

Technical Note: assessing predicted cirrus ice properties between two deterministic ice formation parameterizations (egusphere-2022-1057)

Colin Tully, David Neubauer, and Ulrike Lohmann

Referee #1 Author Response

To Referee #1,

Thank you for taking the time to review our manuscript and for providing useful comments on improving this study. We have quoted each of your general and specific comments below with our response and changes in the text where applicable. We omitted minor changes like typos and word removals. In some cases, our responses were linked to more than one of your comments, which we note below.

Sincerely,
Colin Tully (on behalf of all co-authors)

General Comments

1. **Comment:** There are a few contradictions in this manuscript that need to be addressed. Kärcher and Marcolli (2021) motivated the differential AF approach by arguing that the cumulative AF approach may overpredict heterogenous ice nucleation, yet the box model results featured here show that when ICNC does not agree, the cumulative AF approach underestimates ice nucleation compared to differential AF. The box model results also show that predicted ICNC between the two schemes frequently agree (and at worst show a discrepancy of $< 2X$ in extreme conditions, looking at Fig. 2), yet an argument is made later in the manuscript when explaining impacts of KM21 on cirrus simulated in ECHAM-HAM that this ice nucleation parameterization often increased ICNC compared to ML20. Also, though KM21 sometimes increased ICNC in the box model results, it decreased zonal mean heterogeneously nucleated ICNC compared to ML20 over much of the SH and NH. Perhaps these issues could be resolved with more careful discussion of agreement and significance. Otherwise, I suggest addressing each of the contradictions explicitly in the discussion.
 - a. **Response:** KM21 refers to the parameterization as presented by Kärcher and Marcolli (2021). We formulated a GCM-compatible version (KM21_GCM) to compare to our default approach in ECHAM-HAM (ML20). For greater clarity we replaced the schematic in Figure 1 with a video supplement, in line with your **Comment 21** under the **Specific Comments** section, that explains the differences between KM21, KM21_GCM, and ML20. In summary, KM21 addresses the issue that one should not use cumulative AF with INP-budgeting as it could overestimate the number of newly formed ice crystals. ML20 follows a

cumulative AF approach, but does **not** explicitly budget INPs, instead using a differential ICNC variable. In theory these two approaches should lead to similar results. However, this is only applicable for a single cycle of cirrus ice formation. In a GCM cirrus ice formation is calculated at every timestep, of which there could be several thousand during a typical simulation. In addition, INP concentrations and ice saturation ratio conditions can change across GCM timesteps, among other factors. Therefore, we formulated KM21_GCM to account for these changes across GCM timesteps and to consider the “different ice nucleation behaviors of available INPs”. This is discussed in more detail in the supplementary video.

- b. Regarding the contradictions that you cite in the predicted ICNC, this is related to the formulation of KM21_GCM, the limitations with the box model relative to the GCM, and the rather extreme conditions we tested in the box model. These are either directly addressed or are related to issues you pointed out under **Comments 15, 16, 21, 28, 34, 35, 41, 45 and 46** in the **Specific Comments** section.

2. **Comment:** The author also argue in the abstract and conclusion that ML20 leads to increased interpretability of GCM results, but I think more discussion is needed to support this argument. The formulation of ML20 is indeed simpler, but explicit INP budgeting makes more sense intuitively than the implicit treatment of INP removal in ML20 (e.g., ...”ICNC is updated only if the amount of new ice formation as a portion of N0 exceeds ICNC from the previous sub-timestep”). In short, an argument for increased interpretability could be made for either scheme, so I don’t see this as a clear benefit of ML20, though there are other obvious benefits that are described in the conclusion.

- a. **Response:** We agree that arguments can be made on the interpretability of the results from either approach. The wording that we chose was incorrect in this case. We edited the text in the abstract (see under **Comment 1** in the **Specific Comments** section) and in the conclusions to reflect this correction under **Comment 47** in the **Specific Comments** section.
- b. **Changes in the text:**

“While the KM21_GCM approach with explicit INP-budgeting is closer to first principles when simulating deterministic ice formation in an iterative way following the adiabatic ascent of an air parcel, it requires additional tracers in the climate model. Not only does this require additional memory allocation, and thus greater CPU demand, but it also complicates the parameterization for determining heterogeneous nucleation on externally mixed mineral dust particles in our cirrus sub-model as we must consider changing conditions across GCM timesteps, which also means that there is increased likelihood of unintended errors within the model code. Arguments are emerging that call for a simplification of cloud microphysical processes within GCMs, especially in the case that the simplified model is “equifinal” to the more complex version (i.e. the outcome is similar), (Beven, 2006; Proske et al., 2022). ML20 is a simpler parameterization for deterministic ice nucleation than KM21_GCM as it does not require tracing the maximum AF achieved in a cirrus formation cycle or the INP concentration across GCM timesteps. As our results showed that differences in cloud

properties as well as radiative effects were insignificant, we argue that from the perspective of understanding the impact of cirrus on the climate, the simpler ML20 approach is suitable.”

3. **Comment:** Please also check that all tables and figures appear in the section in which they are first referenced.
 - a. **Response:** This is an easy LaTeX fix that we implemented in the revised manuscript. This will, of course, be addressed for eventual typesetting.

Specific Comments

Abstract

1. **Comment:** I think it is important to point out in the abstract that this study focuses on deposition mode ice nucleation only, possibly in the title, and to allow for the possibility that explicit budgeting could still impact cirrus properties through immersion mode nucleation. As you state on L64, immersion mode is likely the dominant mode of heterogeneous ice nucleation in cirrus. It would also help the reader to briefly describe the difference between explicit and implicit INP budgeting.
 - a. **Response:** These are good points that we neglected. We added the reference to deposition nucleation to the abstract instead of to the title. We also reformulated the middle section of the abstract to differentiate between the explicit and implicit INP-budgeting approaches more clearly. Finally, we added a reference to the fact that this study could be extended to assess the impact our new approach would have on immersion freezing. We also updated Line 64 as new evidence from Froyd et al. (2022) points to deposition nucleation of mineral dust particles as the most abundant source of ice in the upper troposphere, though in their model they excluded immersion freezing. Please find these changes under your **Comment 11**.
 - b. **As you included a few comments on the abstract, we rewrote quite a bit of it.** Therefore, we quote the entire rewritten abstract here for simplicity and refer to these changes in subsequent comments.
 - c. **Changes in the text:**

“Determining the dominant ice formation mechanism in cirrus is still an open research question that impacts the ability to assess the climate impact of these clouds in numerical models. Homogeneous nucleation is generally well understood. More uncertainty surrounds heterogeneous nucleation due to a weaker understanding of the complex physio-chemical properties (e.g. ice nucleation efficiency and atmospheric abundance) of ice nucleating particles (INPs). This hampers efforts to simulate their interactions with cirrus, which is crucial in order to assess the effect these clouds have on the climate system. Kärcher and Marcolli (2021) introduced a new deterministic heterogeneous ice nucleation parameterization based on the differential activated fraction (AF), which describes the number of INPs that activate ice within a specified temperature or ice saturation ratio interval. They argued that this new approach with explicit INP-budgeting, which removes INPs from the total population after they nucleate ice, could help to correct a potential over-prediction of the importance of heterogeneous nucleation within cirrus when budgeting is not considered. We formulated a general circulation model (GCM)-compatible version of the

differential AF parameterization for simulating only deposition nucleation within in-situ cirrus and compared it to the method currently employed in the ECHAM6.3-HAM2.3 GCM that is based on cumulative AF. This default cumulative AF approach does not use explicit INP-budgeting, but instead implicitly budgets for INPs that nucleated ice using a differential ice crystal number concentration variable to calculate whether new ice formation should be added to the pre-existing concentration. In a series of box model simulations that were based on the cirrus sub-model from ECHAM, we found that the cumulative approach likely under-predicts heterogeneous nucleation in cirrus as it does not account for interstitial INPs remaining from the previous GCM timestep. However, as the cases that we simulated in the box model were rather extreme, we extended our analysis to compare the differential and cumulative AF approaches in two simulations in ECHAM-HAM. We find that choosing between these two approaches impacts ice nucleation competition within cirrus in our model. However, based on our five-year simulations, the small and insignificant difference in the top-of-atmosphere radiative balance of $0.02 \pm 0.35 \text{ Wm}^{-2}$ means that the overall climate impact is negligible. We argue that while our GCM-compatible differential AF parameterization is closer to first principles, the default approach based on cumulative AF is simpler due to the lack of additional tracers required. Finally, our new approach could be extended to assess the impact of explicit versus implicit INP-budgeting on the ice crystal number concentration produced by immersion freezing of mineral dust particles as this is also an important mechanism in cirrus.

2. **Comment:** L3: “There is more uncertainty surrounding heterogeneous nucleation processes due to the complex physio-chemical properties of ice nucleating particles.” This is a bit of a general, catch-all statement. The authors could consider a different approach to motivating this study. For example, why is it important to improve representations of ice formation in cirrus? Or, if you want to discuss the complexities of INP properties in the abstract, you should elaborate on how their complex properties (by which I assume you mean the myriad types, ice activation mechanisms/chemistry) relate to poor predictive understanding of INP, and how the poor predictive understanding relates to the challenges of representing ice formation processes in cirrus. In other words, you need to hold the reader’s hand a bit more to help them understand the motivation for studying two different deterministic INP parameterizations.
 - a. **Response:** This is tricky. While we agree that more context is needed, the abstract should be kept as brief as possible, and more detail can be provided in the main text. Therefore, we revised the statement here to be a bit more descriptive and provided more detail in the introduction, which coincides with your **Comment 11** (see changes in the text there). Please see the revised wording for the abstract under **Comment 1**.

3. **Comment:** L4: “...follows a time-dependent (stochastic) or time-independent (deterministic) approach...”. This sounds like a point that would be more appropriate in the introduction. I don’t think this is a necessary distinction to describe in the abstract because the two parameterizations you are comparing are both deterministic.
 - a. **Response:** We agree, and this was excluded from the abstract in favour of more detailed explanations in line with your other comments.

We added this detail in the introduction in the part of the text that corresponds to your **Comment 11**, see changes in the text there.

4. **Comment:** L6: Please define “differential activated fraction”
 - a. **Response:** Agreed. This was added to the text and is quoted in our response to your **Comment 1**.

5. **Comment:** L12: “...as it does not account for INP fluctuations across GCM timesteps.”. After reading the full draft, I return to this line and understand what you mean here, but I think this phrasing is misleading. Looking at Fig. 1 for example, there are “fluctuations” in INP concentrations at each timestep in both schemes. Perhaps “...as it does not account for interstitial INPs remaining from the previous time step”?
 - a. **Response:** Yes, we can see how “fluctuations” is misleading and we agree that more specificity is needed here. We like your wording, so this was amended in the text. See changes in the text under **Comment 1**.

6. **Comment:** L15: “...small and insignificant”. Please clarify what is meant quantitatively by small and insignificant.
 - a. **Response:** We agree this can be clearer, therefore we added the TOA radiative anomaly in the text to make this distinction. Please see changes in the text under **Comment 1**.

Introduction

7. **Comment:** L29: typo at “Climate Change”
 - a. **Response:** Thank you for pointing that out. It is fixed in the manuscript, and you will see it in the tracked changes PDF.

8. **Comment:** L31: “...are mostly well established.”, “mostly” is unnecessary.
 - a. **Response:** We agree, and this was removed from the sentence.

9. **Comment:** L47: “...due to the presence of an INP surface.” Please delete “surface”.
 - a. **Response:** We also agree here, and this was removed as well.

10. **Comment:** L59: “...e.g., on mineral dust (Murray et al. 2021) or on black carbon particles...”. Please delete these two instances of “on”.
 - a. **Response:** This is in line with comment 9 above. These were removed.

11. **Comment:** L65: “In general, as there are more factors that govern the complexities of heterogeneous nucleation than homogeneous nucleation...”. This is another catch-all statement. Again here, please elaborate on how the poor predictive understanding of INP relates to the challenges of representing ice formation processes in cirrus. Also, please define “ice nucleation competition.”

- a. **Response:** We argue that the previous paragraph discusses the issues related to heterogeneous nucleation, but after re-reading a clearer link could be made in the text. We also incorporated changes in line with your **Comment 1**.
- b. **Changes in the text:**

“The theory behind homogeneous nucleation is relatively well understood (Koop et al., 2000; Ickes et al., 2015), with new evidence perhaps suggesting higher freezing onsets at cold temperatures for sulphuric acid droplets (Schneider et al., 2021). However, heterogeneous nucleation in general is still a topic of substantial research (Cziczo and Froyd, 2014; Kanji et al., 2017). Specifically, the ability of certain materials to act as an INP, e.g. mineral dust (Murray et al., 2012), which is likely the most abundant INP species in the atmosphere especially downstream of source regions (Froyd et al., 2022), or black carbon particles (Mahrt et al., 2018, 2020), as well as the characterization of their abundance in the atmosphere (Li et al., 2022). Furthermore, heterogeneous nucleation can occur via several mechanisms. For example, from immersion freezing within a solution droplet or by the deposition of water vapor onto the surface of an INP (Vali et al., 2015; Kanji et al., 2017; Heymsfield et al., 2017), the former of which is thought to be the most common heterogeneous nucleation mechanism in cirrus (Kärcher and Lohmann, 2003), though newer evidence points to the abundance of deposition nucleation in the upper troposphere (Froyd et al., 2022).

Generally, the factors discussed above lead to an overall poor predictability of how INPs influence heterogeneous nucleation mechanisms in cirrus and contribute to uncertainties when simulating these mechanisms in numerical models. This makes it difficult to simulate the impact on ice nucleation competition in cirrus, which influences the ability to reliably estimate the radiative effects of these clouds.

Due to their coarse resolution, general circulation models (GCMs) rely on parameterizations of both homogeneous and heterogeneous nucleation based on laboratory and field-based measurements of ice formation. These parameterizations can follow either a stochastic (time-dependent) approach based on ice nucleation rates or a deterministic (time-independent) approach. For example, homogeneous nucleation of aqueous solution droplets is simulated in the ECHAM-HAM GCM following the stochastic approach by Koop et al. (2000) that is based on simplified assumptions of classical nucleation theory. A common method for simulating deterministic ice nucleation mechanisms is based on the activated fraction (AF, or frozen fraction), i.e. the number of ice-active particles at specific temperature and/or ice saturation conditions out of a population of particles (Vali, 1971; Vali et al., 2015; Vali, 2019; Kärcher and Marcolli, 2021). ...”

12. **Comment:** L69: Please define “AF of available INPs.”

- a. **Response:** We are not sure what you mean by this comment. Is it the AF that is confusing and we should redefine it? Or is it “available” that is confusing? We changed the text to more clearly define AF. We also redefined acronyms in the main text (e.g. INPs). The “AF of available

INPs” is described in the new video supplement to this study, see your **Comment 21**.

b. **Changes in the text:**

“A common method for simulating deterministic ice nucleation mechanisms is based on the activated fraction (AF, or frozen fraction), i.e. the number of ice-active particles at specific temperature and/or ice saturation conditions out of a population of particles (Vali, 1971; Vali et al., 2015; Vali, 2019; Kärcher and Marcolli, 2021).”

13. **Comment:** L68: “...that are based on laboratory measurements of ice formation.”. There are several parameterizations of heterogeneous ice nucleation that are derived from field measurements.

- a. **Response:** This is also a good point. We changed the wording in the text to include both aspects of observational-based measurements.
- b. **Changes in the text:**

“Due to their coarse resolution, general circulation models (GCMs) rely on parameterizations of both homogeneous and heterogeneous nucleation based on laboratory and field-based measurements of ice formation.”

14. **Comment:** L75: “As Vali (1971) explains, on the one hand, the former approach is useful to describe the freezing behavior of a single particle.”. Single particle type? Single INP species? Please rephrase or explain how differential AF approach can also be useful for simulating an INP population.

- a. **Response:** After re-reading this statement and the Vali 1971 paper, as well as conferring with group members who conduct laboratory experiments, we concluded that it is not needed in the context of this study, nor is it correct. The two approaches theoretically are connected by taking the integral of the differential spectra to obtain the cumulative. The way this statement is written it makes it sound like they are disparately different. Therefore, we cut this statement and the following statement starting with “On the other hand...” from the revised text.

15. **Comment:** L80: “For example, if a model explicitly removes INPs from the total available population after each ice formation event and adds them to nucleating ice number concentrations...”. Please rephrase. Do you just mean that INPs are effectively removed from the population when they trigger ice formation?

- a. **Response:** Yes, this is our explanation of INP-budgeting, but we agree this can be clearer as per your comments above. This statement was shortened in the text to make this clearer.
- b. **Changes in the text:**

“However, as Kärcher and Marcolli (2021) highlight, care must be taken when determining which approach to use when calculating the number of ice crystals that can form on INPs. This is especially true for numerical models that simulate the temporal evolution of the ice saturation ratio, based on temperature, to calculate new ice crystal formation, like in ECHAM-HAM (Section 2.1). For example, if a model budgets INPs from the total population

after they nucleate ice, then using the cumulative AF approach may overpredict the number of heterogeneously nucleated ice crystals as it is based on the total number of INPs that could activate between the freezing onset temperature and a given temperature (Vali 1971, 2019)."

16. **Comment:** L82: Please elaborate. It is not immediately clear to the reader how the cumulative AF approach would lead to the overprediction of heterogeneously nucleated ice.

a. **Response:** Yes, we agree here. As this is related to your **Comment 15**, we combined the changes in the text above.

17. **Comment:** L99: Please specify somewhere in this paragraph that this study is limited to deposition mode nucleation.

a. **Response:** We agree that this should be clear throughout the text, so we added the following statements to the paragraph.

b. **Changes in the text:**

"Note, our analysis is applicable only to deposition nucleation mechanisms within in-situ cirrus. Extending the analysis to other ice nucleation mechanisms, namely immersion freezing, is discussed below."

Methods

18. **Comment:** L114: In my opinion, this paragraph should begin with the prior line beginning "Muench and Lohmann (2020)..."

a. **Response:** After re-reading, we agree, and this was moved to the next paragraph.

19. **Comment:** L124: What is meant by "freezing processes"?

a. **Response:** Our wording was also incorrect here. For example, deposition nucleation is not a freezing process (liquid to solid), but rather a nucleation mechanism. For clarity, we refer to homogeneous and heterogeneous nucleation as "modes" and the specific ways in which either of these occur as "mechanisms". We found a couple discrepancies in the text that can help resolve this and you will see them in the tracked changes PDF. Along this line, we amended the specific line you quoted in your comment.

b. **Changes in the text:**

"Kärcher and Marcolli (2021), hereafter KM21, introduced a new method to describe ice nucleation by the number of activated particles."

20. **Comment:** L125: What is $AF(\phi)$? Is AF a function of (ϕ) ? I see ϕ is partially described in L126. Please more explicitly define.

a. **Response:** No, the AF is not a function of phi. It is the symbol we use for the AF in the equations. As this is unclear, we added a "hereafter" in the brackets.

21. **Comment:** Figure 1: The flow and organization of this figure needs to be improved. Please consider putting the resulting ICNC in a single column and creating a single column for the other features of the blocks, such as “total available INP”. You could do the same with subheadings for “Previous INP” and “New INP” for KM21_GCM blocks. The comparable features of each block are not in consistent locations which makes it hard for the reader to follow this figure. Please also specify in the caption whether the INPs in each scenario are interstitial or “total available” or if they are inclusive of previously activated INPs. For KM21_GCM, consider at “+” to indicate that INP from the previous time step are added to the new INP according to AF. Why is the budgeted “leftover” INP in timestep 1 not included in the bottom right blue box for KM21_GCM (i.e., why is $N_{i=1,j=n} = 0$?). Please also add the information about AF on L196 to the Fig. 1 caption. A brief explanation about the different INP treatments at $i=2$ in the caption would also help make this figure read more easily.

- a. **Response:** We agree that this figure is rather confusing for a reader and does not capture the complexity of the issues we are examining in this note. Therefore, we decided to cut this figure in the revised manuscript in favour of a video supplement that walks the reader through the differences between KM21, KM21_GCM, and ML20. Included in the video is an explanation of what we mean by the “different ice nucleation behaviours of available INPs”, related to your **comments 34 and 40**. We replaced the associated text with the schematic to summarise the content of the video. See the revised text below. The video will be uploaded after acceptance of the manuscript, following GMD submission guidelines.
- b. **Changes in the text:**

“The video supplement to this study provides more information on the theoretical understanding of our new KM21_GCM approach and compares it to our default ML20 approach. In summary, we classify different ice nucleation behaviors based on the available INP concentration, following Equation 3. In subsequent GCM timesteps, following the first, we assume that some INPs were ice active in the previous GCM timestep, and are thus removed by subtracting the ICNC that formed previously ($ICNC_{i-1,j=n}$) on INPs ($N_{0,i}$). Out of $N_{0,i}$ we assume some fraction is made up of remaining (“leftover”) interstitial INPs that did not activate ice in the previous GCM timestep ($N_{0,i-1}$) and the remaining fraction contains particles that are new to the system. To obtain the leftover INP concentration, we also subtract $ICNC_{i-1,j=n}$ from the INP concentration from the previous GCM timestep.

In the first instance of ice formation in the current cirrus cycle (current GCM timestep), the leftover INPs nucleate ice according to the differential AF (ψ_j) and the newly available INPs nucleate ice according to the cumulative AF (ϕ_j) as denoted in the numerator of Equation 3. Not only does this approach consider changes in INP concentrations across GCM timesteps, but it also accounts for changes in S_i . For example, if in the current GCM timestep the S_i is drastically lower than that in the previous timestep, then no new ice formation will occur on the leftover INPs. However, ice formation can proceed on the newly available INPs if the S_i is sufficient to produce ice according to the AF calculation following Möhler et al. (2006). If the S_i increases in the current cirrus cycle compared to the previous one, then ice formation

may occur on both the leftover and newly available INPs. Note that Equation 3 also accounts for decreases in INP concentration across GCM timesteps. In such a case the difference term on the left-hand side of the numerator of Equation 3 is set to zero.”

22. **Comment:** L201: “In order to account for leftover INPs as well as INPs that are still included in ice crystals...”. That are removed? Activated and thus removed?
- a. **Response:** As this is related to the specific method of KM21_GCM, we replaced this section in the text with a summary of the new method in the video supplement in line with your **Comment 21**, see above. We also included a summary in the text to accompany the video.
23. **Comment:** L204: “This allows us to properly consider changes...”. Remove “properly”. Track changes?
- a. **Response:** This text was replaced in the revised manuscript to accompany the video supplement in line with your **Comment 21**, see above.
24. **Comment:** Table 1: Please move to the following section where it is first referenced. I noticed this issue with another figure or two. Please check.
- a. **Response:** Thank you for pointing that out. It’s a simple LaTeX fix that you will see in the next version of the PDF. For eventual typesetting, this will of course be addressed.
25. **Comment:** L215: What does “non-exhaustive of the changing conditions...” mean? Please rephrase.
- a. **Response:** It means that the two examples in Figure 1 are not the only two examples of possible INP concentrations and Si values that could be simulated in a GCM. Therefore, they are non-exhaustive. However, as we have replaced Figure 1 with a video supplement, in line with your **Comment 21**, this phrasing is no longer applicable.

Results

26. **Comment:** L229: What is “agreement”? Please explain how you consider agreement quantitatively.
- a. **Response:** Good point. We added a quantitative description of this in the text.
- b. **Changes in the text:**

“Of the 16 tests we conducted, six show agreement between ML20 and KM21_GCM in the predicted ICNC. For these cases we define as a 0% error between KM21_GCM and ML20 for each scenario as denoted by the white shading in Fig. 1.”

27. **Comment:** L233: Please remove “Nevertheless”

a. **Response:** This was removed.

28. **Comment:** L235: Here, and throughout the paper, there is a contradiction that needs to be addressed, whether here or further down in the discussion (but please reference where that discussion begins here). Given that cumulative AF approaches such as ML20 can overpredict ice nucleation compared to explicit budgeting approaches as you described in the Methods, how is it that KM21_GCM is predicting higher ICNC compared to ML20?

a. **Response:** You are right that this is unexpected based on the theory presented in the current methods section. In line with your **Comment 21** (in this document) on Figure 1, we edited the methods section to provide more clarity on this matter.

b. To summarise, there are two issues. Firstly, cumulative AF approaches that use explicit INP-budgeting likely overpredict the number of ice crystals based on the findings by Kärcher and Marcolli (2021). They introduced a new differential AF approach (KM21) to address this specific issue. However, our default approach in the ECHAM-HAM GCM (ML20) uses cumulative AF with implicit INP-budgeting. Finally, KM21 is only applicable in a single formation cycle of a cirrus. In a climate model like ECHAM-HAM this is calculated every model timestep in a sub-model that is called from the cloud microphysics scheme. Furthermore, the INP concentration in the GCM can change between model timesteps. Therefore, we formulated the GCM-compatible version of KM21 (KM21_GCM) to account for interstitial as well as previously activated INPs to compare to the default ML20 approach.

29. **Comment:** L236: “On the one hand, while ML20 considers...” I like this paragraph. The differences between the two treatments are clearly stated.

a. **Response:** Thank you!

30. **Comment:** L251: “Non-zero error between the predicted ICNC for KM21_GCM and ML20 occurs from the start of the second cirrus cycle in the first case (Fig. 3a), and from the start of the third cirrus cycle in the second case (Fig. 3b), where KM21_GCM initially predicts a higher ICNC than ML20.”. Please remove “initially”.

a. **Response:** Agreed as this is applicable throughout the entire second cycle. This was removed.

31. **Comment:** L262: “For ML20, despite a larger AF at the start of the second cirrus cycle, the number of newly formed ice crystals that could nucleate onto the fewer number of available INPs does not exceed the pre-existing ICNC.”. The authors present this condition as the main feature of ML20 that causes

the unexpectedly lower ICNC compared to KM21. Please elaborate on the broader implications. Among cumulative AF approaches, is this condition unique to ECHAM-HAM (where ice does not form unless INPs > pre-existing ICNC)? If not, would you expect other cumulative AF schemes to result in increased ICNC as expected according to KM21 and your explanation in the Methods?

- a. **Response:** We are unaware of other cumulative AF schemes that implicitly budget INPs like we do in ECHAM-HAM. For example, the heterogeneous ice nucleation parameterizations by Barahona and Nenes (2009) and Liu and Penner (2005), that are commonly used in the CAM5 GCM, do not explicitly state whether they follow a similar approach as we do. This implicit budgeting feature follows the implementation by Muench and Lohmann (2020), who re-worked the cirrus sub-model code to simplify various parameterizations and improve code readability. Despite this, we edited the text to make this clearer that this is a feature of the implicit budgeting approach.
- b. **Changes in the text:**

“For ML20, the implicit INP-budgeting approach prevents new ice formation from occurring at the start of the second cirrus cycle, despite a larger AF, as the number of newly formed ice crystals that could nucleate onto the fewer number of available INPs does not exceed the pre-existing ICNC. No new ice formation occurs until the S_i increases sufficiently after nearly 6 minutes.”

32. **Comment:** L279: What is meant by “To emulate the procedure in the GCM...”? Please rephrase.

- a. **Response:** This simply means to copy the method in the GCM code in our box model. We rephrased this sentence.
- b. **Changes in the text:**

“Following the procedure in the GCM, ice formation is not calculated when the large-scale $S_i \leq 1.2$ as the Möhler et al. (2006) AF would be zero.”

33. **Comment:** L291: What is meant by “arguably”? Please elaborate.

- a. **Response:** What was meant by this is that while this case did not show the largest error in the predicted ICNC between ML20 and KM21_GCM, it is notable due to the differences in approaches. For clarity though, we removed “arguably” in the revised text.

34. **Comment:** L313: “Based on our box model results, it is likely that ML20 underpredicts the number of heterogeneously formed ice crystals under cirrus conditions compared to our KM21_GCM approach as it neglects the different ice nucleation behaviors of available INPs.”. Again, please discuss the discrepancy between the result, your expectations for overprediction described in Methods and the motivation for KM21. Also, please elaborate on or rephrase “...it neglects the different ice nucleation behaviors of available INPs.” I am not sure what this means. Please specify what is “different” and

what entities are being compared. Are you arguing that KM21 better emulates the variability in IN-activity between dust populations, or between individual dust particles? If so, you will need to supplement this section with supporting evidence.

- a. **Response:** This is related to previous comments that you made, specifically **comment 21**. We added a new description and a video supplement to explain the differences between KM21_GCM and ML20 in more detail under comment 21. For this statement, we added a reference to the video supplement.
- b. **Changes in the text:**

“Based on our box model results, it is likely that ML20 under-predicts the number of heterogeneously formed ice crystals under cirrus conditions compared to our KM21_GCM approach as it neglects the different ice nucleation behaviors of available INPs (refer to the video supplement).”

35. **Comment:** L322: “...this may not be the case using a GCM.” Do you mean it remains to be seen how frequently these errors occur?
 - a. **Response:** Yes, and after re-reading this sentence, we can remove the last two clauses as our point was conveyed previously in the same sentence.

36. **Comment:** L336: Please briefly describe the false discovery rate method by Wilks (2016) to help the reader understand the significance testing applied. If this is the only significance test applied, please state that Wilks (2016) is what is referred to throughout the rest of the results and discussion.
 - a. **Response:** We agree, and we reformulated and added a couple sentences on this.
 - b. **Changes in the text:**

“The stippling in Fig. 5 displays insignificant data points based on an independent t-test, following the false discovery rate method by Wilks (2016). This approach accounts for high spatial correlation of neighboring grid-points where the null-hypothesis cannot necessarily be rejected. Like Tully et al. (2022c), we calculate a 5% significance based on the inter-annual variability of the five-year simulations. For the remainder of this section, we base significance on this method.”

37. **Comment:** L353: “This is only partially reflected in zonal profiles of cloud fraction and relative humidity (RH) anomalies in Fig. 6, where there are only small positive anomalies in the southern hemisphere (SH) tropics of up to 1 % that are insignificant (as denoted by the stippling).”. What is meant by “partially reflected” if the anomalies are insignificant? Also, please be explicit here and elsewhere throughout the manuscript on what is meant by significant.
 - a. **Response:** We agree this is unclear. The cloud fraction and RH anomalies in the tropics are small and the fact that they are insignificant, as denoted by the stippling, means that it is unclear whether the increase in HOM we found with KM21_GCM is supported

by these anomalies. We revised the text to reflect this. We also added text above in line with your **Comment 36** on being more descriptive on how we define significance in this section.

b. **Changes in the text:**

“It is unclear whether this is reflected in the zonal profiles of cloud fraction and relative humidity (RH) anomalies in Fig. 5. Throughout the tropics we find only small cloud fraction and RH anomalies of around $\pm 1\%$; however, these signals are insignificant as denoted by the stippling.”

38. **Comment:** L357: “There are significant, positive cloud fraction and RH anomalies between 1 and 10 % towards the mid-latitudes and the poles in both hemispheres.” Please add a reference to Figure 6.

a. **Response:** Good point. This was added to the text.

39. **Comment:** L358: “However, the HOM signal is not consistent throughout the SH and is insignificant.” What is meant by HOM signal?

a. **Response:** This refers to the anomaly of homogeneously nucleated ice in Figure 5 (now Figure 4). It is defined on Line 354. However, in the revised text we refer to this as “The increase in HOM” to make it clearer.

40. **Comment:** L363: See previous comment on “different ice nucleation behavior of available INPs.”

a. **Response:** This is addressed in the supplemental video in line with your **Comment 21**. The video is now attached to the manuscript that explains what we mean by this phrasing. In summary, we differentiate between “new” INPs and “leftover” (interstitial) INPs as there is no direct communication from the cirrus sub-model back to the aerosol model. Therefore, in subsequent GCM timesteps, we must assume that some of the available INPs will be made up of those that are new to the system and those that are leftover. In our new KM21_GCM approach, we take the ice nucleation behaviour of these leftover particles into account in the differential AF approach.

41. **Comment:** L363: “...we found that it often allows for higher rates of ice formation in cirrus.”. Did you track ice formation rates in ECHAM-HAM? Or do you mean KM21 results in higher ICNC? This point about frequency is another contradiction with your previous box model results, in which the results showed that the resulting ICNC between the two schemes frequently agree. The authors further argued that the conditions for which the resulting ICNC differed would occur infrequently in a GCM (L320). Please address this contradiction.

- a. **Response:** Yes, we meant that KM21_GCM produces higher ICNC. However, after re-assessing our arguments you are right that our box model does show that these two approaches often agree, or at least show very little error. This was our oversight and for that reason, we reformulated the wording on Lines 320 and 363. This is in line with your **General Comment 1** and **Comment 45** below.
- b. **Changes in the text:**

“Some of the changes in large-scale S_i and INP concentrations we tested in the box model were rather extreme in order to examine differences between KM21_GCM and ML20. However, our box model setup is limited as we assume a constant temperature and updraft velocity. We also did not consider other processes such as ice sedimentation and mixing that would be simulated in a GCM. Furthermore, the KM21 parameterization was developed for a single air parcel within a process model that depicts ice formation within a single cirrus. It does not capture the complexities associated with changes in INP concentrations as well as S_i (among several other factors) across several hundreds of timesteps in a typical GCM simulation. Therefore, we present a short analysis comparing our GCM compatible differential AF parameterization, KM21_GCM, to our default ML20 approach for deterministic heterogeneous ice nucleation in ECHAM-HAM in Section 3.2.”

“There is a much clearer signal in the northern hemisphere (NH) mid-latitudes (roughly 45 °N – 60 °N) where both HOM and HET produce more ice in KM21_GCM than in ML20. While the positive HET ICNC anomaly is consistent with some of our findings from our box model results (Section 3.1) that showed KM21_GCM produced higher ICNC than ML20, it is insignificant for the five years we simulated with the GCM and is only evident in the NH. It is more likely that the GCM results confirm our box model results that show in most cases KM21_GCM and ML20 agree or have a very small error (Fig. 1).”

42. **Comment:** L370: “While there are relatively large, but insignificant changes...”. Please define quantitatively what you consider “relatively large”.
- a. **Response:** After re-reading this, we agree this is not descriptive. We also see that the anomalies themselves are not necessarily that large relative to the ML20 “reference” case. Therefore, we reformulated this in the text.
 - b. **Changes in the text:**

“While the zonal mean HOM and HET ICNC tracer anomalies for KM21_GCM (Fig. 5) are both notable (by at least $\pm 10 L^{-1}$) relative to our reference ML20 simulation, they are insignificant for the five years we tested. Therefore, it is difficult to describe the exact effect of choosing one deterministic ice formation parameterization (ML20 or KM21_GCM) over the other.”

43. **Comment:** L373: “Nevertheless, these changes correspond to only a small positive top-of-atmosphere (TOA) warming effect by around $0.02 \pm 0.35 Wm^{-2}$ that is driven predominately by a weaker shortwave (SW) cloud radiative effect (CRE).” Is there a reference for the cirrus contribution to CRE in ECHAM-HAM? This would be helpful context.
- a. **Response:** We agree that this is a good point for context. Both Gasparini et al. (2016) and Gasparini et al. (2020) quantified the cirrus

CRE in ECHAM. We added these estimates and some context to our findings in the text.

b. **Changes in the text:**

“Despite this finding, the maximum positive and significant anomaly for cloud fraction is 3.6%, which equates to only a small positive top-of-atmosphere (TOA) warming effect by around $0.02 \pm 0.35 \text{ Wm}^{-2}$ that is driven predominately by a weaker shortwave (SW) cloud radiative effect (CRE). Similarly, the global mean net CRE anomaly between the two cases is indistinguishable from zero, $0.00 \pm 0.32 \text{ Wm}^{-2}$. These radiative anomalies are negligible relative to the estimated CRE from cirrus clouds of 5.7 Wm^{-2} and 4.8 Wm^{-2} by Gasparini and Lohmann (2016) and Gasparini et al. (2020), respectively.”

Conclusions

44. **Comment:** L400: “...nor does it consider the different ice nucleation behaviors of available INPs.”. Please clarify.

a. **Response:** This was addressed with the video supplement in line with your **Comment 21** and is summarised in the text in the methods section, see above.

45. **Comment:** L404: “The large-scale S_i conditions and the changes in INP concentrations between cirrus cycles that we tested with our box model were rather extreme and may not occur frequently in a GCM.”. Would it be possible to calculate the frequency of these conditions from the ECHAM-HAM output?

a. **Response:** This wording was an oversight on our behalf. The limited box model simulations were merely to understand the differences between the two approaches with large changes in the starting conditions we used as input. However, these tests were limited as they did not consider all of the possible changes that can occur across GCM timesteps. Therefore, we extended the analysis with GCM simulations. We amended the text to reflect this change.

b. **Changes in the text:**

“We tested rather extreme changes in the large-scale S_i conditions and INP concentrations between cirrus cycles in our box model to examine the differences between the ML20 and KM21_GCM approaches. However, our setup was limited as it did not capture all of the possible conditions and processes that are simulated in a GCM and that are relevant to assessing cirrus climate effects. Namely, we used a constant temperature and updraft velocity in our box model setup. In addition, we did not consider processes such as ice crystal sedimentation and mixing effects (e.g., entrainment). As a result, we extended our analysis of ML20 and KM21_GCM with two additional tests with the ECHAM-HAM GCM. ...”

46. **Comment:** L408: “However, the signal is mostly insignificant for the five years that we tested (2008-2012), and is inconsistent with the findings from our box model simulations, except in the NH.”. Please define “signal” and “mostly insignificant.”

a. **Response:** Agreed. This is unclear, we reworked this statement to specifically refer to the cirrus ICNC tracer anomalies.

b. Changes in the text:

“... As a result, we extended our analysis of ML20 and KM21_GCM with two additional tests with the ECHAM-HAM GCM. We found that choosing one of the two deterministic ice formation approaches has an impact on ice nucleation competition within cirrus. However, the cirrus ICNC tracer differences for both homogeneous and heterogeneous nucleation were insignificant between these simulations for the five years that we tested (2008-2012). These results corroborate our findings from our box model simulations, which showed that out of the 16 tests we conducted six showed agreement (0% error) and an additional three tests showed a small error between ML20 and KM21_GCM of 0.3% (Fig. 1). This likely highlights that the GCM was often in similar regimes over the five years of simulation as the tests in our box model that showed zero or small errors.

47. **Comment:** L414: “Not only does this require additional memory allocation, but it also introduces more room for potential error.” Please elaborate on “room for potential error.”

a. **Response:** Agreed. We reformulated this sentence to combine it with the following one to make this point clearer.

b. **Changes in the text:**

“Not only does this require additional memory allocation, and thus greater CPU demand, but it also complicates the parameterization for determining heterogeneous nucleation on externally mixed mineral dust particles in our cirrus sub-model as we must consider changing conditions across GCM timesteps, which also means that there is increased likelihood of unintended errors within the model code.”

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