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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 $\text{HNO}_3/\text{H}_2\text{O}$ 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 $\Delta G_{\text{act}} = A \times (T \ln S_{\text{NAD}})^{-2} + B$ is fitted to the experimental data with $A = 2.5 \times 10^6 \text{ kcal K}^2 \text{ mol}^{-1}$ and $B = 11.2 - 0.1(T - 192) \text{ kcal mol}^{-1}$. 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 $\sigma_{\text{sl}} = 51 \text{ cal mol}^{-1} \text{ cm}^{-2}$ 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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