

We thank the reviewer for the constructive comments and suggestions for improving this manuscript. The reviewer's comments are in italics and our responses in standard font below.

**General Assessment:**

*In this work the authors improve the representation of ice cloud formation within the CAM model but including previously neglected factors accounting for preexisting ice crystals and subgrid variability in supersaturation. In line with previous works they find that including preexisting ice crystals decreases the ice crystal concentration therefore improving (however not completely fixing) a current bias of the CAM model. They also find that the contribution of heterogeneous ice nucleation to the total ice crystal production increases when accounting for preexisting ice crystals. Additionally the authors test the sensitivity of the anthropogenic indirect forcing to the choice of the ice nucleation parameterization. The study is relevant to the atmospheric community. However the discussion is confusing and the work is difficult to follow. The authors need to clearly state their objective and motivation behind the new implementation. The paper should be structured in a way that is easier to follow it can be published in ACP.*

Based on the reviewer's comments, we have revised the introduction section to clearly state our objectives and motivations behind the new implementation. We reorganized the model description (section 2) and the conclusion (section 6) in order to make the paper more clear. Please find our replies to the reviewer's comments below.

**General comments:**

*There are many grammatical and stylistic errors throughout the manuscript. I have tried to point out some but I'd recommend the authors to careful review the manuscript to make it suitable for ACP.*

**Reply:** Based on the reviewer's comments, all authors now paid special attention to spelling and grammatical checking, and did our best to correct the grammatical and

stylistic errors.

*The approach used in the work needs more justification. It is not clear why the authors implemented the PREICE parameterization. The motivation for inclusion of the subgrid probability distribution and the comparison of several ice nucleation parameterization is lacking.*

**Reply:** To address the reviewer’s comment, we rewrote the introduction section in the revised manuscript.

The purpose of considering PREICE was added as follows: “The presence of PREICE prior to ice nucleation events may hinder homogeneous and heterogeneous nucleation from happening owing to the depletion of water vapor on PREICE. Simulation results from ECHAM with the KL parameterization showed that the PREICE effects lead to cirrus clouds composed of fewer and larger ice crystals (Hendricks et al., 2011; Kuebbeler et al., 2014). Barahona et al. (2014) incorporated the BN parameterization into GEOS5, and modified the original BN parameterization to include the PREICE effects. Model results showed that cloud forcings are significantly reduced due to the effects of PREICE (Barahona et al., 2014)” (the fourth paragraph of section 1).

We clearly pointed out the motivations for inclusion of the sub-grid probability distribution and the comparison of several ice nucleation parameterizations in the introduction section of the revised manuscript: “Analysis of in situ data sets obtained in cirrus clouds found that ice saturation ratio,  $S_i$ , is highly variable both spatially (Jensen et al., 2013) and temporally (Barahona and Nenes, 2011), and that ice nucleation takes place only in a portion of cirrus cloud rather than in the whole area of cirrus cloud (Diao et al., 2013; 2014). However, most GCMs assume that cirrus cloud is homogeneously mixed, and ice nucleation event occurs in the whole area of cirrus cloud (Gettelman et al., 2010; Salzmann et al., 2010; Hendricks et al., 2011; Kuebbeler et al., 2014). Only until recently have GCMs attempted to account for the fraction of cirrus cloud where homogeneous freezing occurs ( $f_{hom}$ ) (Wang and Penner, 2010; Barahona et al., 2014; Wang et al., 2014)” (the fifth paragraph of section 1) and “Studies of anthropogenic aerosol indirect effects

showed that the annual global mean change in longwave cloud forcing from pre-industrial times to present-day estimated from CAM5 with the LP parameterization is 0.40–0.52 W m<sup>-2</sup> (Ghan et al., 2012), much larger than that from European Centre Hamburg model (ECHAM) with the KL parameterization (0.05–0.20 W m<sup>-2</sup>) (Zhang et al., 2012). Therefore, it is imperative to find out whether different ice nucleation parameterizations are the main cause for these differences” (at the end of the third paragraph of section 1).

*Section 2.3: This approach to include preexisting ice crystals seems simplistic and may contradict what is said in Section 2.4. What are the assumptions behind this approach? Do the authors use in-cloud or grid scale ice crystal concentration for their calculation? Do ice crystals sediment out of the cloud during a nucleation event? Since the cloud is not homogeneously mixed, one may argue that homogeneous nucleation occurs in cloud pockets devoid of ice crystals that will have high supersaturation (as assumed in Section 2.4). Maybe only the small ice crystals are transported with the cloud parcel is there a cut-off size for the crystals that affects homogeneous ice nucleation? (See for example Spichtinger and Kramer 2012 and Barahona and Nenes 2011)*

**Reply:** This approach to include preexisting ice crystals (PREICE) is based on the concept of Kärcher et al. (2006), which is derived from an adiabatic rising air parcel. The effect of preexisting ice crystals is to reduce the ice supersaturation in the rising air parcel by depositional growth, and is represented by reducing the updraft velocity driving the air parcel. In the development and application of ice nucleation parameterizations, the sedimentation of ice crystals out of the rising parcel is not considered during a nucleation event. This information has been included in the revised manuscript (in section 2.4 when we talk about the PREICE effect). For the PREICE effect on ice nucleation, the in-cloud ice crystal number concentration and mass-mixing ratio are used. In GCMs, it is assumed that the PREICE are uniformly distributed in the cirrus cloud, and thus the effect of PREICE on ice nucleation is considered only for the portion where ice nucleation occurs, since we assume that homogeneous nucleation takes place spatially only in a portion of

cirrus clouds rather than in the whole area of cirrus clouds (section 2.4). We note that the sub-grid variability of supersaturation (high supersaturation in a portion of the cloud) introduced in section 2.4 results from the temperature (vertical velocity) fluctuations rather than from the inhomogeneity of PREICE spatial distribution in cirrus clouds. We assume that cirrus clouds are homogeneously mixed after a nucleation event, so that PREICE are uniformly distributed in cirrus clouds. The corresponding sentences were added in the revised manuscript: “We note that cirrus clouds are assumed to be homogeneously mixed after a nucleation event” (at the end of new section 2.3) and “ice crystals are assumed to uniformly distributed in cirrus clouds” (in section 2.4 when we talk about the PREICE effect).

In the two-moment stratiform cloud microphysics scheme (Morrison and Gettelman scheme, hereafter MG scheme) in CAM5, ice crystals sedimentation is considered. We note that the ice nucleation process and the sedimentation are not solved together. Numerical splitting is used to treat the two processes separately. The LP, BN and KL ice nucleation parameterizations that were used in this study do not consider ice sedimentation during a nucleation event. Thus, the approach used for calculating PREICE effect also does not consider ice sedimentation.

*Figure 2 is confusing and can be done much better in a quantitative way showing the evolution of  $N_i$  in time.*

**Reply:** Following the reviewer’s comment, we modified Figure 2. Ice crystal numbers in new figure are derived from CAM5 model results that describe the cirrus cloud evolution within a model grid cell (3°N, 75°W, ~198 hPa, ~217K). We note that in GCMs different model grid cells can have different evolution of ice crystal number. Here, in order to illustrate the effect of pre-existing ice, we just show a schematic diagram of  $N_i$  evolution in cirrus clouds.

*Section 2.4: How does the variability in water vapor play a role defining the*

*supersaturation distribution? The model proposed assumes that only temperature fluctuations are important, however several studies have pointed out that their role is secondary. Furthermore, it is not clear whether the PDF is prescribed or changes with the grid cell conditions. The authors should explain exactly and very specifically how Eqs.(8) and (9) are used.*

**Reply:** The MG cloud microphysics scheme in CAM5 does not consider the variability in water vapor because the empirical basis for the underlying water vapor distribution functions and its impact on ice microphysics is not well understood (Morrison and Gettelman, 2008). In this study, in order to calculate the fraction of cirrus cloud where homogeneous freezing might occur, only in-cloud supersaturation ( $S_i$ ) variability from sub-grid scale vertical velocity (temperature fluctuations) is taken into account whereas sub-grid water vapor variability is neglected. We add a note in the revised manuscript: “We note that the in-cloud  $S_i$  variability due to the spatial variability of water vapor is not considered, which can be important based on recent studies (e.g., Diao et al., 2014)”.

Base on the work of Kärcher and Burkhardt (2008), the PDF of in-cloud  $S_i$  is a function of mean in-cloud temperature ( $T_0$ ), mean in-cloud ice saturation ( $S_0$ ) and temperature standard deviation ( $\delta_T$ ). In this study, we assume that  $T_0$  is equal to model grid mean temperature and  $\delta_T$  is applied to the whole grid area.  $S_0$  is assumed to be 1.0 because the water vapor deposition on ice crystals would remove supersaturation inside clouds with a long model time step (30 min). According to measurement-based analysis of Hoyle et al. (2005),  $\delta_T$  is linked to sub-grid scale vertical velocity turbulence ( $W_{sub}$ ), by  $\delta_T \cong 4.3W_{sub}$ . So we can derive  $\delta_T$  from  $W_{sub}$ . In CAM5, the  $W_{sub}$  is diagnosed from the square root of the turbulent kinetic energy (Bretherton and Park, 2009). After getting the  $\delta_T$ , we can derive the PDF of  $S_i$ . Based on the PDF of  $S_i$ , we can find out the  $f_{hom}(S_i > S_{hom})$ . Here, the threshold  $S_{hom}$  is a function of temperature (Kärcher and Lohmann, 2002a,b). This PDF of  $S_i$  was diagnosed based on grid cell conditions, such as  $T_0$  and  $\delta_T$  ( $W_{sub}$ ). So the PDF changes with the grid cell conditions. The corresponding paragraph was rewritten. We specifically explained this approach.

*Is the PDF connected at all to cloud condensate and cloud fraction? Otherwise it seems that the proposed approach may run into inconsistencies. The effect of defining different pdfs for different processes should be assessed.*

**Reply:** The PDF of  $S_i$  is not connected to cloud condensate or to cloud fraction. In CAM5, the ice cloud fraction is diagnosed using the total water (water vapor and cloud ice), based on Gettelman et al. (2010). The formation of ice cloud condensate (growth of ice crystals) is calculated using a relaxation timescale (Morrison and Gettelman, 2008; Gettelman et al., 2010). In other words, neither cloud condensate nor cloud fraction is treated based on PDFs. So we cannot assess the effect of defining different PDFs for different processes at this stage. We note that CAM5 is not able to consistently represent different cloud microphysics processes. For example, in the MG cloud microphysics scheme, cirrus cloud condensate and ice nucleation are calculated separately and sequentially. Actually, ice nucleation process should include the growth of newly-formed ice crystals (cloud condensate).

**Minor comments:**

*Page 17637 Line 1. Change import important.*

Done.

*Page 17637 Line 4. Remove “on other hand”.*

Done.

*Page 17637 Line 11. Remove “then”.*

Done.

*Page 17637 Line 13. Cirrus clouds is plural. Please correct.*

This sentence was removed base on the other reviewer’s comments. We checked all the

sentences that include “cirrus”, and make sure that “cirrus clouds” is plural.

*Page 17637 Line18. Please be more specific about what species are likely to be IN in the atmosphere. (DeMott et al., 2003; DeMott et al., 2011; Hendricks et al., 2011; IPCC, 2007, 2077)*

Done. The sentence (Page 17637 Line 18) was rewritten as follows: “Laboratory experiments and field observations show that various insoluble or partly insoluble aerosol particles can act as IN under cirrus formation conditions, such as mineral dust, fly ash, and metallic particles (DeMott et al., 2003; Cziczo et al., 2004; DeMott et al., 2011; Hoose and Möhler, 2012)”.

*Page 17637 Line22. Enigmatic refers to mysterious. Maybe just say “difficult to understand”.*

Done. “Enigmatic” was removed. The sentence was rewritten as follows: “Understanding the role of different aerosol types serving as heterogeneous IN remains challenging”.

*Page 17638 Line 6-10. This statement is confusing. Please rewrite.*

Done. This sentence was rewritten as follows: “Cziczo et al. (2013) analyzed the residual particle composition (after the ice was sublimated) within cirrus crystals of North and Central America and nearby oceans, and found that heterogeneous freezing was the dominant formation mechanism of these clouds”.

*Page 17638 Line 20-22. This is a badly formulated statement. Please rewrite.*

Done. This sentence was rewritten as follows: “A key component in cirrus cloud microphysics schemes is the ice nucleation parameterization that links ice number concentration to aerosol properties”.

*Page 17639 Line 12-15. This sentence literally comes from nowhere. The authors must justify why they think including a probability distribution of in-cloud supersaturation is required.*

This sentence was removed because we reorganized and rewrote the introduction section. We added a paragraph (the fifth paragraph of section 1) explaining the motivation of including a probability distribution of in-cloud variability.

*Page 17639 Line 12-15. Is this actually in-cloud or it applies to the whole grid cell? What is done for the cloud-free part of the cell.*

This is in-cloud. In CAM5 ice nucleation event is not directly related to cloud fraction (cloud cover), which is diagnosed using the total water (water vapor and cloud ice). In the MG cloud microphysics scheme, only the ice number concentration (in-cloud) was calculated by the ice nucleation parameterization. So we just focus on the ice nucleation process for the cloudy portion.

*Page 17639 Line 15-16. Again, why is it important to compare different parameterizations?*

We added the motivation for comparison between different parameterizations in the third paragraph of section 1.

*Page 17639 Line 23. Say: the version 5.3 of the Community Atmospheric Model.*  
Done.

*Page 17639 Line 24. What does highly parameterized mean?*

Here, “highly parameterized” means “with simplified cloud microphysics”. We added



this explanation.

*Page 17642 Line 5-10. Neither  $S_i$  or  $q_{i,pre}$  are constant during the parcel ascent. What are the assumptions behind Eq.(6) ? This particular approach is not new and has been proposed before (Numerically by Kärcher et al. 2006 and analytically by others). The authors should cite previous works.*

Eq.(3-6) present the evolution of  $S_i$  in an adiabatic rising air parcel with PREICE. In ice nucleation parameterization, ice number from homogeneous freezing is calculated based on sulfate number concentration, grid-mean temperature, and updraft velocity. The corresponding  $W_{i,pre}$  is calculated at the homogeneous freezing saturation threshold ( $S_{hom}$ ) and the properties of PREICE from previous timestep ( $n_{i,pre}$ ). Here, we neglect the change of PREICE size ( $R_{i,pre}$ ) from the beginning of the current timestep to the occurrence of homogeneous freezing event. Correspondingly, some sentences were added at the end of this paragraph:

“We need the  $W_{i,pre}$  for the ice nucleation parameterization. In the LP ice nucleation parameterization, ice number produced from the homogeneous freezing is a function of temperature, sulfate number concentration, and updraft velocity. To calculate the corresponding  $W_{i,pre}$ ,  $S_{hom}$  is used in Eqs.(7-8) (that are Eqs.(5-6) in the discussion paper). The  $n_{i,pre}$  and  $R_{i,pre}$  in Eq.(8) indicate the number concentration and radius of in-cloud PREICE, respectively, from the previous time step. The  $W_{i,pre}$  used for heterogeneous nucleation is calculated based on the same approach, except that  $S_i$  in Eqs.(7-8) is replaced by the heterogeneous freezing saturation threshold ( $S_{het}$ ).”

As mentioned at the beginning of this subsection, our approach is based on the work of Kärcher et al. (2006). The reference “(Kärcher et al., 2006)” was added again here.

*Page 17642 Line 20-25. This paragraph is confusing. Please rewrite.*

This sentence was rewritten as follows: “The most distinct feature of this figure is that

$W_{i,pre}$  is proportional to the PREICE number concentration. When the PREICE number concentration is greater than  $50 \text{ L}^{-1}$  and  $W$  less than  $0.2 \text{ m s}^{-1}$ , the black dotted line (for homogeneous freezing and PREICE radius of  $25 \text{ }\mu\text{m}$ ) indicates that homogeneous freezing can not occur, because  $W_{i,pre} > W$ .

*Page 17643 Line 2. Using the effective radius as defined Morrison and Gentleman (2008) is incorrect (i.e, The third over the second moment of the distribution). To be consistent with Eq.(6)  $R_{i,pre}$  must represent the mean volumetric radius (i.e, the first moment of the size distribution).*

In the revised manuscript, we clearly point out that the effective  $R_{ieff,pre}$  is obtained by using the first moment of ice particle size distribution. The corresponding sentence was added as follows: “The  $R_{ieff,pre}$  is obtained directly by using the first moment of ice particle size distribution. We note that this  $R_{ieff,pre}$  is different from the effective radius used in the radiative transfer scheme which is calculated from the third and second moments of size distribution.”

*Page 17644 Line 17. It is not clear what this means. How is the PDF used exactly (also how does it look like)? Do the authors multiply  $N_i$  by  $P_T(S > S_{hom})$  as done by Kärcher and Burkhardt (2008)? Please be more specific.*

In the default CAM5.3, it is assumed that cirrus cloud is homogeneously mixed. Thus, homogeneous nucleation event is assumed to occur in the whole area of cirrus clouds. Due to the in-cloud variability in ice saturation ratio, homogeneous nucleation takes place spatially only in a portion of cirrus clouds rather than in the whole area of cirrus clouds. In this study, the fraction of cirrus clouds where homogeneous freezing occurs is calculated based on the PDF of in-cloud variability in ice saturation ( $S_i$ ) that is caused by temperature fluctuations.

The sentence (Page 17644 Line 17) was rewritten as follows: “we assume that homogeneous freezing takes place spatially only in the portion of cirrus clouds ( $f_{hom}$ )

where in-cloud  $S_i > S_{hom}$ .”

More explanation of  $f_{hom}$  was added in the introduction section in the revised manuscript. We also specifically explained about how the PDF of  $S_i$  is used in section 2.3. We now added the sentence: “The PDFs of  $T'$  and  $S_i(T')$  can be found in Fig. 3 of Kärcher and Burkhardt (2008).”

$N_i$  calculated from the homogeneous freezing is multiplied by  $f_{hom}$ . We now added a sentence to clarify this: “Because the ice number concentration after an ice nucleation event indicates the in-cloud value, the ice number concentration calculated from homogenous freezing parameterization is multiplied by  $f_{hom}$ ”.

*Page 17645 Line 13. It must be nucleation spectra.*

Done. All “nucleation spectrum” was replaced by “nucleation spectra”.

*Page 17646 Line 1. It is not clear what this means. As expressed in Eq. (5),  $W_{pre}$  is independent of the ice nucleation parameterization. What value of  $S_i$  has been used to calculate  $W_{pre}$ ?*

$W_{i,pre}$  is independent of the ice nucleation parameterization. This sentence was now removed.

The  $W_{i,pre}$  used for homogeneous nucleation is calculated based on the homogeneous freezing saturation threshold ( $S_{hom}$ ). The  $W_{i,pre}$  used for heterogeneous nucleation is calculated based on the heterogeneous freezing saturation threshold ( $S_{het}$ ). We have made this clear in the revised manuscript (section 2.4).

*Page 17646 Line 5. It should be: the parameter that sets.*

Done.

*Page 17646 Line 18-20. Remove the sentence starting with the Default...*

Done.

*Page 17647 Line 20. It should be upper limit.*

Done.

*Page 17647 Line 5-20. The aircraft measurements correspond to a scale much smaller than the GCM resolution. Averaging over a 50 Km grid would be incorrect and contradict the assumption that homogeneous ice nucleation occurs only in a fraction of the grid cell. According to that assumption, the grid scale average vertical velocity cannot be representative of the conditions where nucleation is actually occurring.*

We agree with the reviewer that the aircraft measurements correspond to a scale much smaller than the GCM resolution. As discussed in Zhang et al. (2013), since the GCMs represent statistics at much larger scale, it is difficult to compare directly the GCM results with the in situ aircraft data. Therefore, for a fair comparison, following Zhang et al. (2013), aircraft data are averaged over a 50 km grid to derive the statistics of measured vertical velocity.

It is true that the GCM grid average vertical velocity cannot be representative of the conditions where nucleation is actually occurring. It should be clarified that we didn't use the grid average vertical velocity to compare with the aircraft measurements. Instead, the GCM parameterized characteristic sub-grid updraft velocity ( $W_{sub}$ ), which is an input variable for the ice nucleation scheme, is used to compare with the aircraft measurements. For the aircraft measurements, only the updraft portion is counted to get the mean updraft velocity over a 50km grid.

*Page 17649 Line 22. Would  $f_{hom}$  decrease at low temperature (i.e, high altitude) since total water is also decreasing? Is this accounted for?*

As mentioned in section 2,  $f_{hom}(S_i > S_{hom})$  depends on the PDF of in-cloud  $S_i$ . The PDF of  $S_i$  depends mainly on the diagnosed  $W_{sub}$ . Model results showed that  $f_{hom}$  increases at the tropical tropopause layer (low temperature) since the  $W_{sub}$  is larger there.

In CAM5, the ice cloud fraction is diagnosed using the total water. If the model grid cell is totally cloud-free,  $f_{hom}$  is set to zero.

*Page 17650 Line 7-10. Please explain what the reason for the better agreement is.*

The reason was added. These sentences were rewritten as follows: “Compared to the Default experiment, the Preice and Nofhom experiments predict higher  $N_i$  and show better agreement with observations in this temperature range. As discussed above, the main reason is that the two unphysical limits are removed.”

*Page 17655 Line 3-10. This is a misrepresentation of previous work. The KL and BN parameterizations include transition regimes where heterogeneous freezing is active but not enough to prevent homogeneous ice nucleation. Thus newly formed crystals come from both homogeneous and heterogeneous ice nucleation. This paragraph implies that other parameterizations to from complete homogeneous and that only the LP parameterization has such feature, which is not true. The paragraph must be removed.*

Following the reviewer’s comment, we removed this paragraph in the revised manuscript.

*Page 17657 Line 15-17. It must be mentioned that only variations in supersaturation from temperature fluctuations are taken into account whereas water vapor variability is neglected. Including the latter may lead to a much stronger effect and coupling between different nucleation events.*

We agree with the reviewer’s comment. This is mentioned in the revised manuscript: “We note that only in-cloud  $S_i$  variability resulting from the sub-grid scale temperature fluctuation is taken into account in this study whereas the sub-grid water vapor variability is neglected. Including the latter may lead to a much stronger effect and coupling between different nucleation events”.