

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

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This paper presents results to expand the first paper. I have several points that the authors may want to address:

First, the y-axis in Figure 1 should be the measured nucleation rates in volume units. This can illustrate visually the differences between the nucleation rates (see Figure 6 in paper 1). Ideally nucleation rates for a fixed composition should be plotted as a function of T as shown in Tabazadeh et al. for a direct comparison. When composition is changed to saturation there is an inherent temperature dependence in the saturation

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ratio that causes problems with the types of comparisons shown in Figure 1. The reason Tabazadeh et al. used mole fraction in their parameterization was to avoid the T-dependence of the rate, which is embedded in the saturation ratio. In fact for a binary system Eq. (5) presented in this paper is incorrect. ΔG has a dependence on both a_{HNO_3} (activity of HNO_3 in solution) and a_{water} (activity of water in solution). See Reiss, Chemical Physics, 1950 for the correct formulation. The relations are similar to those used for $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ drop nucleation from the vapor phase.

The authors attribute lines in their figures to our work, which are incorrect. The nucleation code we reported should FIT the experimental data by Salcedo et al., Prenni et al., and Bertram et al. within the experimental range of data (temperature and composition) and the lines they attribute to our work does not seem to fit these datasets.

The sole point of our paper was to bring Salcedo et al. and Prenni et al. data sets in agreement for the same solution composition and temperatures ranges. If the authors insists that the rates are volume-based then they need to explain (1) why the data of Salcedo et al. and Prenni et al. differs by over three order of magnitude when expressed in volume units and (2) why the authors' dataset expressed in volume units agrees with neither salcedo et al. nor Prenni et al. datasets. Also, error bars need to be shown whenever experimental data is plotted on a graph.

Ps: A recent study concluded that homogeneous nucleation of water drops occurs on the surface in acoustically levitated fields (Lu et al., Appl. Phys. Lett., 87, 184107, 2005). You may find this paper of some interest to your analysis in terms of distinguishing experimentally between volume and surface rates.

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