<u>Anonymous Referee #1, 2nd review</u> (Review comments in regular**; response in bold**.)

Regarding the last paragraph under General Comments, in reference to the work of Diao et al., it might be argued that this comment pertains best to heterogeneous ice nucleation (het). In the Lagrangian context of a rising air parcel, since the ice phase will first manifest through het, and if homogeneous freezing (hom) occurs it will happen after het, then it follows that pre-existing ice is present when hom occurs but is generally not present when het occurs for the cirrus sampled in the Diao et al. papers.

Answer:

We agree with the reviewer that homogeneous nucleation happens after the heterogeneous nucleation. Since homogeneous nucleation requires higher critical supersaturations to take place, preexisting ice has a larger effect on homogeneous nucleation (through depleting water vapor in the air parcel).

The ice nucleation phase (phase 2) in the GRL Diao et al. papers shows the highest RHi values (on average) for the ice-containing phases, indicative of ice nucleation. However, Ni is often higher in phase 3 (early crystal growth stage), suggesting nucleation continues at lower RHi values. This seems counter-intuitive, making the Diao et al. measurements more difficult to interpret.

Answer:

In Diao et al. (2013 GRL), the authors commented on the increasing Ni and the wide distributions of Ni during the evolution of ice crystal regions (ICRs) (i.e., early crystal growth stage): "The increasing Nc agrees with previous simulations, where new ice crystals continue to form with continuous uplifting [Spichtinger and Gierens, 2009]. On the other hand, not all ICRs experience the same process, which leads to the wide range of Nc distribution for aged ICRs." Diao et al. (2013) mentioned that the increasing Ni is likely due to the new ice crystal formation as the ICRs expand in space with respect to ISSRs, yet in Diao et al. (2013) it was not discussed whether these new ice crystal formation that contributes to continuous increases in Ni is from heterogeneous or homogeneous freezing. It is possible that (1) entrainment of new ice nuclei would lead to additional heterogeneous freezing beyond the initial nucleation events, or (2) homogeneous freezing occurs after the initial nucleation, or both. In fact, it is possible that both cases may occur since Figure 3a in Diao et al. (2013) actually shows increasing standard deviations of Ni distribution along the evolution of ICRs, with Ni ranging from 100 L^{-1} – 3200 L^{-1} (at SID-2H instrument measurement range of 3-50 micron), indicating not all air masses are undergoing the same increasing rate in Ni.

About the reviewer's comment on the decreasing RHi in early crystal growth stage, we want to point out that the RHi values shown in Figure 2 in Diao et al. (2013) are the spatially averaged RHi values for the whole ISSR+ICR samples (as defined in their Figure 1). They do not represent the local RHi or the maximum RHi inside each ISSR+ICR sample. Freezing of ice crystals is dependent on the local RHi, not the average RHi. Thus it is not counterintuitive that the average RHi value

decreases as water vapor deposition onto ice crystals continues, but as long as some local RHi values satisfy the conditions for heterogeneous/homogeneous freezing, new ice crystals can still form.

Another point regarding Zhou et al. is that the time-step in CAM5 is 30 minutes. When the cirrus cloud updraft w is derived from TKE spectra, it appears that a significant percentage of the cirrus will have sufficiently high w to enable both het and hom to occur in a single time-step. In these cases pre-existing ice for either het or hom makes little sense for the cirrus cloud types sampled by Diao et al.

Answer:
The cirrus sub-grid vertical velocity (Wsub) (i.e., the standard deviation of vertical velocity spectra) in CAM5 is derived from TKE and assumed to be constant within a time step of 30 minutes. First, this Wsub is not already sufficiently high to enable both the homogeneous and heterogeneous nucleation to occur in a single time-step (see Shi et al. 2015 for the comparison of PDF (Wsub) with SPARTICUS observations). Second, our recent study finds that when we use the vertical velocity spectra (i.e., consider the high-frequent fluctuations of vertical velocity) within the 30-min time step in the ice nucleation, the occurrence frequency of homogeneous nucleation is significantly reduced, because the negative values of vertical velocity spectra reduce the ice supersaturation in the air parcel. This result is consistent with Spichtinger and Krämer [2013].

The cirrus cloud modeling work of Spichtinger and Gierens (2009, ACP, Part 2) appears consistent with the observed phases of cirrus evolution in Diao et al. (2013, GRL, p. 3477). The authors might refer to this detailed modeling work, noting that most of the ice supersaturated regions were consumed by ice diffusional growth within 30 minutes, with ice nucleation occurring on much shorter timescales. The question is how often do both het and hom occur in a single CAM5 time-step, and how realistic is the treatment of pre-existing ice in the updated CAM5?

Answer:
Thank the reviewer for pointing us to Spichtinger and Gierens (2009, ACP, Part 2). We will refer to this modeling study in the revision. We note that water vapor consumption by ice diffusional growth in cirrus clouds is treated in the CAM5 cloud microphysics within 30 minutes. We agree with the reviewer that the ice nucleation occurs on a much shorter time scale (a few minutes), is a sub-grid process, and thus has to be parameterized in large-scale models such as CAM5.

We agree with the reviewer that the frequency of both homogeneous and heterogeneous nucleation occur is an important scientific question. This is why we validate our CAM5 simulations against in situ observations by Cziczo et al. (2013). We find that the frequency of homogeneous nucleation agrees better with Cziczo et al. (2013) when we consider the pre-exist ice in the updated CAM5.

Reference

Cziczo, D. J., Froyd, K. D., Hoose, C., Jensen, E. J., Diao, M., Zondlo, M. A., Smith, J. B., Twohy, C. H., and Murphy, D. M.: Clarifying the Dominant Sources and Mechanisms of Cirrus Cloud Formation, Science, 340, 1320–1324, doi:10.1126/science.1234145, 2013.

Shi, X., Liu, X., and Zhang, K.: Effects of pre-existing ice crystals on cirrus clouds and comparison between different ice nucleation parameterizations with the Community Atmosphere Model (CAM5), Atmos. Chem. Phys., 15, 1503-1520, doi:10.5194/acp-15-1503-2015, 2015.