Responses to the Reviewers

Format: The reviewer's comments are quoted in italic. Quotation in red color stands for revised/added text in the revised manuscript.

Overall comment:

We thank the reviewer for the detailed comments, especially after a long time since your first review. We have addressed individual comments from the reviewer in our response. Specifically, we have addressed these following main comments:

1. We improved the data quality of relative humidity measurements by applying a new data quality control procedure for in-cloud measurements with a high amount of supercooled liquid water. That is, when the 1-Hz sample is categorized as liquid-containing (i.e., LCR or MCR) and for that second either CDP probe or King probe reported LWC > 0.001 g m^{-3}, the RH_{liq} values are adjusted to liquid saturation. The RH_i and RH_{liq} distributions are shown in the new Figure 4.
2. We examined the data quality of vertical velocity, especially for cases when the research aircraft was doing vertical profiles or when it stayed at constant pressures. We found no drifting issues with vertical velocity measurements. But to be more cautious, we applied a new data quality control procedure to the analysis of σ_w (i.e., standard deviation of vertical velocity calculated for every 40 seconds), i.e., we remove the σ_w values when the research aircraft experienced dPressure > 10 hPa within those 40 seconds.
3. We also improved the calculation of ice particle number fraction (IPNF), which equals Nice / (Nice + Nliq). Specifically, we used an additional cloud imaging probe – the PHIPS probe, to verify cloud particle thermodynamic phases for each phase and each type of cloud region (LCR, MCR or ICR). We applied a series of new quality control procedures to remove the mistakenly reported ice or liquid from the UWILD 2DS data, which have significantly reduced the number of seconds with IPNF between 0.4 and 1, which are the scenarios that the reviewer was concerned about. New Figures 6 and 8 reflected such change, with IPNF values peak at 0 and 1. A sensitivity test to Figure 6 is also conducted by removing all high IPNF values, and our conclusions remain the same.
4. We revised the title again, reduced the ambiguity in our writing, and rephrased the text that the reviewer had concerns about.
5. We also revised the line colors for any line plots such as new Figures 3, 5, 8 to satisfy the color scheme requirement by the journal of Atmospheric Measurement Techniques (AMT).

Response to comments from the Reviewer

Review of revised "The Transition from Supercooled Liquid Water to Ice Crystals in Mixed- phase Clouds based on Airborne In-situ Observations" by Maciel et al.

Overview
I reviewed this paper more than a year ago, and it took me some time to go through my comments and replies and read the revised text. My original comments were split into three major categories: (a) Methodology and basic assumptions, (b) Data quality, and (c) Clarity or presentation. Many of my comments were addressed and clarified. However, after reading the revised text, I came across another set of issues. Most of the questions fall into the same three categories as stated above.

Recommendation: Unfortunately, the revised paper remains unsuitable for publication in ACP. I suggest another round of revision of the manuscript and addressing the comments listed below.

Major comments

1. This work is focused on the analysis of the link between the phase composition of clouds, cloud dynamics and aerosols. Clouds were split into four categories: pure liquid clouds (P1), conditionally mixed clouds consisting of spatially continuous liquid and genuinely mixed cloud segments (P2), conditionally mixed clouds consisting of spatially continuous segments of ice and (pure liquid and/or genuinely mixed phase) clouds (P3) and pure ice clouds (P4). In the first version of the paper, it was assumed that the direction of changes in the cloud thermodynamic state is as follows: (P1)=>(P2)=>(P3)=>(P4). However, as was indicated in the reviewer's previous comments, depending on the dynamic forcing, interaction between the cloud and ambient environment, and ice precipitation out of the cloud, the phase partitioning may go in any direction. The complexity of the interaction between three thermodynamic phases in clouds and its sensitivity to environmental conditions does not allow for simplified judgment about the evolution stage of the cloud. In this regard, the following statement in the conclusions (line 500-501) "Overall, the method proposed in this work provides a unique perspective to assess various evolution stages of mixed phase clouds, especially the transition from liquid to ice phase" is an overstatement. This paper does not contain discussion of the cloud evolution. All that could be said is that the sampled cloud belongs to the one of the four preselected categories P1-P4. Linkage to the dynamics and humidity obtained from the instant in-situ (Eulerian) observations does not allow for judgment about the history of the cloud environment.

We tuned down the description on evolution and only mentioned the coexistence between liquid and ice in this revised sentence in Section 4: "Overall, the method proposed in this work provides a unique perspective to assess mixed phase cloud properties in both macrophysical and microphysical perspectives, especially for phases when supercooled liquid droplets and ice particles coexist."

2. I have a hard time understanding the term "transition stage" throughout the manuscript. I brought this question up in my previous round of comments, however, I did not receive a clear answer. Employing the term "transition stage" implies that some clouds can exist in a non-transition stage. Generally speaking, any cloud can be described as an unstable colloidal system in a transition stage between water in gaseous and condensed stages (liquid and/or ice). There are several types of instabilities relevant to cloudy environments related to condensation/evaporation (e.g., due to dynamic forcing, entrainment & mixing, WBF, radiation effects, Ostwald ripening), mechanical interaction between particles (e.g., coalescence, aggregation, riming, fragmentation), and sedimentation. Each of these types of instabilities is characterized by its own time scale. Specifically, in relation to this study, the use of the term "transition stage" assumes a discussion of time scales such as time of phase relaxation, glaciation time, and residence time of cloud particles, along with different types of forcing. However, none of these points have been discussed. Therefore, the use of the term "transition stage" is redundant and may be misleading to the reader.

We did a global search and changed the term "transition phase" to just "phase" throughout the text to avoid confusion.

3. Per the previous comment, the title of the paper, "The Transition from Supercooled Liquid Water to Ice Crystals in Mixed-phase Clouds based on Airborne In-situ Observations" is misleading. The paper does not discuss the transition from liquid to ice. In fact, the transition of the thermodynamic phase may go in the opposite direction, i.e., ice to mixed-phase. This was also mentioned in section 3.1 and indicated in Fig 1. This conflicts with the title of the paper, implying a one-directional transition, "liquid to ice".

In our last revision, we changed the title to "Transition between Supercooled Liquid Water and Ice Crystals in Mixed-phase Clouds based on Airborne In-situ Observations".

Since this may still be a bit misleading, we further changed the title of the manuscript to: "Partition between ~~Transition from~~ Supercooled Liquid ~~Water~~ Droplets and ~~to~~ Ice Crystals in Mixed-phase Clouds based on Airborne In-situ Observations".

4. Lines 505-507: "Nevertheless, this method helps to provide a statistical categorization of different transition phases of mixed-phase clouds solely based on Eulerian-view sampling of aircraft data, which enables more detailed examination from a statistical, quasi-Lagrangian view that was not available previously." There was no "quasi-Lagrangian" consideration of mixed-phase in the text, and this statement at the end of the paper is unexpected and confusing. I also have a hard time understanding how quasi-random sampling of clouds (e.g. Eulerian) can be linked to a quasi-Lagrangian consideration. What are the time and spatial scales of the quasi-Lagrangian consideration referred to?

We revised this sentence to stay with the statistical analysis perspective of this approach: "Nevertheless, this method helps to provide a statistical categorization of different phases of mixed-phase clouds solely based on Eulerian-view sampling of aircraft data. Future studies may derive such statistical distributions of phases based on 2-D remote sensing observations and 3-D model simulations. Examining individual phases of mixed-phase clouds may also provide more direct comparisons between observations and simulations."

5. Lines 267-270: "Comparing RHi values in regions with and without ice, phase 2 shows higher RHi for regions with ice, while phase 3 shows higher RHi in regions without ice. This feature can be explained by the fact that higher RHi is required in order to initiate ice nucleation in phase 2, while ice crystals that continue to grow in phase 3 will further reduce RHi magnitude by vapor deposition." This is an unjustified statement. I believe the authors meant the dependence of INP nucleation on supersaturation. However, supersaturation in phase 2 (and any other type of cloud) is limited by saturation over liquid. This fact mitigates or eliminates the dependence of INP nucleation vs. vertical velocity in liquid and mixed-phase clouds. On the other hand, the differences between RHliq in P2 and P3 are within 1-4%. This is smaller than the accuracy of RH measurements (i.e., 6% - 7%). It applies limitations on relating these differences to physical processes, and it can be explained just by the error in RH measurements.

We agree that the previous comment was an overstatement and removed it. In addition, after carefully thinking about the layout of the original Figure 5, we decided to remove this figure because it has many bins with a low number of samples, yet it gives the misleading look as if all the bins share similar significance. Since the relative humidity and vertical velocity are also shown in the new Figure 4, these two figures also become too repetitive. After removing Figure 5, the original text in Section 3.2 has been re-written, with descriptions focusing on new Figure 4 and the new supplemental Figure S4.

6. As can be seen from Figure 3a, the sampling statistics of measurements are distributed quite unevenly across the temperature range and cloud types P1-P4. The lengths of different cloud types in different temperature subranges vary from approximately 850km down to 1km or less. The points with low sampling statistics (e.g., less than 100 of 1Hz samples ~17km) have low statistical significance. This should be clearly discussed in the text.

We thank the reviewer for the helpful comment. We added this discussion in Section 3.1: "The lengths of different phases vary from ~0.2 – 180 km in various temperature ranges, with low sampling statistics (i.e., less than 100 seconds) of continuous in-cloud segments longer than 3.5 km, which indicates a patchy horizontal structure with clear-sky gaps inside the clouds."

7. Figs. 4 and 5 include clouds P2 and P3 with subdivisions in clouds "with ice" and "without ice". I have a hard time understanding what it means. Does it mean that P2 "without ice" are just liquid clouds with mixed-phase cloud regions (MCR) excluded from the data set? Whereas clouds P2 "with ice" are just P2 *clouds? Does it mean that clouds P3 "without ice" are clouds with excluded MCR and ICR? Or just with excluded ICR? Does it mean that P3 "with ice" is just P3 clouds or something else? With this ambiguity in the interpretation of the meaning of P2 & P3, "with ice" and "without ice" I found it difficult to follow the subsequent discussion.*

We added more clarifications in Section 3.2 when describing Figure 4: "For phases 2 and 3, LCR represents seconds without ice particles, while MCR and ICR represent seconds with ice particles. These two conditions (i.e., without or without ice) are separately examined in Figure 4 e-h and m-p." For example, in the diagram in Figure S1, a TCR labelled as phase 3 has 3 seconds of LCR and 4 seconds of ICR, therefore these 3 seconds of LCR represent "without ice", while the 4 seconds of ICR represent "with ice".

8. Fig.3e shows that humidity in pure liquid clouds (P1, indicated by a red dot) is saturated over ice. This contradicts previous studies of humidity in liquid clouds (Korolev and Mazin, JAS, 2003; Korolev and Isaac, JAS, 2006, D'Alessandro et al., J.Clim., 2021). Something is fundamentally wrong here.

We thank the reviewer for this fundamental suggestion. We believe the reviewer was referring to Figure 5e instead of Figure 3e. We have conducted more examinations about the relative humidity measurements and applied a new data quality control procedure, which is described in Section 2.1: "For RH_{liq} lower than 100%, an adjustment to 100% is applied if two criteria are satisfied for a 1-Hz sample: 1) it contains supercooled liquid water and 2) either CDP or the King probe measures LWC greater than 0.001 g m^{-3}."

With this new quality control procedure, the **new Figure 4** is copied below. It shows much improved distributions of RH_{liq} for segments containing supercooled liquid water, such as phase 1 (Figure 4 a), phase 2 (Figure 4 b), and phase 3 without ice (Figure 4 h). The RH_{liq} distributions of these conditions now show high frequencies around liquid saturation at various temperature ranges (new Figure S4 b). Section 3.2 is also re-written. In addition, as mentioned above, the old Figure 5 from the last submitted manuscript (the Nov 25, 2023 version) has been removed to reduce the redundancy with new Figure 4, as well as eliminating the misleading features from ice spatial ratio bins that have too few samples.

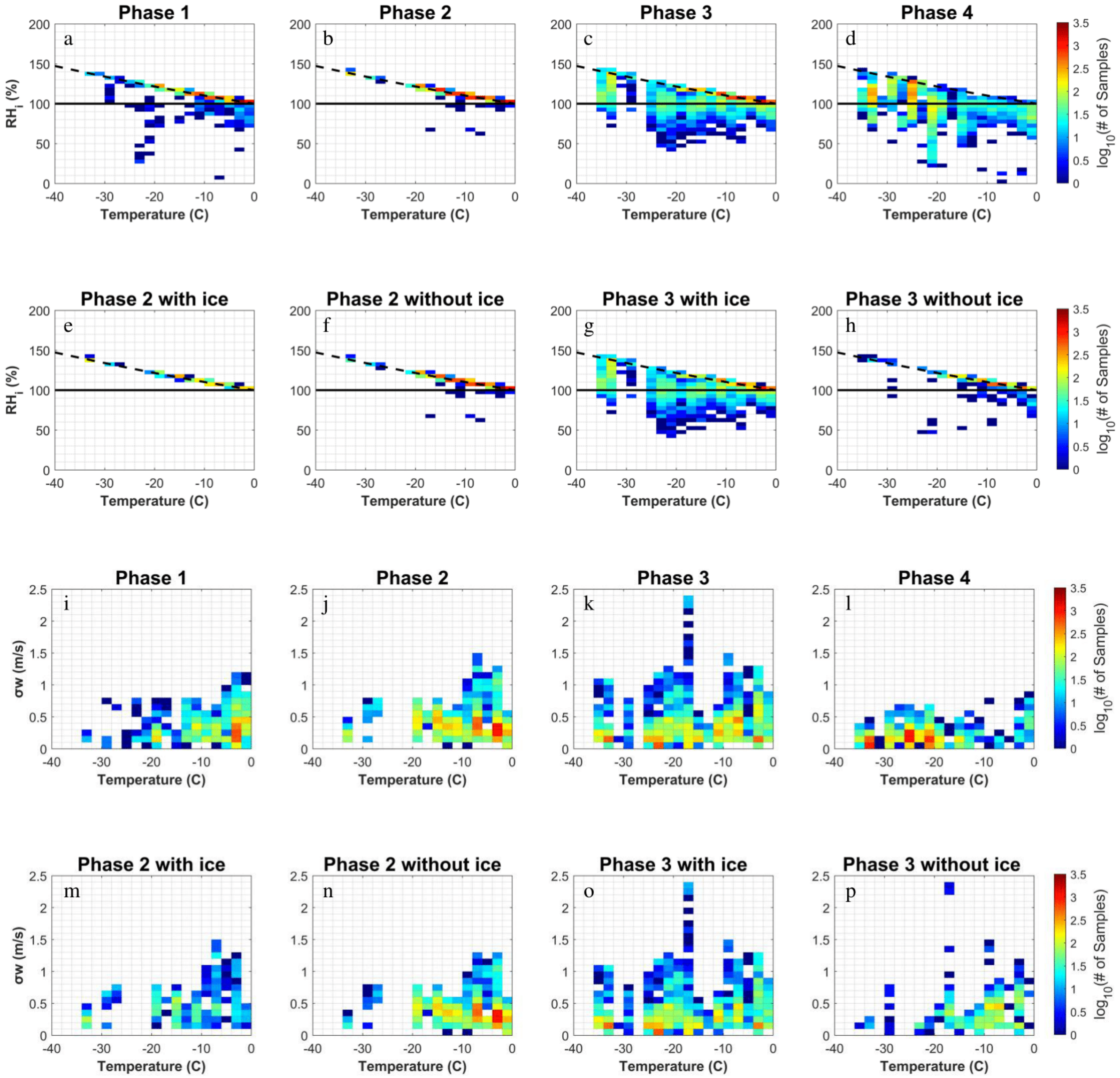


Figure 4. Distributions of (a-h) RH$_i$ and (i-p) σ_w in various phases as a function of temperature. Dashed lines in (a) – (h) indicate liquid saturation.

9. The diagram in Fig.S6a shows that the PDFs of RHi in liquid and mixed-phase clouds from this study are centered at saturation over ice, i.e., RHi=100%. This result is overly concerning. It raises many questions about the accuracy and data quality of RH measurements and results presented in the paper.

We appreciate the concern from the reviewer. Upon the additional quality control to the RH data as mentioned in the above comments, we have also updated that previous supplemental figure, which is now the new supplemental **Figure S4** (copied below). Liquid and mixed phase clouds now show the peak frequency of RH$_{liq}$ at liquid saturation for phases 1 – 3 (Figure S4b).

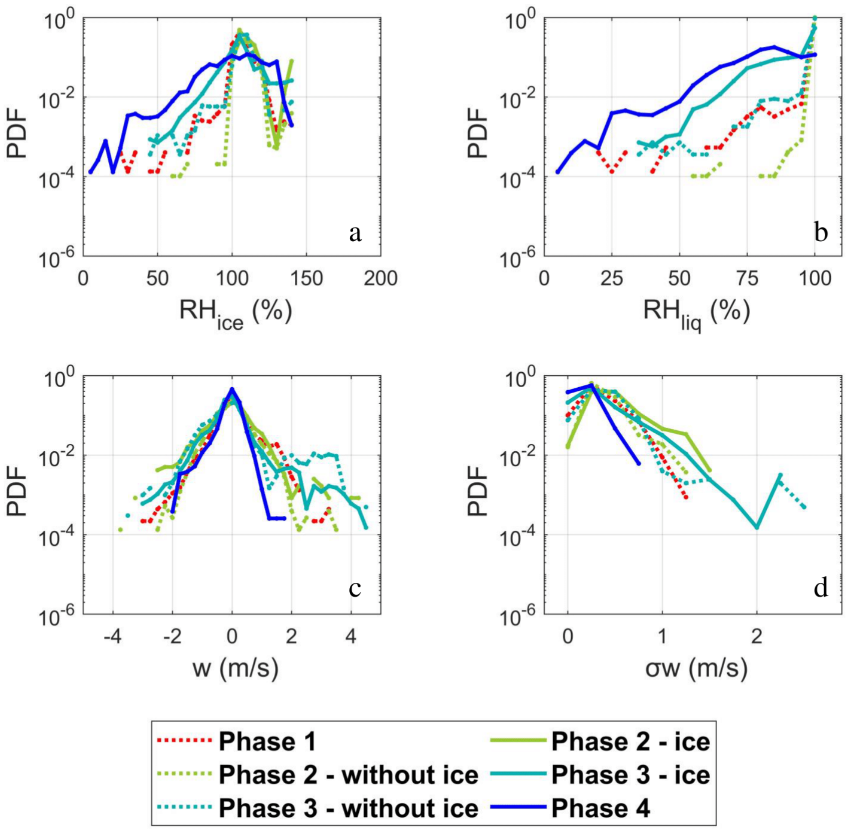


Figure S4. Probability density functions (PDFs) of (a) relative humidity with respect to ice, (b) relative humidity with respect to liquid, (c) vertical velocity (w), and (d) standard deviations of w (σ_w) separated by different phases. Phases 2 and 3 are further separated into seconds with or without ice in this analysis. Both phases 2 and 3 show higher frequencies of updrafts and σ_w compared with phases 1 and 4

10. Could you please double-check that the points with sigma_w=1+ m/s for P2 clouds in Fig.3h are not related to the malfunctioning of the LASEREF? This point looks suspicious and very different from the rest of the points. This is an overly high value, which is relevant for strong convection. If the data quality of these points is justified, could you check the type of clouds?

We thank the reviewer for the helpful comment. We believe the reviewer is referring to the old Fig. 5h, which shows a sudden spike of σ_w value in a specific ISR bin. We took several steps to address this comment.

First, we examined this specific spike with a high σ_w value above 1 m/s. We located this high vertical velocity fluctuation in RF03 from UTC 02:34:39 to 02:34:45, which lasted about 6 seconds. Upon inspection, these few seconds of measurements look good and legit, and they occurred when the plane was flying horizontally. Because of the way that the old Figure 5 was plotted at various bins of ISR, this bin of ISR only has a few seconds of data containing these few seconds of high vertical velocity values, which leads to the high spike of σ_w value. This is also another example that made us realize that the old Figure 5 is not a good representation of the entire dataset, since some bins may only represent a few seconds of data. That old Figure 5 is removed and replaced by the **new Figure 4**. Second, we examined the data quality of vertical velocity, especially during the ascent or descent legs. We applied a new quality control procedure for σ_w value, which will be described in detail in the following comment.

11. Lines 273-275: "For distributions of w in Figure 5 c, phase 1 has slightly higher w than other phases. Phases 2 – 4 show slightly negative average w values, suggesting weak downdrafts as the average condition in these phases." This is a concerning statement. Subsiding clouds at the rate of 10cm/s to 40cm/s (Fig.5c) will dissipate within 5 to 20 minutes with initial LWC=0.1g/m3 at T=-10C. This estimated time scale of cloud dissipation is shorter than the sampling time of the cloud during the flight observations. After my previous comment about vertical measurements, the authors found a negative bias of the vertical velocity measurement in the clear sky at 0.125m/s. Along this way, could you please check the drift of w in the clear sky? Note that measurements of vertical wind during ascending, descending, and any other type of aircraft maneuvering may result in significant biases of w. It is also worth mentioning that the LASEREF is an internal system, and the vertical wind velocity is calculated from the aircraft acceleration, i.e., the aircraft body is used as a sensor. The accuracy of such measurements is relatively low.

That old statement about downdraft was referring to old Figure 5, which has been removed. We further investigated the question from the reviewer, which is, would the vertical motion of the aircraft during ascent or descent cause any drifting in the vertical velocity measurements. We first examined the time series during ascent and descent, compared with the horizontal legs, and we found no indication of drifting of vertical velocity during rapid ascent or descent. Next, we plotted the distributions of vertical winds against the maximum pressure differences (dP) seen in a duration of 10-second, 20-second, 30-second, or 40-second periods, and found vertical velocity distribution is centered at 0 for all these dP ranges (shown in **Figure R1** below). But to be on the cautious side, we removed σ_w values when high dP values (> 10 hPa in 40 seconds) were observed.

We added this comment in Section 2.1: "To minimize the impacts of ascent and descent and the possible associated biases of vertical velocity measurements, we restrict the analysis of vertical velocity fluctuations (i.e., standard deviations of vertical velocity, calculated for every 40 seconds) to segments where the maximum pressure difference (dP) within 40 seconds is less than 10 hPa."

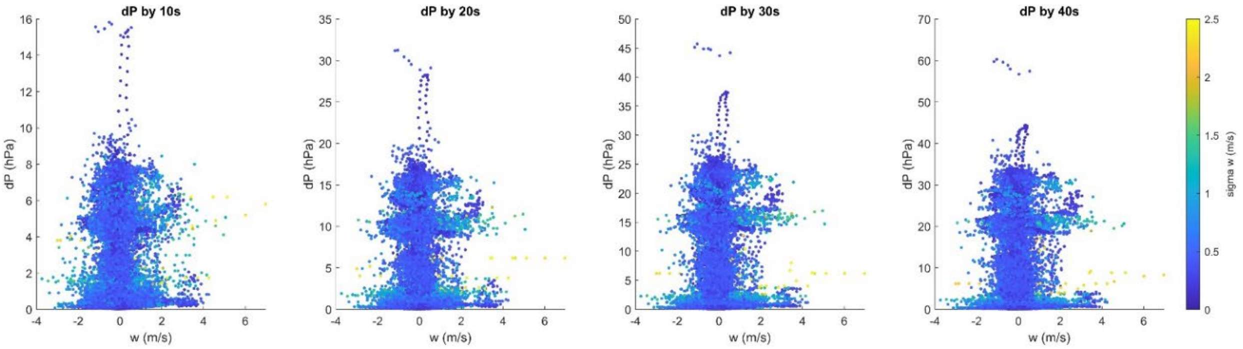


Figure R1. Scatter plots of the relationships between vertical velocity (w, m/s) and the maximum changes of pressure (dP) calculated within a duration of 10, 20, 30, and 40 seconds, color coded by σ_w values.

12. Lines 275-276: "For the σw distribution (Figure 5 d), regions with ice in phase 2 have the highest fluctuations of vertical velocity, indicating that stronger in-cloud turbulence induces high RHi (as shown in Figure 5 a), which further initiates ice nucleation in phase 2." The last part of this statement relates the rate of INP nucleation with the vertical velocity, which induces higher RHi. This is an unjustified statement. Note that humidity in liquid clouds is always limited by quasi-steady supersaturation (i.e., Korolev and Mazin, 2003). In other words, in liquid clouds RHliq ~= 100%+ for a wide range of w. This is suggestive that the rate of the INP nucleation does not depend on w.

We revised Section 3.2 and removed Figure 5. That former comment which was unjustified has also been removed.

13. Lines 278-279: "Such result is consistent with the finding of Buhl et al. (2019) which showed a positive correlation between IWC mass flux and vertical velocity fluctuation, but this study further illustrates that in-cloud turbulence is particularly important for transition phase 2 when ice crystals first start to appear inside MCR, surrounded by supercooled liquid water." There are several observational studies that show a correlation between ice and vertical velocity. However, the statement "when ice crystals first start to appear inside MCR, surrounded by supercooled liquid water" is an overstatement. This study did not present evidence to support it.

We revised Section 3.2 and that former statement was removed.

14. Fig. 5f shows that for P2 and P3 clouds, in most cases RHliq(no ice) <RHliq(with ice). Assuming that "no ice" category means "liquid" the results presented in Fig.5f contradict fundamentals of mixed-phase clouds, i.e., for the same environmental conditions, humidity in pure liquid clouds is expected to be higher compared to that in mixed-phase clouds. I am not sure what the cause of the obtained inequality is. However, in view of the importance of this result, an explanation of this phenomenon is required.

We addressed the concern over RH measurements in our comments above. The new **Figure 4** shows that RH_{liq} peaks at liquid saturation for phases 1 and 2, as well as for 1-Hz samples without ice in phase 3.

15. Lines 324-325: "… because ice crystal growth may occur via various processes in phase 3, such as … vapor depositional growth under ice supersaturation…" This is not true. For RHi=100%, $dM_{ice}/dt=0$.

We revised that sentence: "…probably because ice crystal growth may occur via various processes in phase 3, such as WBF process, glaciation, and/or riming."

16. In my previous comment, I brought up a concern regarding using the ice concentration fraction $\lambda_{ice}=N_{ice}/(N_{liq}+N_{ice})$ *for the analysis on mixed-phase, where* N_{liq} *and and* N_{ice} *are the concentrations of droplets and ice particles, respectively. For most mixed phase cloud* $N_{liq}>>N_{ice}$ *by a few orders of magnitude. Therefore, it is expected that in liquid clouds and mixed-phase clouds* $\lambda_{ice}\approx 0$*, whereas in ice clouds* $\lambda_{ice}\approx 1$*. The diagrams in Figs.7acd are consistent with this prediction, i.e. the cloud particle concentration in mixed-phase clouds is dominated by liquid droplet and therefore,* $\lambda_{ice}\approx 0$*. However, diagrams in Figs.7bf caused questions. These diagrams show* λ_{ice} *vs ice spatial ratio in P3 liquid and ice cloud regions (LCR & ICR). In Fig.7b ice concentration fraction varies in the range* $0 < \lambda_{ice} < 0.4$*, and in Fig.7f* $0.3 < \lambda_{ice} < 0.9$*. This is confusing, since in LCR the ice concentration fraction is expected to be* $\lambda_{ice}\approx 0$*, whereas in ICR* $\lambda_{ice}\approx 1$*. For the sake of argument, consider a case with* $\lambda_{ice}\approx 0.4$*. Then, for the case of LCR (Fig.7b), the droplet concentration typical for the SOCRATES clouds* $N_{liq} = 100\ cm^{-3}$ *the concentration of ice particles will be* $N_{ice} = \lambda_{ice} N_{liq} /(1- \lambda_{ice}) \approx 67\ cm^{-3}$*. To the best of my knowledge, such high concentrations of ice have never been reported in scientific literature. On the other hand, the presence of ice in P3 LCR raises questions about the accuracy of the identification of liquid cloud segments in P3. For the case of ICR (Fig.7f) assume* $N_{ice} = 100 L^{-1}$ *(high end of ice concentration). Then* $N_{ice} = N_{liq} (1- \lambda_{ice}) / \lambda_{ice} = 0.15\ cm^{-3}$*. This is an overly low concentration of liquid droplets. Such clouds are volatile, and they may exist at cloud interfaces, and their lifetime scale is expected to be short. Another question is related to measurements of such clouds. If the measurements were performed in ice clouds, then both CDP and 2DS can be contaminated by shattering artifacts, which can be confused with liquid drops. Please note, that both antishattering tips and antishattering algorithms are not capable of 100% filtering out all shattering artifacts (Korolev et al. JTECH, 2013).*

We appreciate the reviewer for pointing out this issue. We took several weeks to investigate this issue. In a quick summary, we have a few key findings:

1. Almost all the high ice particle number fraction (IPNF) ≥ 0.4 occurred when the UWILD 2DS data were the only contributor to the particle measurements (i.e., a total of 5273 seconds), while the CDP probe reported 0 or NaN. Only 4 seconds are the exception, when IPNF ≥ 0.4 and the CDP probe reported non-zero values.

2. Because the main issue happens with UWILD 2DS data, we applied more quality controls to that dataset. Please note that we initially used the published data from the UWILD paper (Atlas et al., 2021) and the authors acknowledged the fact that at lower temperatures and smaller dimensions there are higher chances of misidentification by the machine learning model.

   We examined cloud images from a new cloud probe, the PHIPS probe, when it has concurrent measurements as the 2DS probe. Please note that the PHIPS probe does not report cloud images for every second when 2DS data have values. Also, the PHIPS probe may have an underestimation of the particle number concentrations compared with 2DS and CDP probes, so we cannot use the PHIPS cloud probe images as a quantitative measure for Nice or Nliq per second. But we use it here to verify concurrent seconds when UWILD 2DS data reported values.

   We found that some of the UWILD 2DS particle phase identifications have misidentified real ice particles (usually small ice fragments) as liquid droplets. This type of misidentification often happens at temperatures below -20°C, when there are numerous small ice fragments with a small dimension (e.g., < 20 micron) and a shape very close to a sphere.

   On the other hand, we also found real valid measurements when IPNF is relatively high, when a few supercooled liquid droplets are surrounded by more ice particles. These samples are often in ICR at higher temperatures. Below in **Figure R2** we show a few examples of PHIPS images when we found IPNF values between 0.2 and 1 and they look realistic.

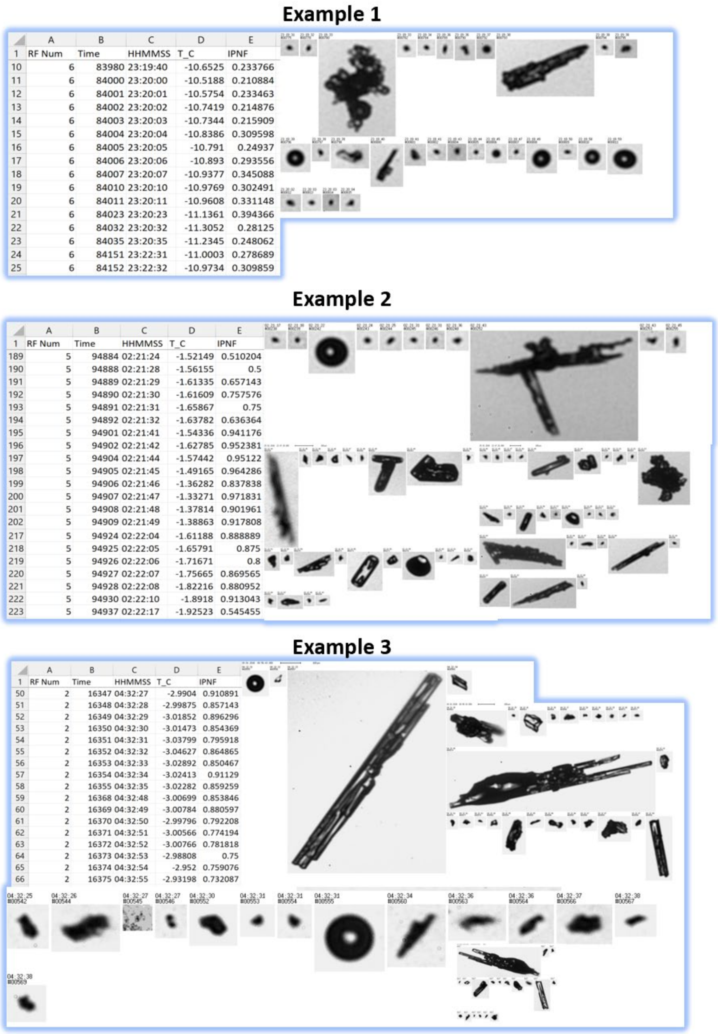
3. With these new examination results using the PHIPS probe, we applied the following quality control procedures to the UWILD 2DS data, added to Section 3.3: "Note that additional quality control procedures are applied to the ice particle number fraction (IPNF) data, because the machine-learning based particle identifications of 2DS data may misidentify small ice fragments as supercooled liquid droplets, especially at lower temperatures. To minimize such misidentifications, the following two quality control procedures are applied, which are developed after inspecting the Particle Habit Imaging and Polar Scattering (PHIPS) airborne cloud probe: (1) for 1-Hz samples of ICR in phase 3 and 4, when temperatures are below -20°C and 0 < IPNF < 1, IPNF is reset to 1 to be pure ice. In addition, for 1-Hz samples of ICR in phase 3, when temperatures are between -20 and -10°C and 0.4 < IPNF < 1, these IPNF values are reset to 1."

4. We conducted a sensitivity test in **Figure R3** below, by completely removing the high INPF between 0.4 and 1 that may be complicated by the difficulties of particle identification of small hydrometeors (i.e., small ice versus liquid droplets). The main findings of positive correlations between the ice particle number fraction and mixed or ice spatial ratio from Figure 6 remain unchanged. In addition, the main finding that phase 3 shows higher slope values than phase 2 is also unchanged. We mentioned this sensitivity test in the text in Section 3.3: "Note even after quality control is applied to IPNF, a small amount of high IPNF values is still seen (e.g., 0.4 ≤ IPNF < 1) in Figure 6 b and f. A sensitivity test is conducted by removing all 0.4 ≤ IPNF < 1 in Figure 6 and the results show consistent conclusions, that is, all phases show positive correlations between IPNF and the spatial expansion of ice-containing regions. In addition, phase 3 still shows

higher slopes of linear regressions compared with phase 2, indicating faster increases of IPNF in phase 3 when pure ice segments start to appear."

5. With these new quality control procedures, we revised the **new Figure 6** (old Figure 7) and the **new Figure 8** (old Figure 9). The main changes can be seen in Figure 8 c, with much higher frequency of ice phase at lower temperatures compared with the old version of Figure 9 c. In addition, we added the new types of analysis showing the frequency distributions of ISR, ice mass fraction (IMF) and IPNF in the top two rows of new Figure 8. For the combined phases 1 to 4 of all in-cloud conditions, the ISR, IMF and IPNF all peak at 0 and 1, with fewer samples in between.

Below we copied Figures R2 and R3, as well as Figures 6 and 8 mentioned above.



Example 1

	A	B	C	D	E
1	RF Num	Time	HHMMSS	T_C	IPNF
10	6	83980	23:19:40	-10.6525	0.233766
11	6	84000	23:20:00	-10.5188	0.210884
12	6	84001	23:20:01	-10.5754	0.233463
13	6	84002	23:20:02	-10.7419	0.214876
14	6	84003	23:20:03	-10.7344	0.215909
15	6	84004	23:20:04	-10.8386	0.309598
16	6	84005	23:20:05	-10.791	0.24937
17	6	84006	23:20:06	-10.893	0.293556
18	6	84007	23:20:07	-10.9377	0.345088
19	6	84010	23:20:10	-10.9769	0.302491
20	6	84011	23:20:11	-10.9608	0.331148
21	6	84023	23:20:23	-11.1361	0.394366
22	6	84032	23:20:32	-11.3052	0.28125
23	6	84035	23:20:35	-11.2345	0.248062
24	6	84151	23:22:31	-11.0003	0.278689
25	6	84152	23:22:32	-10.9734	0.309859

Example 2

	A	B	C	D	E
1	RF Num	Time	HHMMSS	T_C	IPNF
189	5	94884	02:21:24	-1.52149	0.510204
190	5	94888	02:21:28	-1.56155	0.5
191	5	94889	02:21:29	-1.61335	0.657143
192	5	94890	02:21:30	-1.61609	0.757576
193	5	94891	02:21:31	-1.65867	0.75
194	5	94892	02:21:32	-1.63782	0.636364
195	5	94901	02:21:41	-1.54336	0.941176
196	5	94902	02:21:42	-1.62785	0.952381
197	5	94904	02:21:44	-1.57442	0.95122
198	5	94905	02:21:45	-1.49165	0.964286
199	5	94906	02:21:46	-1.36282	0.837838
200	5	94907	02:21:47	-1.33271	0.971831
201	5	94908	02:21:48	-1.37814	0.901961
202	5	94909	02:21:49	-1.38863	0.917808
217	5	94924	02:22:04	-1.61188	0.888889
218	5	94925	02:22:05	-1.65791	0.875
219	5	94926	02:22:06	-1.71671	0.8
220	5	94927	02:22:07	-1.75665	0.869565
221	5	94928	02:22:08	-1.82216	0.880952
222	5	94930	02:22:10	-1.8918	0.913043
223	5	94937	02:22:17	-1.92523	0.545455

Example 3

	A	B	C	D	E
1	RF Num	Time	HHMMSS	T_C	IPNF
50	2	16347	04:32:27	-2.9904	0.910891
51	2	16348	04:32:28	-2.99875	0.857143
52	2	16349	04:32:29	-3.01852	0.896296
53	2	16350	04:32:30	-3.01473	0.854369
54	2	16351	04:32:31	-3.03799	0.795918
55	2	16352	04:32:32	-3.04627	0.864865
56	2	16353	04:32:33	-3.02892	0.850467
57	2	16354	04:32:34	-3.02413	0.91129
58	2	16355	04:32:35	-3.02282	0.859259
59	2	16368	04:32:48	-3.00699	0.853846
60	2	16369	04:32:49	-3.00784	0.880597
61	2	16370	04:32:50	-2.99796	0.792208
62	2	16371	04:32:51	-3.00566	0.774194
63	2	16372	04:32:52	-3.00766	0.781818
64	2	16373	04:32:53	-2.98808	0.75
65	2	16374	04:32:54	-2.952	0.759076
66	2	16375	04:32:55	-2.93198	0.732087

Figure R2. Sample images from the PHIPS probe and the matching time series for the cloud segments with 0.2 ≤ IPNF < 1 in RF2, RF5, and RF6.

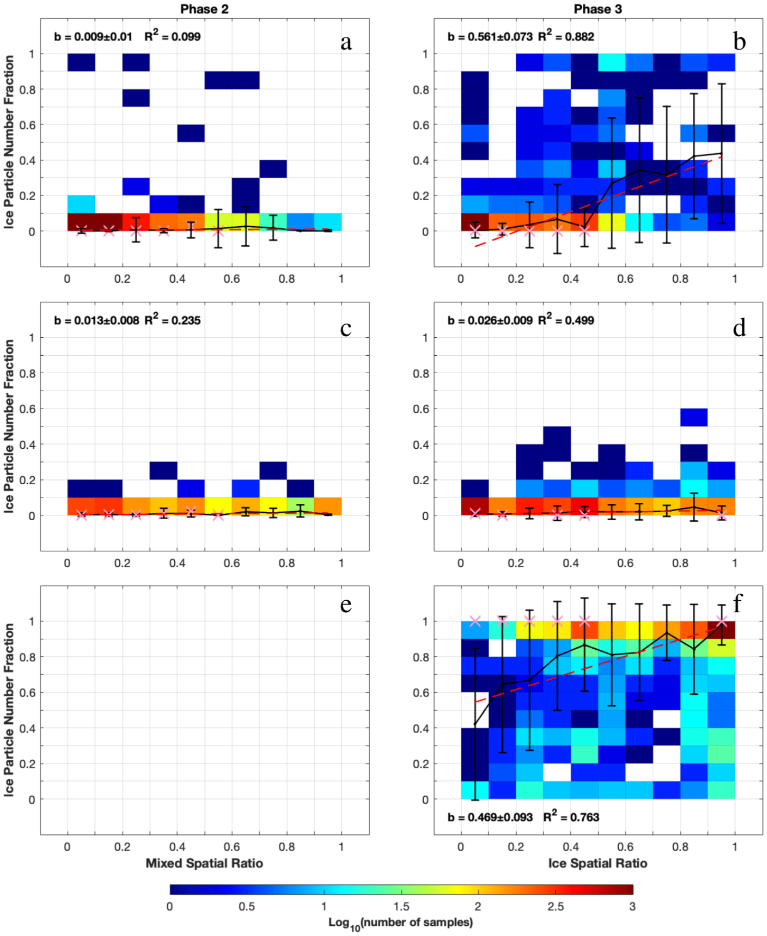


Figure 6. Relationship between ice particle number fraction and mixed spatial ratio or ice spatial ratio, separated by two phases (phase 2 in column 1 and phase 3 in column 2), and by various cloud segments – (a, b) LCR, (c, d) MCR and (e, f) ICR. Average values for each ice spatial ratio bin are shown in black solid lines, with vertical bars representing standard deviations. Linear fit is shown in red dashed line. Average values of generating cells (time series obtained from Wang et al. (2020)) are in pink “X” markers. The slope value b, its associated standard deviation, and the ordinary R-squared value are shown in the legend.

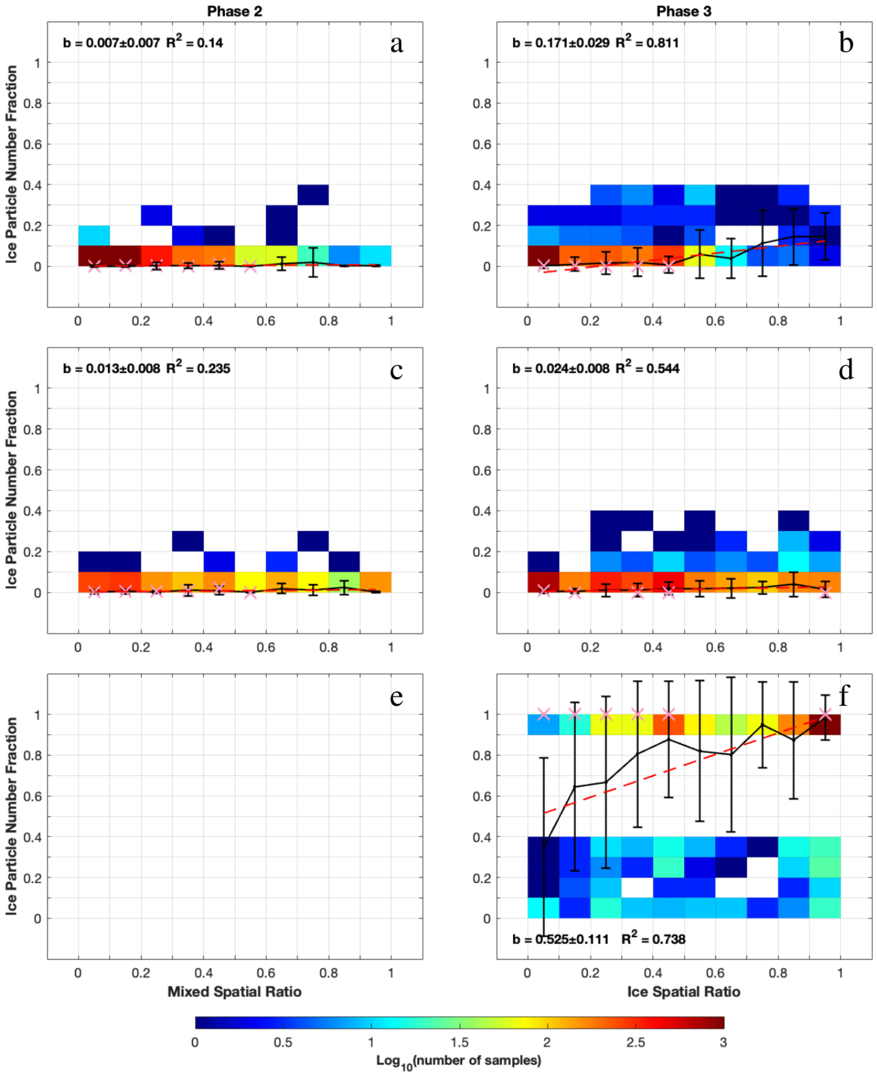


Figure R3. A sensitivity test that is similar to Figure 6, but removing IPNF ≥ 0.4 for LCR and MCR, and 0.4 ≤ IPNF < 1 for ICR. Both phases 2 and 3 show positive slopes for linear regressions, and phase 3 shows higher slope values compared with phase 2, consistent with Figure 6.

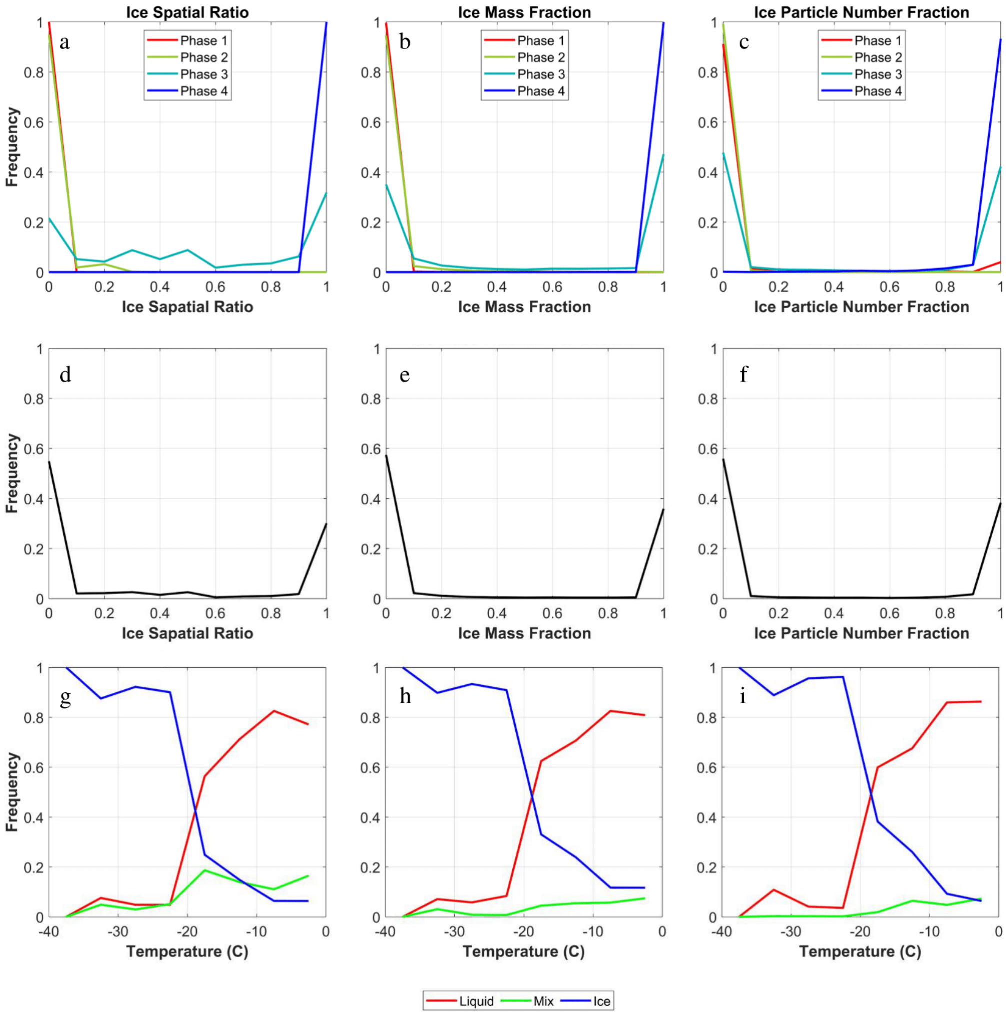


Figure 8. Frequency distributions of (a) ice spatial ratios calculated for individual consecutive TCR, (b) ice mass fraction per second, and (c) ice particle number fraction per second for four phases. (d-f) Similar to (a-c), but for all the phases combined representing the entire in-cloud conditions. (g-i) cloud phase frequency distributions defined based on the respective parameter in each column.

17. The measurements of aerosol particles presented in section 3.5 were conducted by UHSAS inside clouds. I have a serious concern about this approach and the data quality. There is no need to say that aerosol inside clouds is modified by droplet activation, droplet collision- coalescence, scavenging by cloud particles, and precipitating out of the cloud. An example of aerosol processing can be seen in Fig.13b at https://doi.org/10.1175/2011BAMS3180.1. It is also not clear how a 50-second moving average would help eliminate this issue. Such averaging is expected to mix LCR, MCR, ICR, and out-of-cloud regions.

To address the issue of potential contamination of aerosol observations inside clouds, we revised the **new Figure 9** and **new Figure S6** (copied below) by limiting the averaging of aerosol concentrations to clear-sky conditions only. The positive correlations between the average aerosol number concentrations and mixed or ice spatial ratio are still seen in phase 2 and phase 3. In addition, phase 2 shows higher slope values compared with phase 3, which are similar to our conclusions before.

We also edited the discussion in Section 3.5: “Due to the possible complication of in-cloud measurements of aerosol number concentrations, we applied a moving average to calculate logarithmic scales of clear-sky aerosol concentrations at every 50 seconds in Figure 9. Furthermore, the average aerosol concentration is only analyzed if more than half of the entire 50 seconds satisfy the criteria of in-cloud conditions. A coarser spatial averaging using the moving average of clear-sky conditions of every 100 seconds is also shown in supplementary Figure S6.”

All the main findings remain consistent with our previous manuscript, including positive correlations between MSR or ISR with aerosol number concentrations, higher slope values for phase 2 than phase 3, and higher slope values for $N_{>500}$ than $N_{>100}$.

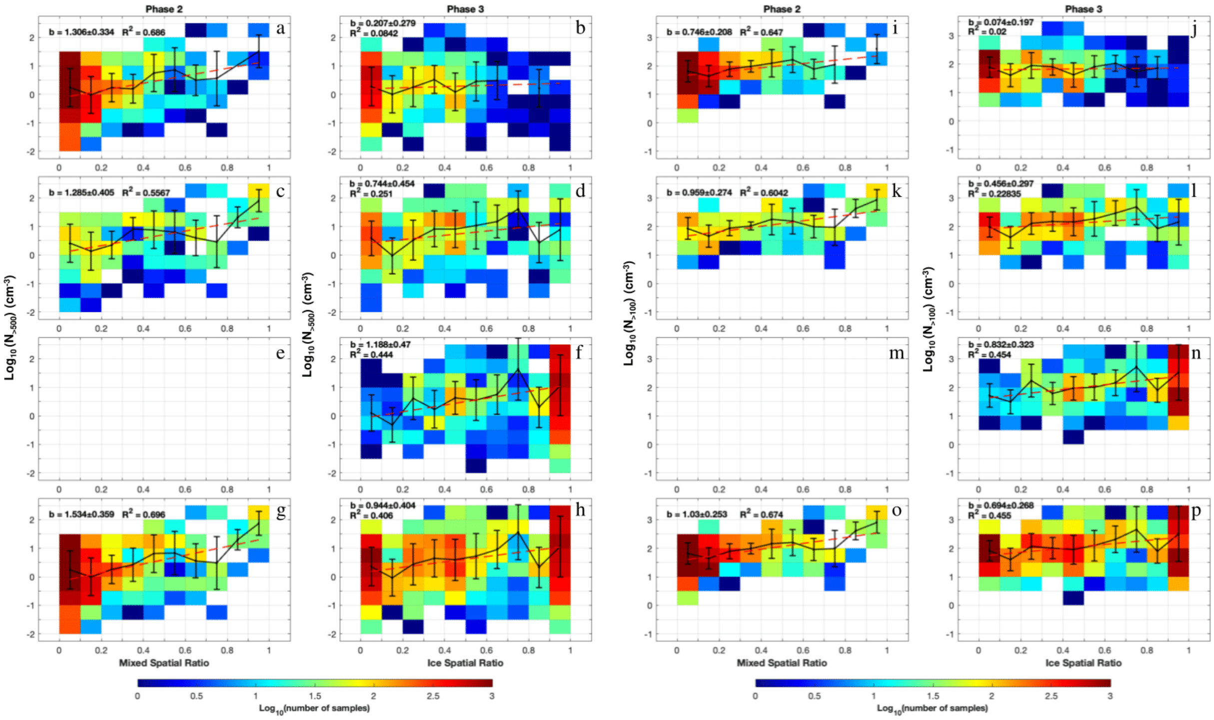


Figure 9. Similar to Figure 6, but showing logarithmic scale (a-h) $N_{>500}$ and (i-p) $N_{>100}$ in relation to mixed spatial ratio or ice spatial ratio, separated by the phases and cloud regions. The first, second, and third rows represent LCR, MCR, and ICR, respectively. The last row represents all cloud regions in a specific phase. The aerosol number concentrations represent the moving average values of every 50 seconds for the clear-sky conditions only.

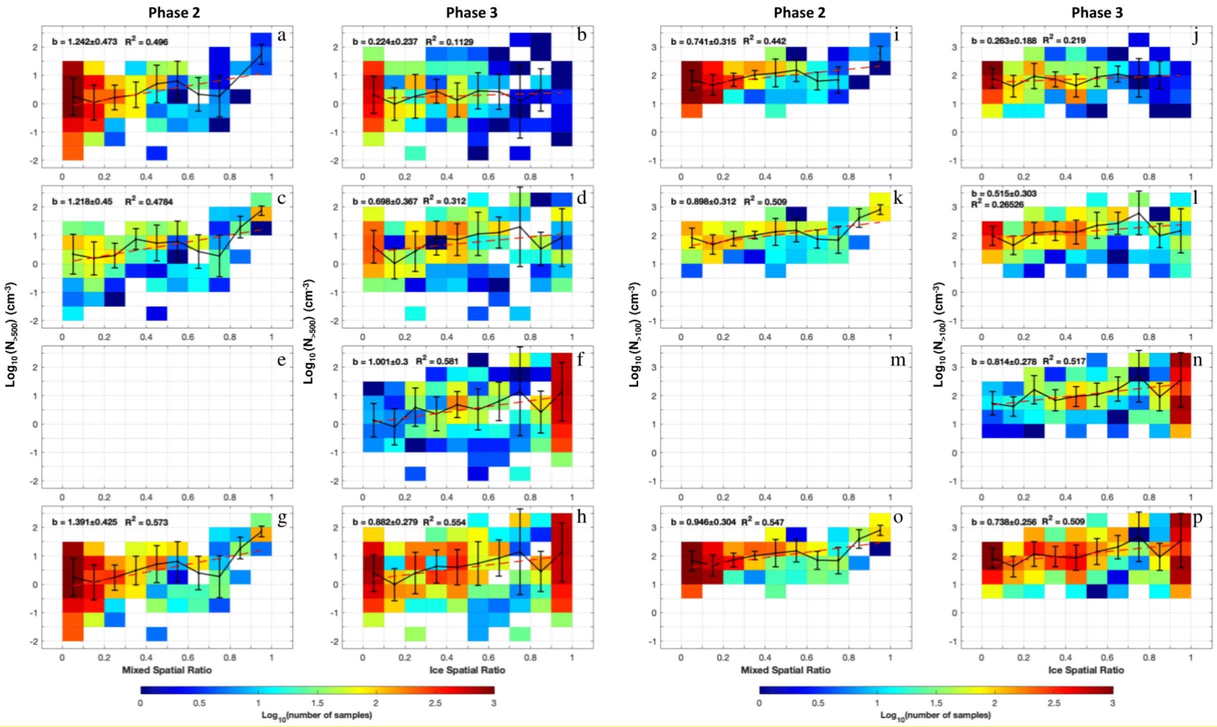


Figure S6. Similar to Figure 9 in the main manuscript but using 100-second moving averages of logarithmic scales of $N_{>500}$ and $N_{>100}$ for the clear-sky conditions only. The results using the coarser scale of aerosol number concentrations are very similar to those shown in Figure 9.

18. Please, replace the reference Korolev et al. JTECH, 1998 by Korolev, A.V., 1998: "About Definition of Liquid, Mixed and Ice Clouds." FAA Workshop on Mixed-Phase and Glaciated Icing Conditions. December 2-3, Atlantic City, NJ, 325-326. This is a result of the error in referencing papers in Korolev et al. AMS Monogr. 2017, which propagated to the present study.

We have replaced the reference Korolev et al. JTEch, 1998 by the following:

Korolev, A.V.: About Definition of Liquid, Mixed and Ice Clouds. FAA Workshop on Mixed-Phase and Glaciated Icing Conditions. December 2-3, Atlantic City, NJ, 325-326, 1998.