2 mol^{-1} and $B=11.2-0.1(T-192)$ kcal mol^{-1}. A and B were chosen to also achieve good agreement with literature data of ΔG_{act}. The parameter A implies, for the temperature and composition range of our analysis, a mean interface tension $\sigma_{s/}=51$ cal mol^{-1} cm^{-2} between the growing NAD germ and the supercooled solution. A slight temperature dependence of the diffusion activation energy is represented by the parameter B. Investigations with a detailed microphysical process model showed that literature formulations of volume-based (Salcedo et al., 2001) and surface-based (Tabazadeh et al., 2002) nucleation rates significantly overestimate NAD formation rates when applied to the conditions of our experiments.

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