Authors’ Response to RC1 and RC2

July 16, 2026

We thank both reviewers for their time and effort spent on our manuscript and for their valuable remarks and suggestions. In the following, the reviewers’ comments are marked in black and our answers in blue. Showcased text changes are shown in ~~*red*~~ (original) and *green* (revised).

Comment of Referee #1

Summary

This manuscript (“Upward transport and segregation of ice-nucleating particles in deep convective clouds”) by Schaefer et al. investigates the transport of particles in deep convective clouds, with a focus upon ice nucleating particles. They utilize the HALO research aircraft to collect measurements of particle number and composition above Europe. They sampled aerosol and cloud residual composition using a single particle mass spectrometer, and collected filters for offline analysis. They primarily present results from a case study sampling a deep convective system, where they sampled the inflow the convective system, in-cloud, and the outflow of the convective system. The convective inflow had high INP concentrations at very warm temperatures (> -15 °C), while the in-cloud had INPs at warm temperatures, but particularly from -14 to -17 °C, and the convective outflow had virtually no INPs at temperatures warmer than 17 °C. They observed that DCC outflow INPs were much higher than would have been expected from the background INP concentrations in the free troposphere/lower stratosphere, indicating that the INPs were transported from the BL. They also observed that the composition of free tropospheric air, in-cloud residuals, and convective outflow varied systematically, though this aspect of the paper is under-developed.

I like this study, think it is interesting, and that it warrants publication in ACP after some revisions. My suggestions are below.

Thank you for your very constructive review and helpful recommendations in general. We will answer the specific comments below. This also includes remarks of reviewer 2.

General comments

First, this paper is primarily about a single case study, with a handful of supporting measurements from complementary flights. The authors make some strong statements about their results, from a single case study and I think that they need to temper them. For instance, in the summary and conclusions, they state that “deep convective clouds act as a major mechanism for lifting INPs from the boundary layer into the free troposphere.” I am not disputing that DCC can transport INPs from the BL into the FT, but calling it a major mechanism is too strong, based upon the results from one research flight. I would also say that they did not find that

INP outflow from DCC "can influence ice formation in cirrus clouds, and after sedimentation downstream, in mid-level mixed-phase clouds." I agree that this may happen, but don't think the authors demonstrated that happening in this study. Wording here and throughout the manuscript should be revised where appropriate.

We have revised and tempered the wording in the conclusion to the following, so that it conveys this study's message appropriately, also taking the comments from reviewer 2 into account. Line 389 - 404:

> *The effect of ~~DCCs~~ deep convection on INPs is showcased utilizing research flight F15, during which INPs were sampled at different stages of ~~DCCs~~ deep convective clouds, i.e., in the convective inflow at low levels, within ~~DCC~~ anvil cirrus, and in the convective outflow into the UTLS. ~~In essence we found clear indications that:~~In essence, our case study reveals two key findings:*
>
> - *~~Deep convective clouds act as a major mechanism for lifting~~ Deep convective clouds can act as a lifting mechanism for INPs from the boundary layer into the free troposphere ~~, where they can~~ . Although not shown in this study these lifted INPs may influence ice formation in cirrus clouds (Luebke et al., 2016) and, after sedimentation downstream, in mid-level mixed-phase clouds.*
> - *During convective transport, INPs are segregated according to their freezing temperatures, i.e., INPs active at warmer temperatures ~~are~~ appear to be preferentially removed by precipitation, while colder-temperature INPs survive transport to the UTLS.*
>
> *Overall, deep convection ~~should be considered as~~ may represent an important transport and transformation pathway of INPs from lower altitudes to the free troposphere which ~~needs to~~ should be explicitly resolved or adequately parameterized in atmospheric and climate models.*

Second, it appears to me that there is no significant difference between the activated fraction of warm INPs in the inflow vs. in-cloud residuals (Fig. 5a). At the very warmest temperatures (-10 ℃) the in-cloud sampling has higher INP concentrations than the inflow, and when the inflow is higher than the in-cloud, it appears to be within the measurement uncertainty.

At warmer temperatures (-10 ℃) the INP concentration is very uncertain due to poor counting statistics of only a few frozen wells in the offline experiment. This uncertainty propagates to the INP fraction, so no reliable statement can be made for INP fraction at -10 ℃. To show this more clearly, we have changed the lower limit of the y-axis in Fig. 5a to show the very large uncertainty of the INP fraction at higher temperature, particularly of the in-cloud sample.

Additionally we have clarified the sentence (Line 308 - 310):

> *~~"Sampling of CPRs within DCCs during other CIRRUS-HL research flights resulted in activated INP fraction spectra of similar shape (see light blue curves, Fig. 5b, shading same as in panel a), i.e., a steep increase between ~ -13.5 and ~ -16.5 ℃ and very few INPs efficient above -14 ℃"~~*
>
> *"Sampling of CPRs within DCCs during other CIRRUS-HL research flights resulted in activated INP fraction spectra of similar shape (see light blue curves, Fig. 5b, shading same as in panel a), i.e., a steep increase between ~ -13.5 and ~ -16.5 ℃ with the fraction of INPs active above -14 ℃ remaining comparatively small"*

We also corrected the sentence in the abstract accordingly while also addressing a comment of reviewer 2, from (Line 8 - 9):

> ~~*While INP-temperature spectra of convective inflow included both INPs active at high ($T > -15$ °C) and low temperatures ($T < -20$ °C), the in-cloud and outflow spectra only featured INPs active at low temperatures.*~~
>
> *INP-temperature spectra of convective inflow included both INPs active at high ($T > -15$ °C) and low temperatures ($T < -20$ °C). In contrast, the outflow spectra only featured INPs active at low temperatures.*

Finally, the single particle composition aspect of this paper is under-developed. For instance, dust particles are observed in-cloud and also at the DCC outflow. Is the mixing state of the dust particles that make it to the outflow systematically different from those found in-cloud? It would be interesting to know whether the authors considered this in their analysis.

Just to be sure that there is no misunderstanding, we would like to point out that we measured the single particle composition of aerosol / residual particles, not of INP. To make this more clear we made changes in the introduction and discussion (Line 65 - 69):

> *In conjunction with particle sampling by HERA, simultaneous measurements of the chemical composition of particles (aerosol particles and cloud particle residuals) by single particle mass spectrometry and further particle properties, such as number size distribution (Voigt et al., 2022), were carried out. These measurements are used to chemically characterize the aerosol particles prevailing, as well as their potential sources. It is not possible to draw conclusions concerning the chemical composition of the INP present due to their low number concentration fraction.*

And line 360 - 364 from:

> ~~*Single particle chemical analysis in CPR suggests that these low-temperature INPs are mostly mineral dust (Fig. 4d). However, total INP concentrations are at least 2 orders of magnitude lower than total CPR number concentrations. Consequently, identifying specific INPs and their respective type (e.g., mineral or biological) is difficult.*~~
>
> *Single particle chemical analysis of CPRs shows an enhanced particle type fraction of mineral dust mineral dust (Fig. 4d). Given that mineral dust particles are very efficient INPs (DeMott et al., 2003) in the relevant temperature range of this study, they are the most likely candidate for the observed freezing activity in the in-cloud sample. However, since total INP concentrations are at least 4 orders of magnitude lower than total CPR number concentrations, the bulk CPR composition alone is not sufficient to unambiguously identify the INP subset.*

Additionally we have expanded the analysis of the single particle mass spectrometry in a new section "Chemical signature of selected particle groups" in the supplement. In there we give more details regarding the single particle mass spectra of BBA and mineral dust, with the latter being a likely candidate acting as INP.

While this additional analysis is interesting in many aspects and generally consistent with the other findings presented in this study, i.e. upward transport and processing of aerosol particles (including INP), with the obtained data of one campaign, we are not confident that we can make further detailed statements about the chemical composition of INP for the reasons mentioned above. We hope to expand upon this analysis of mineral dust chemical composition, mixing state etc. in future as more campaigns with ALABAMA and HERA have been carried out and are planned. Also, another manuscript will be submitted soon with more information about the chemistry of CPR during CIRRUS-HL with a focus on the high fraction of organics in ice crystal residuals.

Specific comments

Line 51: HALO needs to be parenthesized.

Changed accordingly.

Line 63: Cziczo et al. (2004) also measured composition of INPs in the free troposphere.

Yes, the reference is now included.

Line 81: I think it would be worthwhile to change the name of HASI to not include submicrometer. If it has been modified to also measure supermicron particles, it seems like a potential source of unnecessary confusion.

We agree that it is an unnecessary source of confusion; however, we would like to stay consistent with the inlet naming as in the CIRRUS-HL overview paper (Jurkat-Witschas et al., 2025), where it is unfortunately also named HASI. For studies of following campaigns we will try to improve this. To reduce potential confusion we supplemented the name "HASI" with "aerosol inlet" where possible, and changed the sentence (line 82 - 85):

> ~~*Despite what is suggested by the inlet's name, HASI was modified to a single sampling line supplying all instruments to facilitate sampling of supermicron aerosol particles.*~~
>
> *The aircraft aerosol inlet is a modified version of the HALO submicrometer inlet (HASI). It was adapted from a previous setup featuring multiple sampling lines (Andreae et al., 2018) to a single sampling line supplying all instruments to facilitate sampling of supermicron aerosol particles (Grawe et al., 2023) with a total flow rate of up to 80* $l\,min^{-1}$.

Line 124: The sentence starting "A lower size. . . " is awkwardly constructed, rewrite for clarity.

Agreed. We changed the sentence from line(125 - 127):

> ~~*A lower size limit of 500 nm was chosen as the cumulative number concentration for particles > 500 nm was shown to correlate broadly with INP concentrations (DeMott et al., 2010).*~~
>
> *We set the lower size limit to 500 nm, motivated by the broad correlation between number concentrations of particles larger than 500 nm and INP concentrations reported by DeMott et al. (2010).*

Line 133: Vali formula given in SI, worthwhile to reference this fact.

Agreed, reference added.

Line 163: What does S2 refer to? Is it a figure or table?

This was meant to reference Fig. S2. Thank you for spotting this; we changed it accordingly.

Line 221: No closing parentheses.

Parenthesis added.

Line 234: This whole paragraph has a number of missing parentheses. I am going to stop noting these, but please go through and carefully check the rest of the manuscript.

We apologize for this and added the missing parentheses.

Line 248: I thought that the

The editor reached out to the referee about this comment and we were told to ignore it.

Line 287: Figure 5d title is ‘outflow’ not ‘inflow’, and there is no red line. Do you mean 5c?

Yes, both things are corrected now.

Line 387: I think that the authors showed that DCC can act as a lifting mechanism for INPs, but they did not find a clear indication that “they can influence ice formation in cirrus clouds, and after sedimentation downstream, in mid-level mixed-phase clouds.” I agree that this is likely to happen, but don’t think the authors demonstrated that happening in this study. Would recommend rewording this point to clarify the distinction.

As mentioned above (RC1, General Comment 1) we revised the conclusion and changed the section also taking this comment into account (Line 389 - 404)

Figure 4: Panel (a) altitude is in hPa, could you add a scale showing it in m as well. There is also a blue line that looks like an isotherm in this panel, what is it?

We added a scale on the right hand side showing the approximate altitude of the model pressure levels. This pressure to altitude conversion was obtained from fitting GPS altitude to the static pressure measured on the aircraft on that day and is hence somewhat inaccurate but should give a rough indication of altitude.

Supplement: It would be more reader-friendly to add the list of abbreviations to the main text.

We have added the list of abbreviations to the appendix of the main text.

Comment of Referee #2

Summary

This manuscript addresses an important and timely topic—the vertical transport and distribution of ice-nucleating particles (INPs) by deep convective clouds (DCCs). The study presents valuable in situ vertical observations of INPs in the UTLS and around deep convective clouds, a type of field measurement that remains relatively scarce. This confers good scientific merit to the work. However, several issues still require revision, particularly those concerning data availability and the main conclusions. Overall, the manuscript is suitable for publication in ACP after careful revision.

Thank you for your very constructive review and helpful recommendations in general. We will answer the specific comments below. This also includes remarks of referee 1.

General comments

1. I agree with the general physical reasoning that deep convection is capable of transporting boundary-layer INPs upward to upper atmospheric layers. However, the conclusion stating that "Deep convective clouds act as a major mechanism for lifting INPs from the boundary layer into the free troposphere" and "Overall, deep convection should be considered as an important transport and transformation pathway of INPs from lower altitudes to the free troposphere" are overstated, given that this interpretation is exclusively derived from a single research flight. Please revise such strong categorical statements throughout the manuscript to align with the limited observational basis.

We have tempered the wording in the conclusion to the following, so that it conveys this study's message appropriately. This also includes remarks of referee #1 and changes as highlighted above (RC1, General Comment 1).

> *The effect of ~~DCCs~~ deep convection on INPs is showcased utilizing research flight F15, during which INPs were sampled at different stages of ~~DCCs~~ deep convective clouds, i.e., in the convective inflow at low levels, within ~~DCC~~ anvil cirrus, and in the convective outflow into the UTLS. ~~In essence we found clear indications that:~~In essence, our case study reveals two key findings:*
>
> - *~~Deep convective clouds act as a major mechanism for lifting~~ Deep convective clouds can act as lifting mechanism for INPs from the boundary layer into the free troposphere ~~, where they can~~ . Although not shown in this study, these lifted INPs may influence ice formation in cirrus clouds (Luebke et al., 2016) and, after sedimentation downstream, in mid-level mixed-phase clouds.*
> - *During convective transport, INPs are segregated according to their freezing temperatures, i.e., INPs active at warmer temperatures ~~are~~ appear to be preferentially removed by precipitation, while colder-temperature INPs survive transport to the UTLS.*
>
> *Overall, deep convection ~~should be considered as~~ may represent an important transport and transformation pathway of INPs from lower altitudes to the free troposphere which ~~needs to~~ should be explicitly resolved or adequately parameterized in atmospheric and climate models.*

2. I agree with the authors' finding that the in-situ measured INP concentrations aloft are roughly two orders of magnitude lower ($\sim$0.01–0.1 L^{-1}) compared with previous ground-based observations. However, the observed ice crystal number concentrations in the atmosphere are substantially higher than the measured ambient INP levels (3–4 orders). It would be helpful if the authors discuss and elaborate on potential physical pathways responsible (such as secondary ice production...) for such abundant ice formation under low INP abundance in the upper troposphere.

We currently discuss secondary ice production (SIP) in the manuscript as a likely contributor to rapid glaciation of the DCC and as a plausible explanation for the observed sharp segregation of INPs by effective nucleation temperature in the convective outflow and anvil. However, invoking our INP measurements and assumptions of SIP to explain the absolute ice crystal number concentrations in the upper troposphere is a comparison that we deliberately avoid, for the following reasons:

- The INPs we measured are active between -10 and -27 °C and are therefore most likely not the only relevant nuclei for ice formation at UTLS temperatures. Other processes may contribute to ice crystal abundances in the anvil and upper troposphere, including immersion freezing by INPs active at lower temperatures (-27 to -38 °C), deposition nucleation and/or pore condensation freezing, and homogeneous freezing below -38 °C. Disentangling the contributions of these processes from that of SIP is not possible with our data.
- The microphysical pathways governing SIP in deep convective clouds remain poorly constrained by in-situ observations (Lawson et al., 2017) and laboratory experiments (Korolev and Leisner, 2020), and we do not consider the current state of knowledge sufficient to make quantitative statements about its contribution in this specific case.

SIP by definition does not produce or consume insoluble INPs – it multiplies ice crystals without altering the INP budget. Our study focuses on INPs rather than total ice crystal numbers. While SIP likely influences INPs by accelerating cloud glaciation, it is of limited direct relevance to our central analysis.

We acknowledge this complexity explicitly in the manuscript (line 375):

> *The microphysical pathways governing the interaction between INPs and DCC are inherently complex and remain incompletely understood, in part because they are poorly constrained by in-situ observations within active convection (Lawson et al., 2015; Han et al., 2024). Processes such as SIP, ice growth, aggregation, and phase partitioning of hydrometeors may all contribute to the observed INP characteristics, yet current knowledge does not allow these mechanisms to be quantified or disentangled in great detail for this case study.*

We therefore prefer not to expand this discussion further in the manuscript, as any additional statements would be highly speculative given the available data.

3. The authors performed a rather concise single-particle composition analysis, which is presumably aimed at constraining the chemical composition of INPs. However, the manuscript explicitly states "Unfortunately, the proportion of INP in the total number of aerosol particles is too low for establishing a robust link between INPs and specific particle types". Given this inherent limitation, the motivation for carrying out only a simplified single-particle analysis needs to be justified. Standard comprehensive single-particle characterization to unravel aerosol physicochemical processes routinely covers mixing state, surface coating property, core-shell structure, internal/external mixing status and elemental association information. The absence of such supporting information makes Figure 2 and its associated discussion confusing to readers.

Just to be sure that there is no misunderstanding, we would like to point out that we measured the single particle composition of aerosol / residual particles, not of INP. To make this more clear we made changes in the introduction and discussion as outlined in the answer to referee #1.

> *In conjunction with particle sampling by HERA, simultaneous measurements of the chemical composition of particles (aerosol particles and cloud particle residuals) by single particle mass spectrometry and further particle properties, such as number size distribution (Voigt et al., 2022), were carried out. These measurements are used to chemically characterize the aerosol particles prevailing, as well as their potential sources. It is not possible to draw conclusions concerning the chemical composition of the INP present due to their low number concentration fraction.*

And line 360 - 364:

> ~~*Single particle chemical analysis in CPR suggests that these low-temperature INPs are mostly mineral dust (Fig. 4d). However, total INP concentrations are at least 2 orders of magnitude lower than total CPR number concentrations. Consequently, identifying specific INPs and their respective type (e.g., mineral or biological) is difficult.*~~

> *Single particle chemical analysis of CPRs shows an enhanced particle type fraction of mineral dust (Fig. 4d). Given that mineral dust particles are very efficient INPs (DeMott et al., 2003), they are the most likely candidate for the observed freezing activity in the in-cloud sample. However, since total INP concentrations are at least 4 orders of magnitude lower than total CPR number concentrations, the bulk CPR composition alone is not sufficient to unambiguously identify the INP subset.*

The motivation for the simplified analysis of the UTLS background aerosol is mainly to put the INP population into context with the overall aerosol population. To make this clearer we added *To characterize the overall UTLS background aerosol particle population and with that potential sources for INPs,...* to the beginning of the paragraph. (Line 179)

Because both reviewers suggested a more comprehensive single-particle chemical composition analysis, we have expanded the analysis of the single particle mass spectrometry for the case study of F15 in a new section "Chemical signature of selected particle groups" in the supplement. There, we give more details regarding the single particle mass spectra of BBA and mineral dust, with the latter being a likely candidate acting as INP.

While this additional analysis is in many aspects interesting and generally consistent with the other findings presented in this study, i.e. upward transport and processing of aerosol particles (including INP), with the obtained data of one campaign, we are not confident that we can make further more detailed statements about the chemical composition of INP for the reasons mentioned above. We hope to expand upon this analysis of mineral dust chemical composition, mixing state etc. in future as more joint campaigns with ALABAMA and HERA have been carried out and are planned. For more information about the chemistry of CPR during CIRRUS-HL with a focus on the high fraction of organics in ice crystal residuals, another manuscript will be submitted shortly.

4. Figure 6: It needs to be confirmed whether the convective inflow of this studied convective case is indeed located on the western side of the storm. Considering Germany lies in the mid-latitude westerly belt of the Northern Hemisphere, organized mesoscale convective systems generally propagate eastward, with low-level inflow typically feeding the storm from its eastern leading flank. If the inflow position in Figure 6 is plotted oppositely to the structural feature, I suggest horizontally flipping (mirroring left and right) Figure 6 to conform to the canonical dynamical structure of European mid-latitude convection; the current layout appears physically inconsistent.

The figure was intended as a schematic without alignment according to wind directions. Howevaer, to make the figure more intuitively understandable, we mirrored the cloud according to the reviewer's suggestion.

5. 5. Line 400: Data availability. "Processed data from the CIRRUS-HL campaign will become publicly available on the HALO database (`http://doi.org/10.17616/R39Q0T`) at `https://halo-db.pa.op.dlr.de/mission/125` in June 2026. Earlier access to the data is available by contacting the principal investigators of the campaign." As the current calendar has reached June 2026, the processed INP datasets from the CIRRUS-HL campaign are still not publicly accessible via the provided HALO database link and DOI as stated in the manuscript. Please revise the data availability section and ensure the related INP data can be made accessible.

We apologize for providing a false date on this. The actual date for public access to the HALO database is 10 July 2026. By now, it should be publicly available. However, as the HALO database is currently being overhauled thereby frequently unavailable or with very poor performance we have decided to also upload the INP data to a Zenodo repository: https://doi.org/10.5281/zenodo.21156567

Specific comments

1. The manuscript employs an excessive number of abbreviations throughout the conclusion section and the main text, for example: "To distinguish the effect of DCCs in UTLS INPs," "Sampling of CPRs within DCCs during other CIRRUS-HL research flights...". Frequent and dense use of acronyms affects reading fluency and makes the overall narrative less accessible. It is recommended to appropriately replace some overused abbreviations with full terms where appropriate, especially for repeatedly mentioned concepts, to improve readability and logical smoothness.

We reduced the number of acronyms and abbreviations in the revised version of the conclusion, which addressed comments of both reviewers. The appendix of the main manuscript as well as the SI now include tables explaining the abbreviations.

2. In the abstract, the sentences starting with "While INP-temperature spectra of convective inflow...", "In contrast, INPs active at lower..." are quite long and could be split for readability.

Thank you for the advice. We changed the sentences to (line 8):

> *INP-temperature spectra of convective inflow included both INPs active at high ($T > -15$ °C) and low temperatures ($T < -20$ °C). In contrast, the outflow spectra only featured INPs active at low temperatures.*

3. Since the full name of CIRRUS-HL has already been provided in parentheses, for consistency it is recommended to also supplement the full name of HALO upon its first appearance for the readability of manuscript.

Changed accordingly.

4. The caption of Figure 5 could be better structured for readability. It is suggested to revise it by separately describing each panel in sequence, which will help readers understand the figure more easily.

We changed the caption according to your suggestions.

5. Line 163: It is unclear what "S2" refers to in the sentence "and the sampling altitude ranged between 5 km and 15 km at ambient temperatures from 0 to -65 ℃ (see S2)". Please define S2 explicitly (e.g., supplementary Figure S2, supplementary Table S2 or supplementary Text S2) and revise the citation format accordingly.

This was meant to reference Fig. S2, see also the answer to reviewer 1. Thank you for spotting this; we changed it accordingly.

6. Line 396: The slash symbol between "universal" and "generally valid" looks informal and ambiguous for formal manuscript writing. Please replace the slash with appropriate wording (e.g., "or") to polish this sentence.

Changed to *or*

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