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Interactive Comment

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

A. Tabazadeh

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NASA Ames Research Center, Earth Science Division, Moffett Field, CA 94035

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 is correct here and the total number of molecules on the surface is shown in Fig. 1 and not the number of surface-active molecules. Since Knopt et al. give no information on surface properties, it is rather difficult to calculate this along the lines discussed by the referee because one needs to know either the surface tension or the concentration of the surface-active component in the sample to do a meaningful calcu-

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lation. However, a monolayer assumption is a pretty reasonable one to make based on published literature data on what is known about organic films on surfaces of aerosol and cloud particles in the troposphere. The best review article on this subject was published 20 years ago by Gill et al. (1983). Recent studies in fact show a significant lowering of measured particle surface tensions in the atmosphere due to presence of organic films (see Facchini et al. and references therein), confirming what is dicussed in Gill et al.Šs paper many years ago. In this review, Gill et al. provides a rather extensive table (see Table 5 in this articles) on common surfactants that are present in the background troposphere. In addition, Gill et al. provide time constants on how long it would take for a finite surface in the troposphere to acquire an organic coating. Furthermore, as indicated in this letter, organic contamination can also accumulate in laboratory-generated samples through a variety of artifacts, which are present in a terrestrial laboratory (Middlebrook et al., 1997). Based on what is known on the condition of surfaces in the troposphere, it seems to me that the experimenter needs to be extra careful if her/she is interested in measuring freezing rates of particle systems in the stratosphere, where background conditions are quite different. Thus, to address the refereeŠs comment here, I have added references by Gill et al., Facchini et al. and Middlebrook et al., which clearly show water-based surfaces in the troposphere can quickly acquire an organic coating. This is equivalent to assuming a monolayer coverage of an organic film being present in the laboratory, which is basically the assumption used in the construction of Fig. 1.

Point 2: I agree with the referee on this since I myself donŠt like references to conference proceedings in published papers. Thus I have eliminated this reference. Instead I have referenced another manuscript, which is now published online. In this manuscript the mathematical formulation for the dependence of surface composition on particle size in a multicomponent solution is explicitly derived. The reason that the conference paper was cited before is because the results of the above-mentioned published paper were applied specifically to the $HNO3/H2O$ system.

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Points 3 and 4: I have eliminated a few statements that the referee highlights in his/her report. However, given what is described in our recent published paper (Tabazadeh et al., 2002b), it is clear that Knopf. et al. only provided analysis in their charts and graphs, which support their case. In fact sound laboratory data (Disselkamp et al., 1996; Prenni et al., 1998 Ű see Tabazadeh et al., 2002 for full citation), which do not agree with Knopf et al. work, were not mentioned, plotted or discussed in this paper. On the other hand, our analysis (Tabazadeh et al., 2002b) agrees with all published experimental work to date, including those omitted by Knopt et al. for one reason or another. Thus, when I state the experimental results of Knopf et al. are perhaps faulty is not based on an idea, as the referee seems to imply here, but rather it is based on facts (Tabazadeh et al., 2002b) that were basically ignored by Knopf. et al.. However, as I state in the last paragraph of my letter the main purpose of this commentary is not to dual on Knopf. et al.Šs criticism of our previous work (Tabazadeh et al., 2001), but to point out a need for more quantitative studies on this subject. Therefore I have willingly eliminated a few statements that criticized Knopf et al.Šs presentation of our previous work (citation to Tabazadeh et al. (2001) work is now removed) in order to make this letter more focused on what is important, which is careful attention to experimental details that may dramatically alter the rate of droplet freezing in some laboratories.

Spelling errors are corrected.

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

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