

We thank Dr. DeMott for his insightful comments and suggestions. Our responses appear below in italics.

Overall this is an interesting and very useful study for which I suggest only a number of simple corrections or added statements. Although extremely long and somewhat repetitive regarding certain points, it is otherwise well written overall and makes its points well; these are that based on trajectory analyses, Asian mineral dusts reach altitudes where they can affect cold clouds as ice nuclei more often than do Saharan dust particles, but very few trajectories reach homogeneous freezing temperatures without first passing through clouds at warmer temperatures. The implication is that the impact of desert dusts is primarily on mixed-phase clouds, not cirrus, but that any impacts on cirrus can only be understood by studying ice formation by cloud-processed dust. A point that I think was not raised in this regard is that although some fraction of the particles may be lost to ice formation and other scavenging processes (biggest unknown) in the warmer cloud regions, the temperature dependence of ice nucleation by most dusts would seem to favor their persistence to low temperatures where they become much more active as ice nuclei. That is, a large fraction would not be expected to be lost as ice nuclei until temperatures well below -30_C.

We discussed this point on page 4045, lines 16-21: “However, given the distribution of saturation temperatures, it is reasonable to argue that a substantial fraction of dust IN remain inactive during MPC formation at $T > -20^{\circ}\text{C}$ and remain available as immersion or even deposition IN during any subsequent CIRRUS’ formation, perhaps even in some pre-activated form due to cloud-processing (see discussion in Section 3.2)”

However, we have clarified this statement further (in the same place): “However, given the distribution of saturation temperatures, it is reasonable to argue that a substantial fraction of dust IN stay inactive during MPC formation at $T > -20^{\circ}\text{C}$. They might either remain interstitial (becoming available as deposition IN during a subsequent CIRRUS’ formation event), or perhaps even transform into some pre-activated form after liquid phase processing (e.g., becoming immersion nuclei, see discussion in Section 3.2).”

This point is also discussed, albeit very generally, in the first paragraph of the summary.

There are a number of other subtle points regarding the potential formation of ice clouds in ice supersaturated regions at warmer temperatures that lead the authors to make valid suggestions for additional laboratory and field studies.

Specific Comments

Abstract

It is not clearly stated in the last sentence why studies of immersion mode ice nucleation of mineral dusts with “atmospherically relevant” coatings are needed for relevance to classical cirrus.

Clarified the potential paucity of deposition IN and the absence of liquid water, which together diminish the possibility of deposition nucleation and (the less explored) contact freezing in classical cirrus-forming temperature regions.

1. Introduction

Page 4030, lines 13-14: Concerning the statement “. . .although deposition freezing is also observed at warmer temperatures [e.g., Roberts and Hallett, 1968; Zimmermann et al., 2008],” would it be safe to say that deposition nucleation observed in these studies was for quite large mineral dust surfaces? It complicates matters as stated, but perhaps size and surface purity are at play? This occurs to one even without the exercises performed in this paper.

Added “for larger particles and/or higher surface defects.”

Page 4030, lines 19-21: “. . .because the ice nucleation efficiency of mineral dust is thought to decrease when it acquires chemical coatings (containing, e.g., nitric or sulphuric acids, or organics) through processing in liquid clouds.” I have to question why this result is preordained. If many dust particles need to go through a water activation process prior to ice nucleation at many mixed-phase cloud temperatures in any case (e.g., Field et al. 2006), why should coatings matter in the mixed-phase regime? In the cirrus regime it is understood that adding coatings can elevate the RH for ice nucleation by various aerosols via causing a switch in ice nucleation mechanisms [e.g., Moehler et al. 2005; Moehler et al. 2008].

We have clarified the text: In the exploration of dust-MPC effects we looked both for particles that could be coated and those that could be uncoated. Coated particles would participate in immersion freezing at lower temperatures while larger uncoated particles could initiate deposition nucleation and “regular-sized” (~1 um) uncoated dust particles could participate in the less explored contact freezing of water droplets at warmer temperatures.

2 African and Asian Dust Emission Regimes

Page 4031: Is mineral dust emission truly limited seasonally as the first sentence of section 2.1 suggests? Is it even necessary to restrict it so when this paragraph goes on to say that Saharan dust emission exhibits “considerable dust activity throughout the year” and that the Taklimakan is active year-round?

We have made a distinction between large, optically thick plumes visible from space, which definitely have their season, and the less spectacular dust emissions that occur year-round in some dust regions.

3.2 Limitations

Page 4037, lines 8-10: Moehler et al. [2008] have shown that SOA markedly suppresses ice nucleation efficiency of mineral dust IN “at cirrus temperatures.” I consider that there is very little published evidence regarding the role of coatings on freezing in the mixed-phase regime, although this is about to change.

Added “at cirrus temperatures”.

3.3 Data Analysis

Page 4038: A temperature of -40_C seems to be formally too cold to be considered a mixed-phase cloud. The transition usually occurs warmer than this, except perhaps with strong convective cloud forcing. I understand it probably makes little difference to the exercise performed.

Indeed. We tried slightly warmer transition temperatures in the analysis and it made very little difference to the outcome that MPCs are the dominant cloud type.

4.1 Dust/Cloud interactions by cloud type and dust region

Page 4042, lines 11-15: Again, I am not sure why it is a necessary conclusion that the exposure of dust particles to water or solution coatings “will conceivably deteriorate their efficiency in serving as condensation/immersion ice nuclei [Pruppacher and Klett, 1997].” Can you state where in this reference this is discussed?

This is a mistake. Corrected “condensation/immersion” to “deposition or contact”.

Page 4043 discussion: This discussion on temperature details breaks things down by supposed favored ice nucleation regimes. This is fine, even though I feel based on unpublished data that one cannot put a simple border at say -20_C for separating contact-only versus immersion freezing or deposition nucleation.

Added “roughly” to the temperature boundaries given.

Nevertheless, I consider it rather speculative to say that a most important consideration is that “trajectory points reaching the mixed-phase cloud phase space (ii) of Figure 4 must pass through the ice-saturated WARM-HET cloud region (v), where the most efficient dust IN may have already been activated in deposition mode.” I do not think of deposition nucleation as especially effective in the ice supersaturated regime of mixed-phase clouds, but there are some references that support this possibility (e.g., that deposition could occur for some dust as warm as -10_C) that have already been given in the introduction (and perhaps should be repeated here). Actually, such discussion does occur on the following page, so it may simply be that this is a case of a need to reorganize and reduce the overall length of discussion.

We have added clarifying statements in this section.

Page 4044, lines 6-7: It might be useful to distinguish between the actual formation of WARM-HET clouds and the conditions favorable to the formation of such clouds, since they may not form at all. That is “Outside JJA, conditions favorable to the formation of WARM-HET clouds. . .”

We have added four instances of “conditions favourable for”, four instances of “potentially” and 1 instance of “predicted” in this paragraph to draw attention to the uncertain existence of these clouds.

Page 4045, lines 15-16: Coming back to the point about limiting the discussion of simple segregation of mechanistic regimes for ice nucleation, the statement “. . .at which point dust IN start to become fully active in condensation/immersion freezing [Hoffer, 1961]” could be removed as unnecessary.

Done.

Page 4045, lines 22-23: “although sample size may be an important confounding factor (Archuleta et al., 2005).” What is meant by this statement in regard to preactivation studies? Please expound.

We meant “particle size” instead of “sample size”. Dropped the reference to Archuleta et al. [2005] in this paragraph since they investigated particle size effects, not discrepancies between multiple studies, as the sentence may have implied.

Page 4047: Here there is still more discussion of the possibility of WARM-HET clouds and the possible role of deposition nucleation. This seems excessive. Within this discussion, it is stated that Connolly et al. assume “. . .that deposition nucleation could have been underestimated.” What led them to this conclusion? It is not clear if left as a simple statement of fact. Secondly, in the Zimmerman et al. study referenced, does 20-nm imaging resolution afford the ability to clearly detect the many monolayers of water that would represent? I question the strength of this result.

We believe that a full discussion of the laboratory results summarized in Figure 8 is necessary; however, we agree that clarifications and streamlining are needed. Connolly et al. [2009] state in their conclusions that “Deposition nucleation was negligible for temperatures warmer than -12.5C (not shown). In the experiments shown here all of the dust particles in the AIDA acted as CCN, leaving no interstitial dust particles that could act as deposition nuclei. In the atmosphere however, it is reasonable to assume that this would happen and significant deposition nucleation could take place before the formation of liquid drops.” We added more information in this regard to the text. Also, we agree that the 20-nm resolution employed by Zimmermann et al., [2008] is coarse and we have removed the sentence referring to a lack of observed water condensation at high supersaturations w.r.t. water.

Page 4048, lines 1-6: I question the acuity of this statement made in Ansmann et al. [2008] and I will question its repetition here. They did not observe a high number of ice nuclei, but a high number of dust particles, the difference being that only 1 in 1000 or less will be active at -20_C. The remainder of the discussion on this page seems too much for the few valid points made. Would it not be enough to say that in the Ansmann et al. [2009] studies, most of the cloud observations were separated from the primary levels of Saharan dust?

The discussion has been shortened, as suggested.

5 Summary

Page 4054: I question making a broad sweeping statement that “further field campaigns should focus on the Asian region” when in fact the issue of what happens to dust during convective transport remains a significant and complex one. This is especially true of dust surrounding developing tropical cyclones in the Atlantic basin, extremely important storm systems, so I find it wrong to sweep this question away. It has not been well addressed at all with the right sorts of measurements.

This is a good point, which we have adopted in our summary.

References

Möhler, O., Büttner, S., Linke, C., Schnaiter, M., Saathoff, H., Stetzer, O., Wagner, R., Krämer, M., Mangold, A., Ebert, V., and Schurath, U.: Effect of sulfuric acid coating on heterogeneous ice nucleation by soot aerosol particles, *J. Geophys. Res.*, 110, D11210, doi:10.1029/2004JD005169, 2005.