

Interactive comment on “Commentary on ”Homogeneous nucleation of NAD and NAT in liquid stratospheric aerosols: insufficient to explain denitrification” by Knopf et al.” by A. Tabazadeh

Anonymous Referee #2

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This paper criticizes a recent paper by Knopf et al. (2002), where homogeneous nucleation rate coefficients of NAT and NAD in $\text{HNO}_3/\text{H}_2\text{O}$ and $\text{HNO}_3/\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ solution droplets are measured and used to predict nitric acid hydrate production under stratospheric conditions. In their stratospheric analysis, Knopf et al. assume that the nucleation of the hydrates occurs in the bulk phase of the droplets. It is precisely this assumption the one being disputed in this paper, which argues that nucleation in small droplets occurs in the air-solution interface. I think that the paper brings up a very interesting issue for discussion, and it is highly valuable. However, there are some points that need revision:

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1) The arguments given in the second paragraph of the paper are based on Figure 1, which shows the percent of surface molecules of a droplet as a function of its size. I presume that the percent refers to the total number of surface molecules. However, the second paragraph refers mainly to some surface-active molecules which partition preferably on the surface and which can affect the nucleation process. Hence, I think that figure 1 should plot the percent of surface-active molecules present on the surface of the drops (and not the percent of total number of molecules). Even better, it could show the surface-concentration of these active molecules versus droplet size.

2) In the third paragraph, the author writes: "...our recent studies (Djikaev and Tabazadeh, 2002) also show that the level of surface enrichment of a given species in a multicomponent solution is a function of the particle size". According to the references, these studies were presented during a meeting. I think that, since the studies by Djikaev and Tabazadeh (2002) are important to the discussion, it would be useful to summarize them in the text.

3) The following statements in the paper are misleading: a. At the end of the first paragraph the author writes: "Therefore, their criticism of the aerosol freezing rates (Salcedo et al.) used in the modeling study by Tabazadeh et al. (2001) is unjustified" b. In paragraph four the author writes: "...the experimental rates (Salcedo et al.) used in the modeling study by Tabazadeh et al. (2001) may not be completely accurate for the reason discussed in the Tabazadeh et al. (2002b) paper, and not for the reasons given in the Knopf et al. study."

These two statements lead to think that Knopf et al. disagree with the data presented in Salcedo et al. (2001). However, this is false, since Knopf et al. (2002) conclude that:

"Salcedo et al. (2001) have investigated the nucleation of NAD and NAT from binary aqueous nitric acid droplets. We consider their experimental data to be sound and the observed linear relationship, ..., to be valid in the experimentally observed range of saturation ratios."

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Actually, what Knopf et al. (2002) disagree with, is the extrapolation of the rates measured by Salcedo et al. to stratospheric conditions that Tabazadeh et al. (2001) performed in their study. On the other hand, the paper does not support the analysis by Tabazadeh et al. (2001) with any argument to contradict Knopf criticism. For this reason, I suggest these statements to be revised or removed.

4) Paragraph 4 reads: "...for the reasons given above the Knopf et al. experimental results are faulty and not applicable to the atmosphere". I think that there are not enough convincing arguments to prove that the experimental results are faulty. Most of the arguments in the paper support only the idea that the experimental results are not applicable to the atmosphere because the surface of atmospheric particles are different to the surfaces of the samples used by Knopf et al. I suggest that this sentence to be revised or to give more arguments against the experimental methodology.

Technical corrections:

a) Figure 1. The x-axis reads "Spherical sample size in sadius (micron)". Change "sadius" for "radius"

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