**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Homogeneous nucleation rates of nitric acid dihydrate (NAD) at simulated stratospheric conditions – Part II: Modelling" *by* O. Möhler et al.

O. Möhler et al.

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We acknowledge the comments and questions addressed by both referees of our paper. Here we reply to referee #2.

Answer to the specific comments:

The referee is right that the work by Tizek et al. (Phys. Chem. Chem. Phys. 4, 5128, 2002) investigated the formation of α-NAD and β-NAD (and other hydrates) by annealing amorphous nitric acid samples of variable molar composition. It was found that the more stable phase β-NAD was predominantly formed at a nitric acid mole fraction close to 0.33 (stoichiometric composition for NAD). At 180 K and nitric acid molar fractions below about 0.3, a second metastable dihydrate phase, α-NAD, was

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found to be more abundant than β-NAD. We will refer to this work in the introduction of the revised manuscript.

We also agree that a system involving metastable phases is not in thermodynamic equilibrium (good comment made by the referee). It is meant at this point that the supercooled droplets are in a stationary state in equilibrium with the surrounding gas-phase with a constant saturation pressure of nitric acid above the pure liquid phase. We will change the manuscript accordingly. We will also mention in the discussion, that the formation of thermodynamically less stable phases are favoured because of their lower activation barrier, according to Ostwald's step rule.

The interface tension may indeed contribute to the temperature dependence of the activation energy. However, any evaluation of the actual value or the temperature dependence of the interface tension is, to our knowledge, very uncertain. Furthermore, the T-dependence due to the interface tension is more important at lower NAD saturation ratios, S_{NAD}, where the contribution of the germ formation energy to the total activation energy exceeds that of the diffusion activation energy. No experimental data are available at low S_{NAD} and different temperatures, therefore any assumption of a T-dependence without additional information would be highly uncertain here. We therefore decided to use a constant interface energy as a first order estimate of the activation energy in the temperature range of the available experimental data. Because the interface energy normally decreases with decreasing temperature, this approach tends to overestimate the activation energy and therefore underestimates the nucleation rate at lower S_{NAD}. Because this point could also be important for an application of the parameterisation to stratospheric conditions with S_{NAD} normally less that 8, we suggest to add the above discussion in the manuscript.

Our plots show the activation energy as function of S_{NAD}, but with the temperature as parameter. The curves show exactly the dependence on S_{NAD} given in Eq. 5.

The work by Daniel Knopf (J. Phys. Chem. A 110, 5745, 2006) will be cited (see also

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answer to referee #1).

Answer to minor comments:

All minor comments will be considered in the preparation of the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 2119, 2006.

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