Second review of

Prior heterogeneous ice nucleation events increase likelihood of homogeneous freezing during the evolution of synoptic cirrus

Kasper Juurikkala et al.

Previous comments are in black, responses in blue and new comments are in green

New General:

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.

Specific comments:

(S 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 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 the 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).'

(S 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.

(S 21) Line 486ff (new 460ff): '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.

(N 1 - New comment) 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.

(N 2 - New comment) 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?

(N 3 - New comment) Line 377 (of manuscript with changes tracked):

'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.

(N 4 - New comment) 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).