

Dear Dr Krämer,

On the behalf of all the authors, we submit a revised version of the manuscript titled “Effects of preexisting ice crystals on cirrus clouds and comparison between different ice nucleation parameterizations with the Community Atmosphere Model (CAM5)” for your consideration of publication in ACP.

We have responded to all reviewers’ comments and revised the manuscript accordingly. The point-by-point reply to the reviews has been uploaded to the interactive discussion webpage and is attached below. The marked-up manuscript was uploaded as “Manuscript” file. Major changes to the original manuscript were marked in blue.

We hope you find the revision satisfactory and look forward to your favorable decision.

Sincerely,

Xiaohong Liu

Replay to Anonymous Referee #1

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:

The manuscript investigates the effect of preexisting ice crystal on cirrus clouds and its climatic implications. Also the manuscript describes the updated cirrus scheme in CAM5, namely the consideration of in-cloud variability in ice saturation ratio as well as the removal of two unphysical limiters. The manuscript deals with current open issues in the field of cirrus cloud science, adds valuable knowledge and is of high scientific interest. So, I strongly recommend publishing the manuscript in ACP. However, I fear that the written language and the structure of the manuscript need some improvement before publishing. There are several spelling and grammatical errors all over the manuscript and I could not take the effort and mention them all here in my review. I encourage all authors to take a close look to that when revising the manuscript. Also, I think parts of the results (the comparison between different ice nucleation parameterizations) need some deeper investigation rather than speculations. Since the data/information is given in current literature this is possible to do. My more detailed comments and suggestions can be found below.

Based on the reviewer's comments, we corrected the spelling and grammatical errors and improved the manuscript structure. We also made further analysis to understand the differences between different ice nucleation parameterizations.

Major comments:

Page 17637, Line: 14: I think this needs to be rewritten, since it is not totally true that cirrus cloud knowledge is still in its infancy. I think in recent years big steps had been

made forward in both, cirrus cloud measurements as well as cirrus cloud modeling. Please add this and give adequate citations.

Reply: Following the reviewer's comment, the sentence was rewritten and related citations were added. The new sentence now reads as: "In recent years, significant progress has been made in both cirrus cloud measurements and cirrus cloud modeling (e.g., Heymsfield et al., 2005; Krämer et al., 2009; DeMott et al., 2011; Cziczo et al., 2013; Jensen et al., 2013; Diao et al., 2014; Barahona and Nenes, 2011; Jensen et al., 2012; Spichtinger and Krämer, 2013; Murphy, 2014)."

Section 2: Section 2 is not well structured and thus repeats information and/or is difficult to understand. Thus I recommend to 1) change the order of the subsections, 2) use slightly different titles for the subsections and 3) delete some of the redundant repeating information. Detailed information on this issue can be found below (Page 17639-17646):

Page 17639-17640: Subsection 2.1. I suggest to rename this subsection and to put parts of this subsection to 2.2, since descriptions of the cirrus scheme in CAM5 and the ice nucleation parameterization in CAM5 are very close and don't need different subsections. In order to have 2 separate subsections I suggest to have a subsection 2.1 called "CAM5" and 2.2 called "cirrus cloud scheme in CAM5". I further suggest to put the text between page 17640 line 6 and line 16 to subsection 2.2.

Page 17644: Subsection 2.4: It is totally confusing to explain small modifications to the base model after introducing the concept of PREICE, which is the main topic of this study. So, I suggest swapping subsection 2.3 and 2.4. This means of course that the text needs to be changed accordingly. I further suggest to rewrite text on page 17641, line 15-17 as follows: "To account for the effect of preexisting ice we introduce preexisting ice into our model CAM5 based on the concept of Kärcher et al., 2006, which is based on the concept of an adiabatic rising air parcel."

Reply:

- 1) Based on the reviewer's comments, the structure of Section 2 was changed: Subsection 2.1 titled "CAM5" introduces cloud schemes, aerosol scheme, and radiation scheme in CAM5. Subsection 2.2 titled "Cirrus cloud scheme in the standard CAM5" describes the cirrus scheme and ice nucleation parameterization in CAM5. Redundant repeating information was removed.
- 2) We swapped subsection 2.3 and 2.4. The text was modified accordingly. For example, the first paragraph in the new subsection 2.3 was rewritten as follows: "In this study, several modifications have been made in the ice nucleation scheme in CAM5. First, the effect of PREICE is taken into account (subsection 2.4). Second, ...". In the new subsection 2.4, the text on page 17641 line 15-17 was replaced by "To account for the effect of preexisting ice (PREICE) we introduce PREICE into CAM5 based on the concept of Kärcher et al. (2006)".

Page 17643: Description of Figure 2: I don't understand how you derived these numbers? These numbers should be dependent on vertical velocity, temperature, supersaturation. . . Please add this info and explain in more detail.

Reply: In the discussion paper, the numbers in Figure 2 are used for illustration purpose only. In the revised manuscript, we modified Figure 2 with ice crystal numbers derived from CAM5 model results that describe cirrus cloud evolution within one model grid cell (3°N, 75°W, ~198 hPa, ~217 K). In this experiment, the updraft velocity is set to 0.2 m s⁻¹, and the sulfate number concentration is set to 100 cm⁻³. Heterogeneous nucleation is not taken into account. The simulation is run 3 months. Just one cirrus cloud evolution process is shown here.

Page 17653-17654: The differences in the effects of PREICE between different models (CAM5, GEOS5, ECHAM5) but using the same ice nucleation parameterization are very large. I think this needs some more detailed investigation and explanation. You speculate that the input parameters like Wsub, RH_i and aerosol number concentration used to drive

the ice nucleation parameterization are different in the above-mentioned models. In Kuebbeler et al., 2014 aerosol number concentrations are presented, thus a comparison with that data is possible. Also, how are processes influencing the ice number concentration like sedimentation, accretion, aggregation, etc. realized in the different models? Besides, what is about freezing details like critical supersaturation for heterogeneous freezing? Which freezing mechanisms (immersion, deposition) are used in the models? All these can influence your results and should be investigated/discussed in more detail.

Reply: Following the reviewer's comment, we made additional analysis and add more discussions on why the simulated PREICE effects between CAM5, GEOS5, and ECHAM5 are different.

With the same BN ice nucleation scheme, the PREICE effect in CAM5 is stronger than that in GEOS5. In GEOS5, the heterogeneous nucleation mechanisms include immersion and deposition ice nucleation on dust, black carbon, and soluble organics (Barahona et al., 2014). The global mean ice number concentration (N_i) produced from the heterogeneous nucleation and its percentage contribution to the total N_i are $\sim 22 \text{ L}^{-1}$ and $\sim 30\%$, respectively (see Fig. 7 in Barahona et al., 2014). In the CAM5.3 simulation with BN, N_i from the heterogeneous immersion nucleation on the coarse mode dust and its percentage contribution to the total N_i are 5.1 L^{-1} and 9.4% , respectively, which are significant lower than that from GEOS5. As a result, in CAM5.3 there are less IN competing with the homogeneous ice nucleation and PREICE has a relatively larger impact. This might be the main reason why the PREICE effect from CAM5.3 with BN is stronger than GEOS5.

With the same KL scheme, we see a stronger PREICE effect in CAM5 than in ECHAM5-HAM. In the model used in Kuebbeler et al. (2014), the ice nucleation process requires that the model grid is supersaturated with respect to ice ($RHi > 100\%$). Furthermore, the depositional growth of ice crystals is treated based on the model grid RHi . If a model grid is supersaturated and a large number of PREICE is present, the

depositional growth of these PREICE will remove the supersaturation in the grid, and thus prevents the following ice nucleation. In other words, the ice deposition growth (the cirrus clouds scheme condenses mass on PREICE) in ECHAM5 also takes into account the effect of PREICE by removing the supersaturation. However, in CAM5 the treatment of deposition growth of ice crystals only relaxes the grid RHi towards 100%, and does not remove the supersaturation well below 100% (Morrison and Gettelman, 2008). In other words, the ice deposition growth process cannot prevent the ice nucleation process from happening. As compared to ECHAM5, ice nucleation processes frequently occur in CAM5. Thus, the effect of PREICE during the ice nucleation process, which is represented by reducing the updraft velocity driving ice nucleation parameterization, is weaker in ECHAM5.

We have also compared the number concentrations of heterogeneous IN, sulfate particles and ice crystals presented in Kuebbeler et al. (2014) with those from CAM5. Freezing mechanisms (immersion, deposition) in GEOS5 and ECHAM5 are also discussed. The corresponding text can be found in the third paragraph of section 4.

Page 17655, line 26 to end of page: Numerous laboratory based studies have shown that not all dust particles can act as IN and thus an upper limiter for heterogeneously freezing particles does indeed make sense. As far as I know, only Hendricks et al., 2012, Kuebbeler et al., 2014 and some studies looking at the potential of soot as IN in cirrus clouds have considered this. I would be curious to see how your results change if you also put an upper limiter to dust particles in your model. Also I assume that this might indeed be the reason why the results differ so strongly between models. However, as already mentioned above you could easily check this, when plotting aerosol data of your model using KL parameterization and comparing to Kuebbeler et al., 2014.

Reply: The default CAM5 uses dust in the coarse mode as potential heterogeneous IN and there is no upper limiter applied. In the model it is assumed that dust is internally mixed with sulfate, and thus only immersion freezing of dust particles is considered.

Furthermore, soot contribution to the heterogeneous IN is turned off. Therefore, it is assumed that all dust in the coarse mode can act as IN (immersion freezing) in the default CAM5. In CAM5, the global averaged heterogeneous IN (i.e., dust in the coarse mode) number concentration in cirrus clouds is 10.4 L^{-1} . 71% of IN number occurs in the range of $1-10 \text{ L}^{-1}$, 25% of IN number in the range of $10-100 \text{ L}^{-1}$, and 3% of IN number in the range of $>100 \text{ L}^{-1}$. In this study, we do not focus on dust or soot effect on cirrus clouds. So we didn't modify the default dust number concentration used in the heterogeneous nucleation parameterization. To compare different ice nucleation parameterizations under the same aerosol condition, for BN and KL parameterizations, we also used all coarse mode dust (no upper limit) as the potential heterogeneous IN.

We note that, our earlier study (Zhang et al., 2013) using the standard CAM5 model with LP and BN parameterizations has studied the influence of the upper limiter (f_a) to dust IN. Results from the BN parameterization with $f_a=100\%$ are similar to those from the LP parameterization ($f_a=100\%$). With a larger f_a (100% versus 5%), not only are more crystals produced by heterogeneous nucleation but also the homogeneous nucleation is suppressed and contributes considerably less to the total ice crystal production. In the Northern Hemisphere, where the main sources of dust aerosols are located, a larger f_a (100% versus 5%) leads to considerably less ice crystals. That study used the standard CAM5 without considering the preexisting ice effect.

Following the reviewer's comment, we performed a simulation using the updated CAM5 model with the BN parameterization and applied an upper limiter (5%) to the coarse mode dust particles. Model results show that the change in total ice number is moderate because of the low number concentration of coarse mode dust ($<10 \text{ L}^{-1}$) used to drive the ice nucleation parameterization. This suggests the limiter to the coarse mode dust might not be the main reason why the results differ so strongly between the models (CAM5 with BN versus GEOS5 with BN). We note that the contribution from heterogeneous ice nucleation to total ice crystal number concentration is $\sim 30\%$ in GEOS5, while it is only 9.4% in CAM5.3. Based on our sensitivity tests, the effect of PREICE increases with

increasing contribution from the homogeneous nucleation. This might be the main reason why the PREICE effect in CAM5.3 (with a higher homogeneous nucleation contribution) is stronger than GEOS5.

Page 17657-17658: Discussion and conclusions: Conclusions should shortly describe what was done in the manuscript and review the most important findings of this study. To me, this sections is again not well structured. I suggest rewriting this section in a way that each paragraph describes one important finding. (In more detail, I suggest to put text from page 17657, line 8-12 as well as from page 17568, line 8-13 to the last paragraph since it all deals with findings of the comparison between different ice nucleation schemes.)

Reply: Following the reviewer's comment, we reorganized this section. The model modifications (for PREICE and f_{hom}) and evaluation are discussed in the first paragraph. The PREICE effect on the relative contribution of homogeneous nucleation versus heterogeneous nucleation is discussed in the second paragraph. The comparison between three ice nucleation parameterizations is discussed in the last paragraph. We moved the text from page 17657, line 8-12 as well as from page 17658, line 8-13 to the last paragraph.

Page 17657-17658: Discussion and conclusions: I think one very interesting finding of our study is the fact that the 3 different parameterizations agree surprisingly well in the representation of N_i and the contribution from heterogeneous ice nucleation to the total ice nucleation. However, BN and KL parameterizations used in CAM5 give strongly different results than BN and KL in GEOS5 and ECHAM5. So, probably these results are more driven by input parameters (w, T, RH_i) and the assumptions of aerosol distribution (immersion vs deposition freezing, size of aerosols, etc.). I think this is an interesting results and should be stressed more here.

Reply: Following the reviewer's comment, this finding is stressed more in the conclusion

section, such as “The differences among this study, Barahona et al. (2014) and Kuebbeler et al. (2014) can be more driven by differences in meteorological input parameters (W , T , RHi), the assumptions of aerosol distribution (immersion versus deposition freezing, aerosol characteristics, etc.), and the methodology of parameterization implementation, than ice nucleation parameterizations themselves.”

Minor comments:

Page 17636, Line: 24: Please include “the”: As a result, the experiment. . .

Done.

Page 17637, Line: 19: Please include the following reference: Hoose and Möhler, Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, Atmos. Chem. Phys., 12, 9817-9854, 2012

Done.

Page 17638, Line 3: “reduces” rather than “reduce”

Done.

Page 17638, Line 4: you could add here some more detailed information, such as: If homogenous nucleation is prevented totally from occurring or if the rate of homogeneously nucleated ice crystals is reduced depends on several parameters, such as number of aerosols, supersaturation, temperature, vertical updraft.

Following the reviewer’s comment, we added some new text regarding this issue: “If homogenous nucleation is prevented totally from occurring or how the rate of homogeneously nucleated ice crystals is reduced depends on several parameters, such as number of heterogeneous IN, temperature, vertical updraft (Liu and Penner, 2005;

Kärcher et al., 2006; Barahona and Nenes, 2009)”.

Page 17638, Line 13: I think the proper reference here is rather Hendricks et al 2011 than Lohmann et al., 2008. Please replace or add that.

Done. We added “Hendricks et al., 2011”.

Page 17638, Line 15: Please add the latest IPCC report here.

Done. We added “IPCC, 2013”.

Page 17638, Line 29: Please use “may hinder” instead of “hinders”

Done.

Page 17639, Line 2-8: Since you are using the KL and BN parameterizations in your study I think these two references (Kuebbeler et al., 2014 and Barahona et al., 2013) can be explained in a little more detail here.

A detailed explanation about these two references (Kuebbeler et al., 2014; Barahona et al., 2014) will be given in section 2.5 and section 4. Here in this paragraph, we just focus on the effect of PREICE. We note that Barahona et al. (2013) has been published in GMD this year. So “Barahona et al., 2013” was changed to “Barahona et al., 2014”.

Page 17639, Line 8: Please give the according reference here.

Done. We added the according reference “Barahona et al., 2014”.

Page 17639, Line 11-13: I don't understand this sentence. Please explain in a little more detail.

This sentence was removed here. The detailed explanation about “occurrence probability of homogeneous freezing events in cirrus clouds” was added in the previous paragraph: “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).”

Page 17640, Line 3: Please remove “also”.

Done.

Page 17640, Line 20: Why do you neglect deposition freezing? Several studies have shown that this is the more efficient ice nucleation mechanism.

We agree with the reviewer that deposition nucleation on pure dust particles can be an efficient ice nucleation mechanism. In CAM5, dust is internally mixed with sulfate in the aerosol treatment. Thus, there is only immersion freezing of dust particles considered in the standard CAM5. In this study, we use the default heterogeneous nucleation mechanism in CAM5.

Page 17641 Line 3-7: I don't understand this sentence. Please explain. Page 17641 Line 8: What does aai stand for in Naai?

Cloud droplet activation usually happens at the cloud base or for the newly-formed

clouds at every cloud level, as represented in CAM5. In comparison, ice nucleation can happen inside preexisting cirrus clouds, because relative humidity with respect to ice (RHi) up to or even more than 120% are frequently observed inside cirrus clouds (Krämer et al., 2009). To make the sentences (Page 17641 Line 3-7) clearer, we modified them as “The cloud droplet activation in warm liquid-phase clouds only occurs at the cloud base of preexisting clouds or in all levels of newly-formed clouds, as represented in CAM5. In contrast, ice nucleation is allowed to happen in all levels of preexisting cirrus clouds in CAM5 if the nucleation thresholds are met because RHi up to or even more than 120% are frequently observed inside cirrus clouds (Krämer et al., 2009).”

The “ aai ” in “ N_{aai} ” stands for the aerosol activation to form ice crystals (i.e., ice nucleation) in the cirrus microphysics scheme. We note that ice crystals can also come from the convective detrainment in CAM5.

Page 17641 Line 8: What does “current” mean? Ice crystals from previous timestep? Ice crystals from other sources?

The “current” indicates ice crystals from previous time step. The corresponding sentence was changed to “New ice crystals will be produced if the in-cloud ice number concentration, N_i , from the previous time step falls below N_{aai} ”.

Page 17641 Line 25: There seem to be problems with many of the equations. There are weird symbols in equation 1,3,7,8,9.

All equations are double-checked. The equation editor in Microsoft Word 2013 (for Mac) is used for this manuscript. Using Preview (Mac PDF viewer), formulas look normal.

Page 17646, Line 7: what is f_{hom} ? It wasn't introduced yet.

The “ f_{hom} ” has been introduced in previous subsection 2.4 “Thus, we can find out the fraction of cirrus cloud, f_{hom} , where ...”. In the revised manuscript, the “ f_{hom} ” was

mentioned several times in previous section 2.3. For clarity, we changed this sentence to “The f_{hom} used for the LP parameterization, as discussed in subsection 2.3, is also used for BN and KL parameterizations”.

Page 17647, Line 17: I suggest to rewrite the part “, there are no W_{sub} data larger than 0.24 m s^{-1} ” as follows: “the cut-off in Default is not exactly 0.2 m s^{-1} but 0.24 m s^{-1} .”

Done. The sentence was changed as suggested.

Page 17648, 1st paragraph: To me, NoPreice fits observations best with only a small shift towards too high ice number concentrations. Can you comment why?

Compared to the Preice and Nofhom experiments, the occurrence frequency of higher N_i ($>100 \text{ L}^{-1}$) from the NoPreice experiment is increased significantly. It seems that NoPreice fits observations best. We note, however, that the observed N_i is from in situ measurements, while the modeled N_i represents the averages over the whole area of cirrus clouds within a model grid cell ($\sim 100 \text{ km}$). In addition, although measurements during the SPARTICUS campaign have significantly reduced the shattering of ice crystals, it is unclear whether the very high N_i ($>1000 \text{ L}^{-1}$) is affected by the shattering artifact. When considering these issues, it is difficult to judge that NoPreice fits observations best. We have added these notes in the revised manuscript.

Page 17649, Line 18: So far as I know, most models have problems getting the correct trend of N_i with T suggested by Krämer et al., 2009. I think here it should be mentioned more clearly that the modeled trend of N_i tending to increase with decreasing temperature is the contrary of what is observed.

Following the reviewer’s comment, we more clearly pointed out that the modeled trend of N_i tending to increase with decreasing temperature is the contrary of what is observed.

Page 17650, Line 9-11: I think here you are a bit too optimistic about the performance of Preice. To me, Nofhom is as good as Preice.

Nofhom agreed better with observations as compared to Preice. The sentence (Page 17650, Line 9-11) just focused on the comparison between updated CAM5.3 and Default CAM5.3. Following the reviewer's comment, this sentence was replaced by "the Preice and Nofhom experiments show better agreement with observations ...".

Page 17652, Line 18-19: I suggest to rewrite "equals to W_{sub} minus $W_{i,pre}$ " as follows: " $(W_{eff}=W_{sub}-W_{i,pre})$ "

Done.

Page 17653, Line 5-8: Please rewrite as follows:" NoPreiceBN, PreiceBN, NoPreiceKL and PreiceKL experiments are also analyzed, but not shown here, because the effects of PREICE from experiments using BN and KL parameterization are similar.

Done. The sentence was rewritten based on the reviewer's comment.

Page 17654, Line 23-25: You write ". . . N_i is reduced in low-level cirrus. . .". I don't fully understand what you mean; reduced compared to what? Please be more specific.

We rewrote the sentence as follows: "One distinct feature of N_i distribution patterns from these experiments is that N_i reduces towards lower altitudes".

Page 17656, Line 6-8: To me, changes in CDNUMI of KL are only smaller than LP and BN between 30-60N. In regions larger than 60N or smaller than 30N all parameterization are rather similar. Please be more specific here.

We rewrote the sentence as follows: " Δ CDNUMI from the PreiceKL experiment is

smaller between 30°-60° N as compared to other experiments. In regions higher than 60° N or lower than 30° N, all experiments are rather similar”.

Page 17656, line 11-13: Why are changes in CDNUMI in Preice between 60-80N of opposite sign than the other experiments? This should be mentioned and explained here.

This was mentioned and explained as follows: “ Δ CDNUMI from the Preice experiment between 60°-80° N (negative) has the opposite sign than the other experiments (positive). However, these changes are generally within the ranges of two standard deviations”.

Page 17656, line 20: It looks like changes in IWP are not statistically significant. Can you comment on this?

We added more comments on this as follows: “Compared to Δ CDNUMI, the fluctuation of Δ IWP is more complicated because many other microphysical processes (especially in mixed phase clouds) can impact Δ IWP. Furthermore, the changes in cloud properties caused by aerosol indirect effects may modulate atmospheric circulation and water vapour transport, and then impact IWP in other regions. Thus, Δ IWP from all experiments are not statistically significant”.

Page 17657, Line 8-9: This sentence is confusing. Make clear, what is same and what is different between the 3 parameterizations.

The method used for calculating $W_{i,pre}$ in the LP parameterization is exactly same as the KL parameterization. Also, the PREICE effect in the BN parameterization is considered by the same concept of KL parameterization. Our box model calculations show that $W_{i,pre}$ calculated from BN and KL parameterizations are very similar under the same condition. This sentence (now in the third paragraph of section 6) was rewritten as follows: “Both LP and BN parameterizations consider PREICE effects based on the concept of KL parameterization”.

Page 17568, Line 11-13: please explain why.

We added the explanation why there are large differences in the PREICE effect between CAM5 and GEOS5 (now in the third paragraph of section 6).

Figures:

Figure 2: The clouds look rather like cumulus clouds than like cirrus clouds. I strongly suggest to change that.

Done. We changed that. Cirrus clouds are now indicated by ovals.

Figures 3,4,5,9: The colors of the two experiments Default and NoPreice are not distinguishable on printed paper. Please use a different color for one of both.

Done. The color of NoPreice was changed to orange.

Figure 9: You are presenting changes (!) of the variables LWCF, SWCF, CDNUMI and IWP. You are missing a delta sign in the upper left corner of each of the four plots in front of the variable.

Done. A delta sign was added in front of the variable.

Replay to Anonymous Referee #2

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⁻² (Ghan et al., 2012), much larger than that from European Centre Hamburg model (ECHAM) with the KL parameterization (0.05–0.20 W m⁻²) (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 (δ_T). In this study, we assume that T_0 is equal to model grid mean temperature and δ_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), δ_T is linked to sub-grid scale vertical velocity turbulence (W_{sub}), by $\delta_T \cong 4.3W_{sub}$. So we can derive δ_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 δ_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 Line13. 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.

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 Karcher 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 L^{-1} and W less than 0.2 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".