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

General comment:
In this study the representation of ice crystal number concentrations in the CAM GCM is investigated. As a reference concentrations as obtained by in situ measurements are used. Ice crystal formation at low temperatures (T < 235 K) depends crucially on local dynamics. Since in largescale models subgrid scale motions cannot be represented by definition, the relationship between ice crystal formation and vertical motions must be parameterized. In this study the authors investigate different possible parameterizations and their impact on the resulting ice crystal number concentrations in the CAM GCM.

In general, this is an interesting and important contribution to ice cloud research; thus, this study is an appropriate contribution for ACP. However, there are some issues, which should be clarified before the manuscript can be accepted for publication. Therefore I recommend major revisions of the manuscript. In the following I will explain my concerns in detail.

Major points
1. Sedimentation in GCMs
A crucial process for the evolution of ice clouds in the tropopause region is sedimentation of ice crystals. As known from many studies using models in different configurations (box models, column models or even full 2D/3D model) sedimentation can shape the evolution of ice clouds in a very crucial way. From the manuscript it is not clear how sedimentation is treated in the used cloud parameterization of the GCM and how this parameterization would influence the results. Thus, the authors should add some text about the treatment of ice crystal sedimentation in the model. In addition, and more important, the authors should try to carry out some sensitivity studies changing the treatment of ice crystal sedimentation (e.g. changing the terminal velocities, if they are treated explicitly in the cloud scheme). This would lead to a better understanding of the interaction of different ice cloud processes in the model. In a consequence it might be that ice nucleation is less sensitive to vertical velocity representations, since sedimentation tends to smear out strong changes in number concentrations and enhances the effect of pre-existing ice.

Answer: In CAM5, the mass- and number-weighted terminal fall speeds for ice number and mass are obtained by integration over the particle size distributions with appropriate weighting by number concentration or mixing ratio and are then applied to the entire population of ice particles in each grid (Morrison and Gettelman 2008). We added this information in the paper:
"…sedimentation of ice is included in the GCM (CAM5 in this study) by applying the mass- and number-weighted terminal fall speeds which are obtained by integration over the particle size distributions with appropriate weighting by number concentration or mixing ratio..."

We ran a series of sensitivity tests by doubling the terminal velocities for all COMP cases in the paper for one year, yet the differences between the doubled terminal velocity cases and their corresponding base cases are relatively smaller than the differences caused by using different sub-grid updraft velocities or different water accommodation coefficients. Thus we decided not to include these results in the paper. Below is one example showing the results from the WTKE_COMP cases with three different terminal fall velocities (base case: Sed-1X, doubled terminal fall velocity: Sed-2X and 10 times terminal fall velocity: Sed-10X). The doubled fall velocity case slightly increases the ice number at the higher temperature range (T>205K) probably due to the increased sedimentation of ice from above. The more extreme case with 10 times terminal fall velocity does show a significant decrease of ice

number at T<215K. But the decrease is still smaller than using α = 1 or using WGARY/WLARGE as the sub-grid updraft velocity.

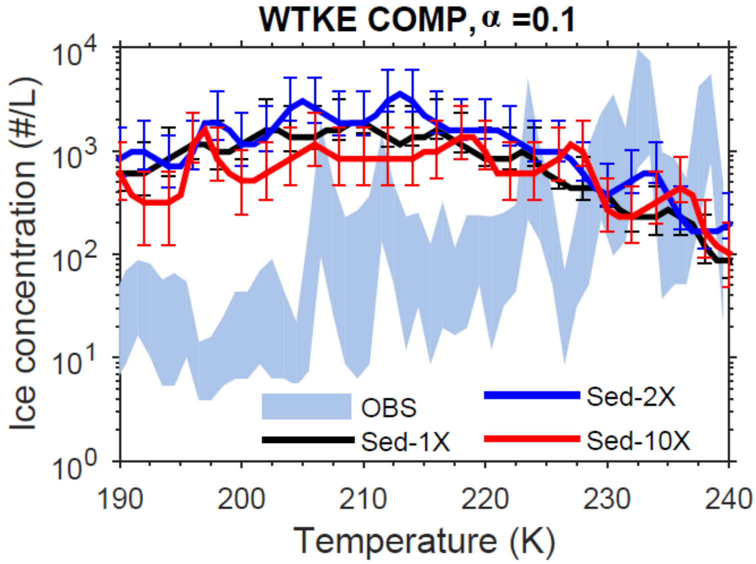

Fig 1. Same as the WTKE-COMP case in Fig 5a but with different terminal fall speeds for ice.

2. Combination of different approaches seems arbitrary

In the last part of the manuscript it is suggested to use a mix of different representations of vertical velocities (or cooling rates, respectively) in order to represent ice crystal number concentrations in a better way. The combination of WGRID and WTKE by using a simple temperature criterion seems to be too simple. As indicated in minor point 6 below, the use of WGRID for the cloud parameterization is recommended by Spichtinger and Krämer (2013) only for a special regime of strong stratification (i.e. tropical tropopause layer). A simple temperature criterion changing the vertical velocity at the threshold T_c = 205K will not work, since the key property is the strong stratification, which occasionally coincides with low temperatures in the TTL. In addition, it is not clear (see minor point 4 below) what the TKE scheme is doing in the upper troposphere. Therefore, the use of WTKE is still questionable, although it might reproduce meaningful ice crystal number concentrations but maybe due to the wrong reasons. In fact, this issue should be clarified first, although this might be beyond the scope of the study. From a practical point of view, I recommend to use a dynamical criterion to split the different regimes (WGRID vs. WTKE), e.g. a threshold using bulk stratification as Brunt-Väisälä frequency calculated on model resolution. Of course, sensitivity due to such a criterion should be explored.

Answer:

First of all, the moist turbulence scheme of CAM5 now simulates cloud-radiation-turbulence interactions in an explicit way and is operating in any layers above as well as within PBL as long as moist Richardson number is larger than a critical value, 0.19. So similar to what it does in the PBL the TKE schemes also vertically transport heat/moist/condensates/momentum/tracers by symmetric turbulences in the upper troposphere but with a much smaller magnitude.

We agree that the use a mix of different representations of vertical velocities lacks theoretical or observational support at the current stage. We pointed out this in the manuscript. We chose T_c = 205K as the threshold to show that some potentially missing

physics below this temperature like the glassy SOA may make this temperature critical. We agree that it could be that this temperature simply coincides with the strong stratification in near the TTL. Following the suggestion of the referee, we used a critical value of the modelled Brunt-Väisälä frequency (0.01 s^{-1}) as the threshold to split the different regimes (WGRID vs. WTKE). The simulated in-cloud ice number at T<205K also improves but not as much as that when we used the critical value of T (205K) to split the different regimes (WGRID vs. WTKE). We now included this new result in the manuscript. A smaller critical value of the modelled Brunt-Väisälä frequency (0.008 s^{-1}) can further decrease the simulated ice numbers but has no effect on the temperature dependence slope.

<u>Minor points:</u>

1. Missing references

In the introduction references about cirrus cloud distributions and properties are missing, especially new results from satellite evaluations. For instance, new global distributions of ice clouds could be derived from CALIPSO and CloudSat. Thus, it would be appropriate to include some new references, e.g. Stubenrauch et al. (2010) and Sassen et al. (2008). Concerning the issue of the net radiation effect of cirrus clouds, also some newer references should be included (e.g., Chen et al. 2000; IPCC 2013 report).

Answer: We now included these references.

2. Measurements of IN in the upper troposphere

In laboratory experiments the ability of different types of aerosols was investigated, especially the formation of ice crystals at glassy particles. However, in situ measurements of heterogeneous INs are difficult and especially the existence of glassy particles or precursors (e.g. organic material) is still not proven by in situ measurements and this should be mentioned in the introduction.

Answer: We added this information in the introduction:

"The ability of different types of aerosols acting as IN was investigated in laboratory experiments (e.g., the formation of ice crystals at glassy particles). However, in situ measurements of heterogeneous IN are difficult and especially the existence of glassy particles is still not proven by in situ measurements."

3. Mass accommodation coefficient

Skrotzki et al. (2013) showed that the mass accommodation coefficient should be in the range $0.1 \leq \alpha \leq 1$. Also model studies (e.g. Kay and Wood, 2008, see also references in Skrotzki et al., 2013) indicated that the low values as reported by Magee et al. (2006) are not representative for cirrus clouds. Please add some text to clarify this issue.

Answer: We now clarified this issue in the manuscript:

"Laboratory measurements support values from 0.006 (Magee et al. 2006) to unity (Skrotzki et al. 2013). Skrotzki et al. (2013) constrained the value in the range of 0.2-1 with majority of other lab studies also supporting a value > 0.1 (see Table 1 in Skrotzki et al. (2013)). Kay and Wood (2008) showed that α is $\geqslant 0.1$ for small ice crystals forming at high ice supersaturations and the α (=0.006) from (Magee et al., 2006) may only be appropriate for large ice crystals or at low ice supersaturations. In this study we will test two values of α (0.1 and 1)."

4. TKE scheme in upper troposphere

TKE schemes are included in GCMs in order to parameterize processes in the planetary boundary layer in a meaningful way. It is not clear what these schemes do in the free troposphere. Actually, it is even not clear that TKE schemes produce the correct kind of turbulence" in the upper troposphere at the right regions (e.g. at regions with low stability, strong shear etc.). Thus, the use of such schemes for parameterizing subgrid scale motion is quite questionable, although this kind of parameterizations is used often in many different models. The authors should comment on that issue and add some text in the manuscript, especially, since the model results indicate that the use of this parameterization does not provide meaningful results for ice clouds.

Answer: The TKE is diagnosed by CAM5's new moist turbulence scheme (Bretherton and Park 2009) which replaces the old dry turbulence scheme in its previous versions. Compared to the dry PBL scheme, the new moist turbulence scheme simulates cloud-radiation-turbulence interactions in an explicit way and is operating in any layers above as well as within PBL as long as moist Richardson number is larger than a critical value, 0.19. We added this information in the manuscript.

Regarding the use of TKE for parameterizing sub-grid scale motion, the manuscript already points out "*It is possible that CAM5 may overestimate the TKE in the upper troposphere at temperatures less than 205K, but an analysis of this is beyond the scope of this paper.***"**

5. Dominance of heterogeneous ice nucleation

The dominance of heterogeneous nucleation is still questionable. In fact, measurements from convective regions in the subtropics as reported in Cziczo et al. (2013) are certainly not representative for the whole upper troposphere and especially not for mid or high latitude conditions. Also the relevance of biological particles at cirrus level is not clear, since Pratt et al. (2009) could provide only one flight at about 7 kilometres (i.e. at temperatures T > 240 K), which is probably not representative for the whole upper troposphere. The authors should add some text, which makes clear that the importance of heterogeneous nucleation for the cold temperature regime (i.e. T < 235 K, which is mostly discussed in the manuscript) is still under discussion and not clear at the moment.

Answer: We added this in the manuscript:

"…and even the importance of heterogeneous nucleation for the cold temperature regime (i.e., T < 235 K) is still under discussion and not clear at the moment. "

"Yet measurements in Cziczo et al. (2013) are mainly from convective regions in the subtropics and are certainly not representative for the whole upper troposphere, especially not for mid or high latitude conditions. Also the relevance of biological particles at cirrus level is not clear, since Pratt et al. (2009) could provide only one flight at about 7 kilometres (i.e. at temperatures T > 240 K), which is probably not representative for the whole upper troposphere."

6. WGRID is only valid for strong stratification

In Spichtinger and Krämer (2013) a special kind of cirrus clouds in the tropical tropopause layer (TTL) was investigated. The dynamical regime for these cirrus clouds is characterized by very low vertical updrafts (w < 2 cm s^{-1}), by low temperatures (T < 205 K) and, most important, by strong strati_cation (i.e. high Brunt-Väisälä frequencies). The latter one is the key property for the investigated regime, leading to short nucleation events and thus low ice crystal number concentrations. For weaker stratifications this effect vanishes. Thus, the use of large-scale vertical velocities for the ice nucleation scheme is only meaningful for such strongly stratified regions (as recommended in the article by Spichtinger and Krämer, 2013). This issue should be clarified in the text (see also major point 2).

Answer: we added this in section 3.2:
"One caveat from above results is as the dynamical region studied in Spichtinger and Krämer (2013) has a very special condition (namely it is characterized by very low vertical updrafts (< 2 cm s-1), low temperatures (T<205K) and strong stratification (i.e. high Brunt-Väisälä frequency)) using the large-scale grid resolved updraft velocity may not be valid for weaker stratifications or when WGRID is not small enough."

7. WGARY seems to be wrong.
The results from the simulations suggest that the use of temperature fluctuations as parameterized by Gary (2006, 2008) does not produce meaningful results. Although this pathway of parameterization is suggested in many publications and is used in some GCMs, it should be stated more clearly in the manuscript that this parameterization.

Answer: From Fig. 4b, when the effects from vapour deposition onto pre-existing ice particles, a larger water vapour accommodation coefficient (α=1), and SOA IN are all included, using WGARY is able to produce the observed lower ice numbers. As showed in this study, there a lot of other uncertainties (i.e., mass accommodation coefficients, pre-existing ice effect, glassy SOA IN), so it may be pre-mature to come to the conclusion that WGARY should be used in a GCM as a choice for sub-grid updraft velocity.

Reference
Bretherton, C. S., and S. Park, A new moist turbulence parameterization in the community atmosphere model, J. Climate, 22, 3422–3448, 2009.