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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ice nucleation and cloud microphysical properties in tropical tropopause layer cirrus" *by* E. J. Jensen et al.

P. DeMott (Referee)

pdemott@lamar.colostate.edu

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General Comment

In this article, Jensen et al. describe investigations and calculations related to understanding the persistence and microphysical composition of TTL cirrus. While not a new topic, some new insights on the processes at play are offered, in particular the apparent inability to explain ice crystal observations via dynamical constraints and assuming that the homogeneous freezing process dominates ice formation at the low temperatures involved. The paper is well written, including an excellent discussion of the role of wave phase on the outcome of ice formation, and most arguments are well-qualified. Since my assessment is that this is a fine article that is suitable for publication with minor revisions, I do not intend my later comments to be taken as severe criticisms.

I do, however, feel justified in asking that the ammonium sulfate (actually ammonium sulfate/organic) heterogeneous freezing story to be tempered/qualified somewhat because of our continuing strong lack of information about the details of this process (see specific comments). While plausible based on present knowledge, I am concerned that this hypothesis is being carried far beyond our current knowledge of how this freezing process occurs, how complex compositions impact it (or override it), what proportion of particles otherwise looking chemically the same participate, and how this depends on temperature and relative humidity. I think that the paper mostly makes a strong case for the existence of some heterogeneous ice nucleus. I also wish to mention that at least one other potential ice formation process, preactivation, perhaps deserves mention as a possible source for ice nucleation.

Specific Comments

1) Abstract: Here I feel that you must add a caveat that this mechanism remains to be fully quantified for the size distribution of ammonium sulfate particles actually present in upper troposphere. More below.

2. Expectations of ice nucleation theory: This title is not quite accurate since a parameterization is applied for testing homogeneous freezing. Second, the authors might state that the application of a spectrum of gravity waves leads to about an order of magnitude increase in ice concentrations.

3. Section 4.2 Ice crystal size distributions: In the last sentence of this section, can a comment be made on if the assumption of gravity wave influenced trajectories in model simulations does or does not lead to broader size distributions? It would seem likely that multiple nucleation events would lead to this result, even if the concentrations are ultimately overestimated. Was this simulated?

4. Section 6 - TTL Ice Nuclei: I suggest that this section could be titled "Potential TTL Ice Nuclei" as the true source requires validation still.

a.Page 20649, lines 13-15: "...few (~5 L^{-1}) mineral dust particles were detected in the PALMS TTL measurements; these concentrations are well below the observed ice concentrations reported above." "Well below" could be quantified as "about ten times lower than" correct? If they are effective ice nuclei at lower RH_{ice}, would they impact the ensuing ice nucleation and the breadth of ice crystal distributions in the parcels to any degree? I would assume little impact on nucleated ice since they could only lower the ice concentration to their value, not some value intermediate to homogeneous freezing concentrations. This point is worth reiterating. But could there be an impact on broadening the ice size distribution? Another few questions though: How is this estimate of the amount of ice possibly due to mineral dust arrived at using PALMS data? For example, is this based on the proportion of all particles that display a mass spectral signature of dust that is then applied to a separately measured aerosol size distribution? My impression is that PALMS has a lower size limit for assessing composition and that the efficiency of defining different particle types may differ, so was a lower size limit applied and is there not some uncertainty in applying these data to make a quantitative estimate?

b. Page 20649, lines 25-26: "...ice crystals are not nucleating on a subset of the aerosol population that has a particularly unusual composition (Froyd et al., 2009b)." But do you know anything about the particular mixing state of the particles?

c. Page 20650, line 1: What about preactivation? When homogeneous freezing occurs anywhere, as it assuredly occurs in convection some other high cirrus, many particle have been ice including sulfates and some insoluble particles (perhaps even just small cores). When the ice crystals evaporate, the ice is not necessarily all lost from the nooks and crannies of particles even at long times with RH_{ice} below saturation. This is commonly referred to as preactivation. Although a loose lower limit of RH_{ice} of 50% has been ascribed to survival of preactivated IN based on past studies (e.g., *Roberts and Hallett*, 1968), this phenomenon has not been adequately characterized for the atmosphere because of the difficulty in doing so. It probably deserves more laboratory

study as well. TTL cirrus seems one of the more likely situations where this process could be at play. Since both processes have been observed in the laboratory, is there a reason that crystalline sulfate freezing is more likely?

d. Page 20650, line 1: Additionally, I suggest a qualifier be added to the statement here such that, "Recent laboratory studies have suggested that a small proportion of solid ammonium sulfate..." The reason is that the laboratory studies (those referenced and others) suggest only that some small fraction of crystalline sulfate, probably less than 1 in 1000 of all particles, will freeze heterogeneously. As near as I know, there is no knowledge of how this freezing fraction depends on temperature or how it depends on sulfate particle size. One could infer that such freezing has only been observed in studies for supermicron particles, yet the number of supermicron particles in the size distribution assumed valid for the TTL in the present study is only about 3 per liter I think. Thus, a great deal of uncertainty must surround the relevance of this freezing process for the atmosphere, and I assume that the reason you have not attempted to model it is due to the lack of sufficient detail regarding the specific ice nucleation properties (proportion of particles, efficiency versus RH, etc...).

e. Page 20650, lines 19-20: Same comment basically. Suggest writing, "Hence, heterogeneous freezing on some as yet undefined fraction of solid ammonium sulfate particles could easily occur before deliquescence in the cold uppermost tropical troposphere. Details of such a process remain to be quantified." This is just an additional qualification to the very nice discussion of the following two paragraphs.

5. Section 7 - Implications of heterogeneous nucleation for cloud processes, page 20652, lines 20-22: "In contrast, heterogeneous nucleation produces relatively low concentrations of ice crystals that can grow to relatively large sizes" But is this proven for distributions of sulfate aerosols? Do they possess a broad range of ice nucleation behaviors required for this result? All it seems possible to conclude is that there is some small population of particles that are acting heterogeneously or at least that is the result that would be consistent with observations. You do not know exactly what

they are and why they freeze that way, correct? Perhaps this comment is satisfactorily handled in the excellent statement made on page 20653, lines 25-27, but I had to read beyond this initial statement to find the other one.

8. Conclusions, page 20655, line 14: I only suggest you summarize here the type of additional knowledge that may be necessary to prove this conclusion.

Technical corrections

Page 20648, line 20: particles

References

Roberts, P., and J. Hallett (1968), A laboratory study of the ice nucleation properties of some mineral particles, Quart. J. Roy. Meteor. Soc. 94: 25-34.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20631, 2009.