

Interactive comment on “Homogeneous nucleation rates of nitric acid dihydrate (NAD) at simulated stratospheric conditions – Part I: Experimental results” by O. Stetzer et al.

Anonymous Referee #1

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General comment.

This paper reports the results of laboratory studies of homogeneous nucleation of nitric acid dihydrate (NAD) out of binary $\text{HNO}_3/\text{H}_2\text{O}$ aerosol droplets under conditions representative of the polar stratosphere. The paper addresses an important scientific issue of the formation of solid phase particles in polar stratospheric clouds (PSC). Solid type PSC particles play an important role for stratospheric chemical ozone depletion through denitrification. Small number concentrations of relatively large PSC particles have frequently been observed in the stratosphere above the ice frost point tempera-

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ture (T-ice), and there is currently an ongoing scientific debate how these particles may form in air parcels which have not experienced temperatures below T-ice. Therefore, the results of the laboratory investigations are important to improve our understanding of the nature of PSCs, and the paper is of general interest to the atmospheric scientific community. The paper is well written, and I can recommend it to be published in ACP after some minor comments have been addressed as listed below.

Specific comments:

p. 2103, paragraph in lines 4-25: You indicate some errors (10-25%) in the estimates of the surface area of the liquid droplets from different assumptions about the parameters in a lognormal size distribution. Does this also take into account what you write in p. 2095, line 24- p. 2096, line 7 about the detection efficiency of the small particles sizes, including the mentioned correction factors and assumptions about the refractive index? It would be interesting to have an estimate on the total errors in the calculated nucleation rates, including uncertainties of other measurements entering the calculations, perhaps also indicated in figures 6 and 7.

p. 2100, line 14 and p. 2094, line 22: NAT was actually observed in the atmosphere in the experiments mentioned in the Larsen et al (1004) study and in other similar investigations (e.g. Voigt et al., Science 290, 1756-1758, 2000; Schreiner et al., J. Geophys. Res., 108, (D5), 8313, 10.1029/2001JD000825, 2003; Deshler et al., J. Geophys. Res., 108, 10.1029/2003JD0003479, 2003). So NAT does form in the stratosphere. As far as I read, the main point in the Larsen et al. study was that hydrate particles formed in the atmosphere in small number concentrations under sub-saturated conditions with respect to ice, i.e. above T-ice, with somewhat smaller nucleation rates than previously estimated. This seems to be consistent with your lab measurements, but these important points seem to be missing in your discussion. Whether NAT formed instantaneously out of freshly nucleated NAD or some time after the nucleation event seems to be of minor importance in the mentioned analysis of the atmospheric observations. I guess you only observed NAD for about 1 hour in one experiment? Is this enough to

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make firm conclusions about formation of NAT out of NAD in the stratosphere?

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 2091, 2006.

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