

Interactive comment on “Ice supersaturations exceeding 100% at the cold tropical tropopause: implications for cirrus formation and dehydration” by E. Jensen et al.

T. Koop

thomas.koop@uni-bielefeld.de

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I would like to briefly comment about the ice nucleation experiments performed by Wise *et al.* (2004). In her comment A. Tabazadeh states that:

“The authors may want to cite a new paper by Wise et al. In this study they show that some organics can in fact nucleate ice at a warmer temperature than sulfate at the same water activity (see Wise, JGR, 2004). These experiments show that it is not the water activity per say that controls the rate of nucleation (of course there is

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always a strong dependence on water activity because that is related to the critical ice saturation terms in the classical nucleation model), but perhaps the surface tension of the ice nuclei formed in the organic solution is somewhat different than the surface tension of the ice nuclei formed in a sulfate solution.”

This is in contrast to the concluding statements in the paper by Wise *et al.* (2004). A brief excerpt from their abstract reads:

*“At levels detectable by our experimental technique we find that the freezing temperatures and critical ice saturation ratios of each system were identical, for a given water activity of the solution, even though the solutions contained varying fractions of inorganic and organic components. Further experiments showed that the freezing behavior of pure dicarboxylic acid particles was identical to that of the other systems studied if the water activity was identical. Although the apparent freezing temperatures reported here are substantially warmer than those predicted by the water activity based nucleation theory of T. Koop *et al.*, we find that solution water activity defined the freezing conditions for the systems studied.”*

Wise *et al.* (2004) did not find significant differences between the freezing of ammonium sulfate and dicarboxylic acid particles. However, the apparent freezing temperatures of both, ammonium sulfate and dicarboxylic acids, were higher than model predictions by Koop *et al.* (2000). With respect to the latter fact it is important to note that even for pure $(\text{NH}_4)_2\text{SO}_4/\text{H}_2\text{O}$ particles Hung and Martin (2001) and Hung *et al.* (2002) report that ice nucleation data from different experimental setups cannot be reconciled using a single J-function:

“...the T_f^ values reported for ice freezing events in $(\text{NH}_4)_2\text{SO}_4/\text{H}_2\text{O}$ particles for several*

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experimental apparatus do not appear to be selfconsistent” ... “Although this paper appears to reconcile qualitatively the AFT-IR results for (NH₄)₂SO₄/H₂O aerosols, Figure 10b shows further work is necessary to reconcile AFTIR, OM, DSC, and CFD measurements.”

Hence, these papers show that at present the data from the different setups cannot be reconciled using a single J-function. As a consequence, the ice nucleation model by Koop *et al.* (2000) has no choice but to disagree with some data (and agree with others) and, thus, predicts lower apparent freezing temperatures than observed in the AFTIR experiments by Wise *et al.* (2004), irrespective of whether (NH₄)₂SO₄ or dicarboxylic acids are investigated. This is also clearly stated by Wise *et al.* (2004):

“Because discrepancies in the conditions required for ice nucleation in particles containing ammonium sulfate from different experimental apparatus have yet to be resolved, the purpose of this paper is to qualitatively test the Koop et al. [2000] hypothesis for organics mixed with ammonium sulfate using a FTIR technique which employs freely floating particles.”

However, it is also very clear that the studies by Wise *et al.* (2004) on pure (NH₄)₂SO₄, glutaric acid, maleic acid, and l-malic acid particles as well as mixtures of these acids with (NH₄)₂SO₄ support our model in that *“the solution water activity defined the freezing conditions for the systems studied here”*.

References

H.-M. Hung and S. T. Martin, *J. Geophys. Res.* 106, 20379 (2001).

H.-M. Hung, A. Malinowski, and S. T. Martin, *J. Phys. Chem. A* **106**, 293 **(2002)**.

T. Koop, B. Luo, A. Tsias, and T. Peter, *Nature* **406**, 611 **(2000)**.

M. E. Wise, R. M. Garland, and M. A. Tolbert, *J. Geophys. Res.* **109**, D19203, doi:10.1029/2003JD004313, **(2004)**.

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