Atmos. Chem. Phys. Discuss., 9, C7151–C7156, 2009
www.atmos-chem-phys-discuss.net/9/C7151/2009/

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.

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Received and published: 16 November 2009

Response to Interactive Comment (P. DeMott, referee)

We appreciate the constructive review, and we agree with the comments. The corresponding revisions to the manuscript have improved it considerably. Specific responses are given below.

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.

We have added such a caveat to the abstract.

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.

We have changed the title to "TTL cirrus microphysical properties predicted by homogeneous nucleation theory". The relative enhancement of ice concentrations caused by waves depends on the background cooling rate applied. We show that if a moderate cooling rate (3 cm/s) is applied, the ice concentration is increased considerably (Fig. 6). If slower background cooling were applied, the enhancement would be even larger (in a relative sense).

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?

Waves generally do not dramatically broaden the size distributions. In the simulations with waves, we typically do not get multiple nucleation events; the first nucleation event usually quenches the supersaturation, preventing subsequent nucleation. We find that the only way to get broad size distributions is to have slow, continuous nucleation that produces relatively few ice crystals. This scenario is consistent with heterogeneous nuclei having a variety of activity dependencies on temperature and supersaturation. We have added a discussion of these issues to the manuscript.

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.

Agreed. We have changed the title as suggested.

a.Page 20649, lines 13-15: ". . .few ($\simeq$5 L^{-1}) mineral dust particles were de-

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tected 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 RHice , 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?

Yes, the dust concentrations are about ten times lower than the measured ice concentrations. As mentioned in the following paragraph, the dust particles do not seem to be enhanced in the PALMS ice crystal residual measurements compared to the interstitial or out-of-cloud aerosol measurements. Therefore, surprisingly, the PALMS measurements suggest that the dust particles are not preferentially acting as IN. The concentration of ice crystals attributable to dust is determined as you describe, using the FCAS measurements of aerosol size distribution. The dust concentration estimate is for diameters $>$ 200 nm. In theory it is possible that PALMS is missing an unknown fraction of dust aerosol that are $<$ 200 nm. However, dust concentrations decline below 500nm, and dust aerosols are extremely rare at $<$ 300 nm.

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

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al., 2009b)." But do you know anything about the particular mixing state of the particles?

Since PALMS measures composition for individual particles, it is sensitive to particle mixing state. Almost every particle that formed ice was an internal mixture of neutralized sulfate and organic carbon. These sulfate-organic internal mixtures were also the dominant background particle type. As discussed in Froyd et al. (2009b), those particles that formed ice were not distinct in terms of organic/sulfate mass ratio, oxidized organic content, sulfate acidity, nitrogen content, or size. This is an important point that we have added to the manuscript.

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 RHice below saturation. This is commonly referred to as preactivation. Although a loose lower limit of RHice of 50has 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?

We agree that preactivation is worth mentioning. However, we also note that during the thin cirrus sampling during CRAVE, there was no convection reaching anywhere near the altitude of the clouds in the region of the observations. In fact convective-influence analyses (trajectories tracing the airmasses sampled back to where they last encountered convection) indicate that the TTL cirrus air sampled had not encountered convective outflow during the previous 2 weeks (Froyd et al., ACP, 2009). Back trajectories indicate that maximum temperatures experienced by the air sampled during the previous two weeks were about 5-15 K warmer than the temperatures during the TTL cirrus sampling. Assuming the air was saturated with respect to ice during the cloud sampling, the preactivated IN would have to needed to survive periods with RHice of 8-42%. Therefore, it seems unlikely that preactivated IN dominated the ice nucleation in these clouds.

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. . .).

We fully agree with these comments, and we have added caveats to the manuscript to address these uncertainties.

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.

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Done.

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.

We have changed "heterogeneous nucleation produces" to "heterogeneous nucleation could potentially produce".

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.

We have added the following: "Considerable additional research is needed to determine whether solid ammonium sulfate particles are important for TTL cirrus nucleation, including laboratory studies with appropriate particle sizes and compositions, and field measurement that determine both particle composition and phase."

The technical errors noted has been corrected.

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

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