

***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***

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Reply to Interactive comment by Anonymous Referee # 1

I thank the referee for taking the time to review this letter. Below I will provide a point-by-point response to his/her comments.

Point 1: The referee may be tricked here by assuming what is shown in Fig. 5 is raw nucleation rate data. In fact Knopf et al. collected no new nucleation rate data for the range of solution compositions studied. What they show are possible upper limit freezing rates, making a volume-based assumption. Because Salcedo et al. and

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Bertram et al. report real experimental freezing rates and the data reported by Knopf et al. are possible upper limit rates, one cannot really compare these two quantities visually or otherwise. Nevertheless, I don't see a good visual correlation in Fig. 5. In fact I see discontinuities in energy functions shown in Fig. 5. As for contamination, the rates compared in Knopf. et al. paper come from different labs, where droplets were prepared in differ fashions. Thus, I doubt that they all had the same level of contamination per particle since the droplet size range from different laboratories varied by 5 orders of magnitude. As I stated in my commentary, the larger the droplet size, the more contamination can affect the rate of the freezing process. In addition, as described in our JPC paper (Tabazadeh et al, 2002b), there are a number of laboratory papers from Tolbert's group, which don't agree with the results show in Fig. 5 of Knopf et al.. For some reason, the laboratory datasets that don't agree with the conclusion of Knopf et al. were not cited or discussed in this paper.

Point 2: It may worth pointing out that no where in the Tabazadeh et al. (2001) paper a procedure is described for extrapolating Salcedo et al. experimental rates as suggested in the Knopf. et al. paper. In Tabazadeh et al paper, rate functions given in Salcedo et al (2001) were used, without any alterations, in a microphysics model to calculate their impact on the denitrification process. Salcedo et al. and Tabazadeh et al. papers are unique because they both take a first step in the right direction. The Salecdo et al. paper is unique because it reports, for the first time, a useable rate function for the nucleation process. The Tabazadeh et al. paper (2001) is also unique because it conducts model investigations, using nucleation rate functions, and not a number of various assumptions and sensitivity studies to look at the denitrification process. To improve science in this area one needs to take the second step of quantifying the process of freezing in the laboratory, and then using the quantified rates in a microphysics model in a sensible fashion. Conducting routine laboratory experiments, which provide null results, and modeling investigations, which rely on sensitivity studies, cannot really help us much in advancing science in this area. If possible, I prefer not to write a direct statement in the commentary regarding extrapolations of laboratory data.

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Comment

Problems with such extrapolations are discussed in depth both in the Salcedo et al. and Tabzadeh et al. (2002a, b) papers. As we have not conducted any extrapolations in our 2001 Science paper, it is inappropriate to write a statement in the commentary implying that we have extrapolated rate functions in this work. Also, based on the comments of referee 2, I have now removed discussions and citations to Tabazadeh et al. 2001 study.

Point 3: The statement is changed.

Point 4: The statement is removed.

A statement is added to the end of the commentary along the lines suggested by the referee.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 827, 2003.

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