Atmos. Chem. Phys. Discuss., 6, S1404–S1407, 2006
www.atmos-chem-phys-discuss.net/6/S1404/2006/


Atmospheric Chemistry and Physics Discussions



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

**O. Stetzer et al.**

Received and published: 27 June 2006

We appreciate the referee's comments and answer as follows:

Page 2093, first paragraph (-line 4): References have been added.

Page 2094, first paragraph: A reference has been added.

Page 2094, line 13: Duft and Leisner extrapolated their results to a lower limit of 4 $\mu$m in radius which is equivalent to what we wrote.

Page 2094, line 22: Reference to Tabazadeh et al. Science, 2001 has been added.

Page 2095, line 25: We will add a graph to illustrate the fit of a size distribution against WELAS data and add some text to explain how we derived the correction factors.

Page 2096, 1st paragraph of ”Experimental Implementation” and page 2097, first paragraph: The time to reach a dynamic equilibrium is in the order of several minutes (as mentioned on page 2097, line 10). However, there is no real equilibrium with the walls since these "pump off" nitric acid and water constantly. The walls should not influence particle production since the heated inlet tube reaches into the chamber by about 1 meter.

Page 2096, line 24: Could you specify ”dry” conditions? It is mentioned in the first paragraph of the next page: around 10 % RHi.

Page 2097, line 12-17: Uncertainties for FTIR derived compositions are plotted in figures 3-5, panels E.

Page 2098, line 4: We don’t seem to understand your question here. Growth of NAD crystals can only happen due to water and nitric acid vapor transport from droplets to crystals via the gas phase, very much like the Bergeron-Findeisen process for ice. We think we explain exactly this on page 2098 briefly and later on page 2100, lines 15- in more detail.

Page 2100, line 11-14: We added a reference to Worsnop (1993) regarding the stability of NAD particles and also to Tabazadeh (2001) for the suggested conversion of NAD into NAT in a model study. Actually, Tabazadeh referenced Worsnop (1993) for the idea of NAD to NAT conversion, however, Worsnop only suggested a conversion into NAT on preexisting ice crystals in the presence of NAD on the last page of this paper. This discussion has been moved to the discussion section where it fits better.

Page 2100, line 24: The size thresholds for the detection of NAD were set so that the larger end of the droplet size distribution is not yet counted as NAD. In other words:

We moved the threshold down as much as possible while avoiding to have false counts of large droplets as NAD crystals (e.g. NAD concentrations after the droplet generation - and evaporation of ice crystals - must be zero). This will be clarified in the text, and



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also illustrated in a new figure.

The background concentrations before the experiments were measured with a CPC, thus a whole size range of particles (10 nm – 3 $\mu$m) contributes to the background we report. On the other hand, NAD particle concentrations were derived by integrating all size channels above a given threshold by an optical detector (WELAS). This instrument did not measure any particles at all before the aerosols were produced. This will be clarified in the text.

Page 2101, 1st paragraph: Starting with the production of aerosols, the AIDA chamber was always supersaturated with respect to NAT, hence NAT could not evaporate under these conditions. If the ice crystals at the beginning of some experiments would be covered with NAT these would have grown into large NAT crystals which we did not observe.

Page 2102-2103: See comment for page 2095, line 25.

Page 2104, line 15: Discussion is modified and now mentions that only data with similar molar fractions should be compared directly. We consider the agreement with other data still as good if one considers the scatter between individual measurements. This scatter (typically over more than one order of magnitude) is a known problem of experimental determined nucleation rates and is partly a consequence of the strong temperature dependence of J.

Page 2104, line 16: Discussion is extended to clarify this point. Regarding the relation between J and delta G we refer to the second part of this paper where this is discussed thoroughly.

Page 2105, line 5: We accidentally omitted the data by Salcedo et al. for X=0.278 and X=0.25. These data have been added to a revised version of Fig. 6. So our data lies in the range of previously reported data.

Page 2105, line 14: The work by Duft and Leisner is certainly an important contribution



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to the discussion about volume versus surface nucleation so it would be inappropriate to not mention it in this work. We do agree that a comparison is difficult given that the nucleating phase and the droplet sizes are different. But it is worth mentioning that theirs and our work come to the same conclusion anyhow.

Page 2106, 2nd paragraph: Unfortunately we are not able to characterize the mode of ice crystals well enough to derive an estimate for an upper limit for the nucleation of NAD or NAT on the ice surface. By FTIR we can derive an ice total volume of 560 $\mu$g/cm3 for E1 (at t=810 s) but the information about size is ambiguous and thus will be the calculation of a total surface area which would be needed to estimate nucleation rates.

Page 2106: Discussion of possible NAD->NAT conversion has been moved to the discussion section and added to the conclusions.

Technical corrections:

All technical corrections have been applied except for:

Page 2094, line 2: ”parameterize” left as is, since the whole document uses american english. Some remaining british english words have been corrected, though.

References: We are unable to find any missing initials?

Figure 7: Markers have been added to identify individual experiments.

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