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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Heterogeneous formation of polar stratospheric clouds – Part 1: Nucleation of nitric acid trihydrate (NAT)" *by* C. R. Hoyle et al.

Anonymous Referee #1

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This paper describes a new NAT nucleation formalism involving foreign nuclei inside ternary liquid particles which is tested using the Zurich Optical and Microphysical box Model (ZOMM). The paper provides a short review of the history of NAT nucleation theories. It then puts forward a new heterogeneous NAT nucleation model, which has a physical background. The model results are compared with CALIPSO data with a fairly good agreement.

The paper often compares its results with those from a model in which a constant nucleation rate is assumed. Of course, such a model is not physically realistic, even though it has been used in several studies. The main point of the paper is that a constant

nucleation rate does not reproduce the data well, whereas a rate following nucleation theory, which has low nucleation rates when the temperature is just below TNAT and a rising rate as the temperature declines, or more properly the supersaturation rises, works better. While this is a useful point to make, I would have preferred to see the heterogeneous model compared with other models in which ternary solutions homogeneously nucleate. Like the current model, these could have been tuned if needed to best fit the data. One suspects that the main difference between using homogeneous and heterogeneous nucleation would occur late in winter as nuclei containing particles were preferentially removed.

The following questions and comments need to be addressed:

P7991 L9. The title of this section will confuse some readers. I think you mean you have developed a theory for NAT nucleation out of STS on foreign nuclei which is based on the theory for ice nucleation on a solid. This title makes it seem you are going to talk about ice nucleation, or nucleation of NAT on ice.

P7996 L20 Why would half the particles contain no foreign nuclei? I think there are data by Dan Murphy showing that a large fraction of the sulfuric acid particles have some meteorite in them. Also why would this nucleus be left as a solid instead of being attacked by the acid. There are recent papers by Neely et al and Bardeen et al which discuss the micrometeorites and their interactions with the background stratospheric aerosol layer. "Implications of extinction due to meteoritic smoke in the upper stratosphere" (R. Neely, J. M. English, O. B. Toon et al.,) Geophys. Res. Lett., 38 L24808, doi 10.1029/2011Gl049865, 2011. "Numerical simulations of the three-dimensional distribution of meteoric dust in the mesosphere and upper stratosphere" (C. G. Bardeen, O.B.Toon, E.J. Jensen, D.R. Marsh, and V.L. Harvey), J. Geophys. Res., 113, D17202, doi:10.1029/2007JD009515 (2008).

Fig 5 what to the contours mean? What does the word "wave" above the color bar mean?

Fig. 6 what does a temperature of -2 mean in the contours?

P8003 L25. The text suggests the reason for low STS was because the ERA temperature was too warm to form STS. However, there are a couple of other reasons that might apply in this case. The model results also overrepresented Mix2 (conclusion at P8006 L2) indicating that too much NAT nucleated. NAT would uptake the HNO_3 from STS because the vapor pressure over NAT is lower. The other reason could be the initial number density of the particles was too low which resulted in a low surface area density. A way to support the temperature bias idea is to apply a temperature reduction in the model and look at the STS formation.

P8004 L7. The paragraph about temperature fluctuations indicated an improvement in Fig. 9 of PSC formation compared with Fig. 8. But I didn't see a big difference. This paragraph is confusing. Did you decide that you needed to permanently change the value of gamma'? If so how does this impact the results you have previously discussed? What value do you think should be used? I think the paper needs a table which gives the values for the parameters that you conclude are final best choices.

P8006 L17. I assume you mean at "higher mean temperatures"

P8013. In Fig. 1, T_{dew} needs to be explained. Also SBS and STS do not have clearly defined boundaries. I'd think it is really STS that is leading to NAT. Otherwise you have no N to make NAT.

P8015. In Fig. 3, the figure shows different areas in the graph, which represent STS, MIX1, MIX2 or ICE. These areas need to be marked and explained in the figure caption for clarity.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 7979, 2013.