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

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General:

This manuscript examines the formation mechanism of a synoptic cirrus observed during the MACPEX field campaign. The observations suggest homogeneous freezing as the dominant nucleation mechanism, which is investigated through simulations with the UCLALES-SALSA model.

Generally, the manuscript deals with the topic of the influence of heterogeneously freezing particles (so-called INPs, mineral dust in this case) on the homogeneous freezing process. The theory of the microphysics of this process is well understood and the study does not provide new microphysical insights. What is presented are sensitivity studies with a model in which the measured properties of a cirrus, namely that it is formed by pure homogeneous freezing, can be confirmed by theoretical considerations. That means in summary the agreement between theory and observations is shown. Specifically, the study shows that pure homogeneous freezing took place in the upper layers of the observed cirrus, because the previously existing INPs had already frozen out earlier.
The study also examines in general the influence of INPs on the formation process - heterogeneous or homogeneous - of cirrus clouds in the specific dynamic situation of the observed cirrus clouds. The influence of small-scale fluctuations of vertical velocity is also discussed.

Overall, the study is interesting and timely and the methods used are appropriate, making the manuscript suitable for publication in ACP.

(G 1) However, I find the title a bit exaggerated and not quite appropriate for the study, even though I understand that the title is intended to arouse curiosity. I suggest toning down the title to something like

'Prior heterogeneous ice nucleation events shape homogeneous freezing during the evolution of synoptic cirrus'
or
'Prior heterogeneous ice nucleation events enable / allow pure homogeneous freezing during the evolution of synoptic cirrus'

or, completely different: 'The influence of heterogeneous ice nucleation on cirrus evolution: a case study of an observed homogeneously formed cirrus'

which to my feeling better reflect the content of the study. This content could/should also be better elaborated in the paper.

(G 2) In general, I have to say that the study could/should be significantly improved in terms of writing. In its current form, the reader cannot follow the content fluently - it took me a long time to put the puzzle together, and if I hadn't had to review the paper, I certainly wouldn't have read it to the end....

Often (a) references to previous work is missing; also,
(b) the model description is too brief.
For more detail see the see specific comments.

Further, (c) the manuscript is not well structured, which makes it unnecessarily difficult to understand. As outlined in the specific comments, I recommend to

- distribute Section 5.4 to Section 4 and Sections 5.2/5.3 and
- move Fig. 12 behind Figs. 11 and 13.

This would give the manuscript a clearer structure and Fig. 12 - the summary of the results - would be the final figure to the paper, which I think is a better place then now in between more detailed results.
A further idea might be to then – merge sections 4 and 2.3 to have only one section describing the model and the simulated scenarios.

My final assessment is that the paper needs fundamental revision, in terms of the focus and findings of the study, the introduction to the topic, the incorporating of previous work and a more fluent structure.

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).

(S 2) Line 38: '*Synoptic cirrus clouds primarily form through two dominant mechanisms: heterogeneous and homogeneous freezing.***'**

This applies not only to synoptic, but to all cirrus.

(S 3) Line 38ff: *'Heterogeneous ice nucleation … In contrast, homogeneous freezing …'*

Please provide references for heterogeneous and homogeneous freezing**.**

(S 4) Line 48ff: '*Ice nucleation in cirrus clouds is strongly influenced by the abundance of INPs, which regulate how efficiently heterogeneous ice nucleation can suppress homogeneous freezing and activity.'*

Please provide references for the competition between heterogeneous and homogeneous freezing, i.e. influence of INP number and vertical velocity on the onset of homogeneous freezing. however, the present study lacks references to previous work. Here are some examples, but the list is not exhaustive.

– Spichtinger, P., and D. J. Cziczo (2010), Impact of heterogeneous ice nuclei on homogeneous freezing events in cirrus clouds, J. Geophys. Res., 115, D14208, doi:10.1029/2009JD012168.

– Rolf, C., Krämer, M., Schiller, C., Hildebrandt, M., and Riese, M.: Lidar observation and model simulation of a volcanic-ash-induced cirrus cloud during the Eyjafjallajökull eruption, Atmos. Chem. Phys., 12, 10281–10294, https://doi.org/10.5194/acp-12-10281-2012, 2012.

– Krämer, M., Rolf, C., Luebke, A., Afchine, A., Spelten, N., Costa, A., Meyer, J., Zöger, M., Smith, J., Herman, R. L., Buchholz, B., Ebert, V., Baumgardner, D., Borrmann, S., Klingebiel, M., and Avallone, L.: A microphysics guide to cirrus clouds – Part 1: Cirrus types, Atmos. Chem. Phys., 16, 3463–3483, https://doi.org/10.5194/acp-16-3463-2016, 2016.

– Kärcher, B., Jensen, E. J., & Lohmann, U. (2019). The impact of mesoscale gravity waves on homogeneous ice nucleation in cirrus clouds.
Geophysical Research Letters, 46, 5556–5565. https://doi.org/10.1029/2019GL082437

The authors miss to discuss this paper, although it also compares MACPEX observations with simulations.

– Kärcher, B., DeMott, P. J., Jensen, E. J., & Harrington, J. Y. (2022).
Studies on the competition between homogeneous and heterogeneous ice nucleation in cirrus formation. Journal of Geophysical Research: Atmospheres, 127, e2021JD035805. https://doi.org/10.1029/2021JD035805

(S 5) Section 2.3 UCLALES-SALSA

a) The information about the model is very limited. I recommend a short description of which parameterizations for heterogeneous and homogeneous freezing are used and how the processes (ice particle growth, evaporation, sedimentation, aggregation, ...) are treated in the model.

b) Further, please provide information about the sizes of the ice bins.

(S 6) Figure 4: What time resolution do the measurements shown have?

Also, why not showing the IWC (ice water content) in addition ? There was a total water instrument on board (CLH), so together with the HVW gas phase water you have a good IWC information.

(S 7) Figures 2, 5, 6:

To give the reader a better overview, I recommend making one plot from Figures 2 and 6, showing the cirrus clouds from Figure 6 and the flight path from Figure 2. Please also draw the backward trajectory from Figure 5 in this plot.

(S 8) Line 184f: '*In Fig. 8a, the measured Ni is presented and it exceeds the concentration of mineral dust particles or other potential heterogeneous INPs, providing strong evidence that homogeneous freezing played a significant role in shaping the Ni distribution*.'

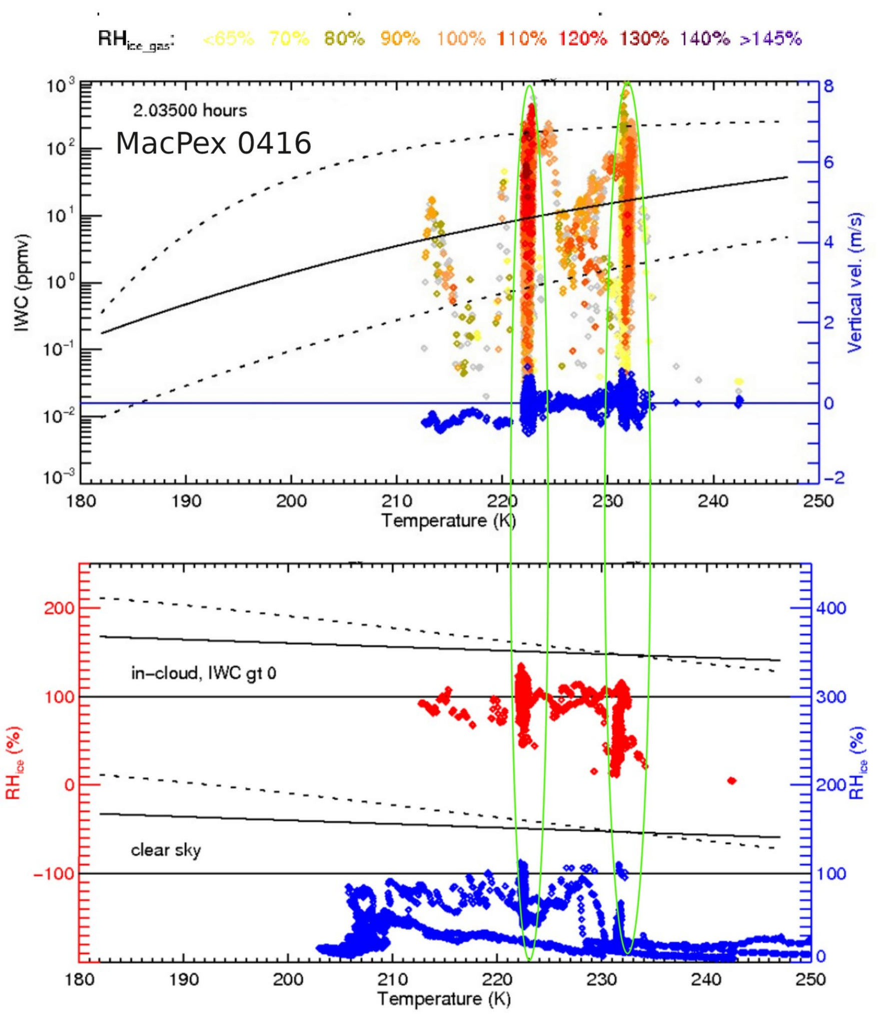
As you stated earlier in the paper (page 3) 'The limited reliable observation capability of the 2D-S probe above 15 µm restricts obtaining accurate information about young cirrus clouds with high number concentrations of smaller-sized particles…' . Now you use Ni to conclude that homogeneous freezing occurred, correctly arguing that Ni is higher than the concentration of potential heterogeneous INPs. However, I think it should be mentioned again here that Ni does not correspond to the actual ice particle concentration since the small ice particles are missing.

(S 9) Line 187f: '*The MMS measured Si in Fig. 8B …*' Isn't Si from HWV measurements?

(S 10) Line 189ff: '*Observing high Si required for homogeneous freezing is inherently challenging, especially within fully developed cirrus clouds, as the available humidity rapidly decreases following a homogeneous freezing event. …*'

What you have written is certainly correct. However, it is possible to find homogeneous events in the measurements, if they are not too old, because they have a signature of high IWCs together with high Si.

I couldn't resist looking at the MacPex 1Hz data from this flight (see plots below) and indeed I would interpret the two events circled in green as homogeneous freezing events – IWC and Si go up to high values and the vertical velocity fluctuations are also quite high. The in-cloud RHi of the younger event around 223K is close to the homogeneous freezing threshold, while the event around 232K appears to be already aged with lower RHi. Outside of the cirrus, RHi is only slightly above saturation, suggesting that cirrus formation probably started with heterogeneous freezing, followed by a subsequent homogeneous freezing event.



(S 11) Line 200f: ‘*The median Ni is about an order of magnitude lower in the lower parts of cirrus which could be explained by following factors:*
– Homogeneous and heterogeneous ice nucleation produces higher number of ice when the temperatures are lower (Jensen et al., 2013b), leading to higher Ni in the upper parts of cirrus.’

I think the difference in nucleation rates is not that large in this temperature range… the next points sounds better

‘– The WB-57F collected statistically significant data between 9–11.2 km, covering only the lower portion of the cirrus cloud, where Si is strongly influenced by sedimenting ice crystals. At 10 km, the formation of new ice crystals is more unlikely than at 11.2 km, as most ice at this altitude likely originated from higher layers. Competition for available water vapor further reduces the potential for ice nucleation.’

(S 12) Line 251ff: *‘Ahola et al. (2020) implemented various freezing mechanisms, … . By default, UCLALES-SALSA uses homogeneous and heterogeneous ice nucleation parametrization schemes based on Khvorostyanov and Curry (2000), however, due to the heterogeneous ice nucleation schemes being stochastic (time dependent), it was concluded that ice nucleation could be greatly over-estimated in this particular study. To overcome this issue, a deterministic, time-independent deposition nucleation parametrization developed by Ullrich et al. (2017) for uncoated mineral dust particles was implemented to SALSA. The reparametrization was created by using a fit to ice nucleation activity of several mineral dust particles presented in Kanji et al. (2011). For this scheme, tracking of activated INP fractions is necessary since deterministic parametrizations base their ice nucleation activity on original INP population. Homogeneous freezing is implemented based on the temperature and Si relation presented in (Koop et al., 2000).*

a) I would move this paragraph to Section 2.3 UCLALES-SALSA, the information is missing there, see my point **(S 5).**

b) One could also consider moving the entire Section 4 to Section 2.3.

(S 13) Line 274f: *‘These runs are referred to as STND (standard) and AGED respectively from hereafter.’*

Please provide a table in which the conditions of all simulation set ups (STND, AGED, ADJ, HOM) are summarized.

(S 14) Figure 12: Distributions of simulated and observed Ni are shown.

a) An important point not mentioned in the paper is the size range over which the simulated Ni is calculated. Only the 2D-S size interval (> 15 um) should be considered. Otherwise the simulated Ni are not comparable with the measurements.
If smaller ice crystals are included in the simulated Ni, the analyses should be repeated for the appropriate size interval.

b) Figure 12 is not discussed until after Figure 13 - I recommend that it is only shown after Figure 13, as it sums up the results of the study.

(S 15) Line 409ff: *'Among all cases in this study, the HOM simulations show the closest statistical resemblance to the 2DS measurements, proving that without any influence of heterogeneous ice nucleation the Ni exceed the clear air concentration of mineral dust most efficiently.'*

This is true for the upper layer of the cirrus cloud, but not for the lower, where the AGED fits best to the measurements. This is not clear from the text.
As this part of the manuscript is of great importance, I recommend that the text be revised accordingly.

(S 16) Figure 14, Section 5.4 : Frequency distributions of vertical wind.

I strongly recommend to show this this Figure earlier. While reading the discussion of Figs. 11 and 13, I have been wondering the whole time how the fluctuations of the vertical wind in the model correspond to the measured ones.

It would fit in in Section 4, or, as recommended in S 12 / S 6, all the relevant information on the simulations in Section 2.3?

(S 17) Line 435-439: *'It was stated previously that the maximum Ni achieved in HOM cases was not clearly correlated to the imposed large-scale w. The cooling within the supersaturated layer was primarily influenced by the large-scale w; however, small-scale turbulence induced local variations in temperature and humidity. …. '*
The whole paragraph would be better included in the discussion of Figs. 11 and 13.

(S 18) Section 5.4.1: This section (including Fig.15) would also be better included in the discussion of Figs. 11 and 13.

(S 19) Figure 15: For better comparison, please synchronise the y-axes (temperature and Ni) of panels (a) and (b).

(S 20) Line 482f: *'…. the cirrus clouds on April 16th, 2011, were predominantly formed through homogeneous freezing, …'*

… the top layer of cirrus clouds on April 16th, 2011, were predominantly formed through homogeneous freezing, …

(S 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?

(S 22) Did I miss it or is there no information about the heterogeneous freezing threshold?