Prior heterogeneous ice nucleation events increase likelihood of homogeneous freezing during the evolution of synoptic cirrus – Responses to RCs of second review

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We thank the editor for handling the review of our work. Our detailed responses to the RCs are provided in the following sections.

1 Response on RC1

General comment:

The manuscript has improved significantly. The authors invested considerable effort into the study, and their work has paid off. The paper is now nearly ready for publication. However, I still have a few comments -both on previously raised issues and on some new points- which are listed below.

Reponse to G1: We thank the reviewer for providing valuable insight and fix suggestions in the second review of the manuscript. We have clarified each comment below.

Specific comments:

Comment 1: Line 33f: *'Over the past few decades, several key measurement campaigns (e.g., Krämer et al., 2009; Voigt et al., 2017) have been conducted in the UTLS.'*

Please add more recent work here:
- Krämer et al., 2009 reported multiple campaigns, the following studies could be added:
Krämer et al., 2016 (ACP), Krämer et al. 2020 (ACP), Patnaude et al., 2021 (ACP), Ngo et al., 2024 (ACP). Voigt et al. (2017) presents a single field campaign (ML-Cirrus), the following studies could be added:
i.e. Pan et al. (2010) (START08, BAMS), Wendisch et al. (2016) (ACRIDICON-CHUVA, BAMS), Jensen et al. (2017) (ATTREX, BAMS), Pan et al. (2017) (CONTRAST, BAMS). These campaigns are included either in Krämer et al. (2020) or in Ngo et al. (2024) (or both).

Response: Suggested campaign studies are added to the list of references as follows.

Lines 33-35 (in the revised manuscript): *'Over the past few decades, several key measurement campaigns Added references e.g., Pan et al. (2010, START08,BAMS), Jensen et al. (2013b, MACPEX), Wendisch et al. (2016, ACRIDICON-CHUVA), Jensen et al. (2017, ATTREX,BAMS), Pan et al. (2017, CONSTRAST) have been conducted in the UTLS.'*

New comment: That's not what I meant - now only a few of all to campaigns are mentioned... I suggest
'Over the past few decades, a number of key measurement campaigns have been conducted in the UTLS, which are compiled by Krämer et al. (2016, 2020) and Ngo et al. (2025).'

Answer to the new comment The text has been edited in the suggested form.

Comment 3: Line 38ff: *'Heterogeneous ice nucleation . . . In contrast, homogeneous freezing ...'*

Please provide references for heterogeneous and homogeneous freezing.

Response: Reference added as in the response to comment 2. *...(Pruppacher and Klett, 1997)'*

New comment: This is a fairly old reference; since then, the understanding of heterogeneous and homogeneous freezing has evolved considerably. I highly recommend citing more recent references here – and/or point to new Section 2.3.

Answer to the new comment The text has been edited in the suggested accordingly with followin form: 'Cirrus clouds primarily form through two dominant mechanisms: heterogeneous and homogeneous freezing (Pruppacher and Klett, 1997; Cziczo and Froyd, 2014; Kanji et al., 2017).' Also references were added to the definition of homogeneous freezing: 'In contrast homogeneous freezing occurs in the absence of INPs and takes place when aqueous solution droplets freeze at temperatures below the -38C threshold for pure water and at high Si (e.g., Schneider et al., 2021; Koop et al., 2000).

Comment 21: Line 486ff:
'Simulations with measured mineral dust concentrations (STND) showed an almost complete absence of homogeneous freezing. This suggests that prior heterogeneous nucleation events likely depleted the heterogeneous INPs from certain layers of the cirrus clouds, particularly in the colder upper regions.'
Something is weird here ... why does complete absence of homogeneous freezing suggest that prior heterogeneous nucleation events likely depleted the heterogeneous INPs?

Response: The heterogeneous ice formed due to the number of heterogeneous INPs measured was preventing Si from reaching high enough for homogeneous freezing in the simulations. A clarification was added to the leading sentence in the following form: *'Simulations with measured mineral dust concentrations (STND) showed an almost complete absence of homogeneous freezing due to heterogeneous ice preventing supersaturation over ice from reaching the critical threshold level for homogeneous freezing.'*

'. . . almost complete absence of homogeneous freezing...' still reads strangely in the context, because the message of the manuscript is that the measured ice crystals formed predominantly homogeneously.

If I understand it correctly, the result that 'the measured ice crystals formed predominantly homogeneously' comes from the ADJ simulations and the homogeneous freezing does not occur in the STND runs. The difference is the moisture profiles. The moisture profiles for ADJ, which are responsible for the homogeneous freezing, can be seen in Figure 10 (see my comment on Figure 10 below). The difference is that in the STND runs Si does not reach the homogeneous freezing threshold, whereas in the ADJ runs it does. Therefore, it should be clear from the text that the subsequent dynamic situation must be favorable so that homogeneous freezing can take place after the heterogeneous exhaustion of the INPs. Since this is the core message of the manuscript, here an attempt to rephrase

this section as I would understand it better (and hope it is correct: 'We investigated the role of heterogeneous ice nucleation with the UCLALES-SALSA model, and the results showed that prior heterogeneous ice nucleation increases the likelihood of homogeneous freezing during subsequent ice nucleation events. Simulations with measured mineral dust concentrations (STND) demonstrated that heterogeneous ice nucleation events likely depleted the heterogeneous INPs from certain layers of the cirrus clouds. In addition, ice supersaturation is suppressed below the homogeneous freezing threshold, particularly in the colder upper regions, resulting in an almost complete absence of homogeneous ice nucleation. Under dynamic conditions allowing supersaturations to reach the homogeneous freezing threshold, such as in the ADJ scenario, this prior depletion indirectly enabled the occurrence of homogeneous freezing in later stages.

Answer to the new comment Thank you for suggesting an alternative wording. We agree that this part of the text was challenging to convey clearly. We have adopted your suggested version, as it aligns well with the intended message of the manuscript.

New comments

Comment 1: Figures 9 and 13: I preferred the previous versions of the Figures, particularly since the model scenarios (STND and AGED / ADJ and HOM) were listed above the two columns. Now, you have to figure the scenarios out from the caption. Please reinsert the abbreviations of the scenarios to the right of the rows.

Response: Abbreviations added to the rows accordingly both in Figs 9 and 13.

Comment 2: Figure 10: In the Figure caption it is mentioned
' ... (b) ice saturation profiles from the beginning to the end of the simulation using the STND setup.' I think that the ADJ scenario is meant ?

Response: This Figure shows the time evolution of the dust and humidity profiles in a STND run. The ADJ uses this end state of STND as a reference initial state. We replaced the end of the sentence to be more clear: ' in a STND run', instead of 'using the STND setup'.

Comment 3: Line 377: *'Also the homogeneous freezing is known to be mostly insensitive to the concentration or size distribution of available aerosols, ... '*

Homogeneous freezing is sensitive to the size distribution of the aerosols present, in particular at cold temperatures and higher updrafts. as shown in Baumgartner et al. (2023). Baumgartner, M., Rolf, C., Grooß, J.-U., Schneider, J., Schorr, T., Möhler, O., Spichtinger, P., and Krämer, M.: New investigations on homogeneous ice nucleation: the effects of water activity and water saturation formulations, Atmos. Chem. Phys., 22, 65–91, https://doi.org/10.5194/acp-22-65-2022, 2022.

Response: The text has been modified to also reflect the recent findings on the sensitivity of homogeneous freezing to the size distribution by following form: *Homogeneous freezing has often been described as largely insensitive to the concentration or size distribution of available aerosols (Kärcher and Lohmann, 2002; Jensen et al., 2010). However, more recent studies (Baumgartner et al., 2022) indicate that under certain conditions, particularly at very cold temperatures and high updraft velocities, the aerosol size distribution can influence freezing. This choice is justified because the resulting number of solution droplets available for homogeneous freezing remains substantially higher than the concentration of heterogeneous INPs, such that homogeneous freezing still produces higher ice concentrations. Our analysis therefore focuses on how heterogeneous ice nucleation shapes the evolution*

of S_i, while also comparing the relative contributions from homogeneous versus heterogeneous ice nucleation.

Comment 4: Line 555f (of manuscript with changes tracked): *'This reflects the known sensitivity of homogeneous freezing to the magnitude of vertical velocity.'* I suggest to add here *' and also the weakening of homogeneous nucleation events due to prior heterogeneous ice nucleation (Spichtinger and Cziczo, 2010).*

Response: The text was edited according to the suggestion.