Responses to the Reviewers

Format: The reviewer's comments are quoted in italic

Section number in the response refers to the revised manuscript with tracked changes

Quotation in red color stands for revised/added text in the revised manuscript

Overall comment:

We thank the reviewer for the detailed comments. We have addressed individual comments from the reviewer in our response. Some main changes include:

1) Adding a new Figure 2 (which is originally Figure S1 showing the schematic diagram);
2) Adding a supplemental Figure S4, showing a sensitivity test on the impact of length scales;
3) Revising new Figure 5 by combining the original Figure S4 into the original Figure 4;
4) Revising the figure of particle size distributions which becomes the new Figure 6 and new Figure S5 in the revised manuscript;
5) Adding a new Figure S6, showing the linear regressions applied to individual seconds of samples instead of average values inside each bin;
6) Adding clarifications in our revised writing to address most of the remaining minor comments.

Response to comments from the Reviewer

Review: Partition between Supercooled Liquid Droplets and Ice Crystals in Mixed-phase Clouds based on Airborne In-situ Observations

This study analyzes the in-situ observation of mixed-phase clouds collected during the SOCRATES field campaigns over the ocean. Each cloud segment is categorized into four phases: 1) liquid, 2) mixed phase/liquid, 3) mixed-phase/liquid/ice, and 4) ice. The dependency of microphysical cloud properties, dynamical properties, and aerosol properties on each of these phases is examined. The paper introduces mixed and ice spatial ratios to describe the evolution of the phases.

This paper presents an intriguing approach for analyzing in-situ data of mixed-phase clouds. However, there are concerns regarding the equal treatment of cloud segments between 0.2 km and 180 km (see major comment). The quality of the presentation could be enhanced by focusing on fewer figures and discussing these figures more comprehensively (see minor comments).

Recommendation: I suggest reconsidering the paper after making major revisions.

Major:

Cloud segments vary in length from 0.2 to 180 km. What is the likelihood that a short cloud segment is incorrectly classified as liquid (phase 1) due to the low measurement volume missing ice crystals? Is there a possibility that the two edges of a long cloud segment interacted with each other? If not, what justifies treating them as one quantity in the analysis? How do the results depend on the length of the segments? Would splitting long cloud segments into smaller pieces (e.g., 1000 m), where cloud particles interact with each other, be advantageous?

We can see why the previous writing may have caused the confusion. To clarify, our analysis is indeed using every second of data within a consecutive total cloud region (TCR). That is, if a TCR segment contains 10 seconds, then there will be 10 samples at 1-Hz resolution, while a segment containing 100 seconds will produce 100 of 1-Hz samples. We revised the Y axis label for new Figure 4 (below) to distinguish the number of 1-second samples (panel a) from the number of TCR segments (panel c). That is, Figure 4 a shows the number of 1-second samples to be used in all the following analysis (i.e., Figures 5 – 10), while Figure 4 c shows the number of TCR segments associated with various length scales. The count of TCR segments and the distributions TCR lengths are only used in Figure 4 c, not in latter sections.

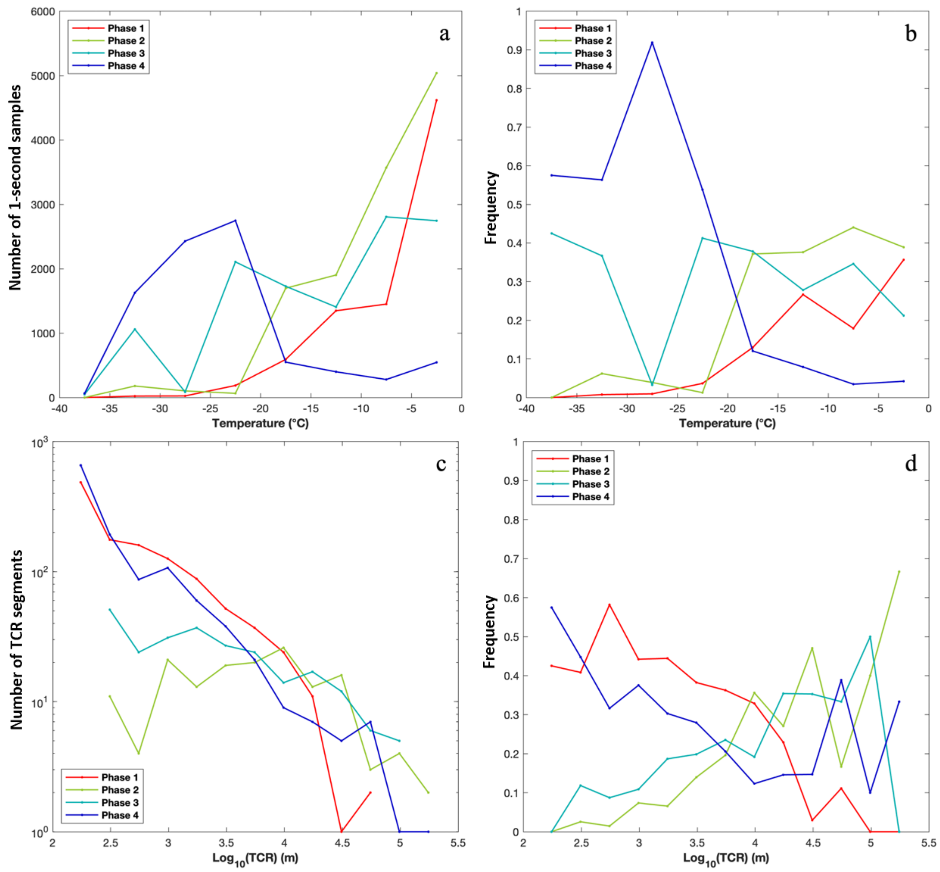

Figure 4. Distributions of 1-Hz samples in four phases at various temperatures in the top row. (a) Number of 1-second samples and (b) frequency of 1-second samples in each phase within various temperature bins. In (b), the frequency of 1-second samples in each phase is normalized by the total number of 1-second samples of all phases in each 5-degree temperature bin. Distributions of various lengths of TCR segments are analysed in the bottom row. (c) Number of TCR segments and (d) frequency of cloud segments in each phase associated with various lengths in log10-scale. In (d), frequency is calculated as the number of segments of a specific phase divided by the total number of segments in each $10^{0.25}$ bin.

To address the comments of "*Is there a possibility that the two edges of a long cloud segment interacted with each other? If not, what justifies treating them as one quantity in the analysis?*" and "*Would splitting long cloud segments into smaller pieces (e.g., 1000 m), where cloud particles interact with each other, be advantageous*", the in-cloud segments (i.e., TCRs) are already separated into 1-second samples when being analyzed for all the analyses shown in revised Figures 5 – 10. To improve the clarity of the text and figure, we revised supplemental Figure S1 and moved it to the main text as the **new Figure 2** (copied below). New descriptions were added in Section 3.1: "An illustration of the identification of TCR is shown in Figure 2. In that example, 1 second of LCR, 2 seconds of MCR, and 4 seconds of ICR are adjacent to each other. Then the 1 LCR sample, 2 MCR samples, and 4 ICR samples all belong to the same TCR, which produces a total of 7 seconds of samples. All the 1-Hz samples within the TCR will be used in the analysis in Sections 3.3 – 3.8 (i.e., Figure 4 a and b, Figures 5 – 10)."

In addition, we tried to highlight the differences between the definition of LCR, MCR and ICR and the definition of TCR in Section 3.1: "In the first step, each second of observations are categorized into four conditions, including a second of clear-sky condition, liquid cloud region (LCR), ice cloud region (ICR), or mixed-phase cloud region (MCR). LCR is defined as a one-second sample where only supercooled liquid droplets were observed, while ICR is defined as a one-second sample with only ice crystals. MCR is a one-second sample with occurrence of both ice and liquid. In the second step, a total cloud region (TCR) that can potentially contain multiple seconds with a combination of LCR, ICR and MCR is identified, which basically is a consecutive in-cloud segment surrounded by clear-sky conditions. In other words, LCR, ICR and MCR are defined at the scale of each second, while TCR is defined at the scale of a consecutive in-cloud segment which can contain more than one second."

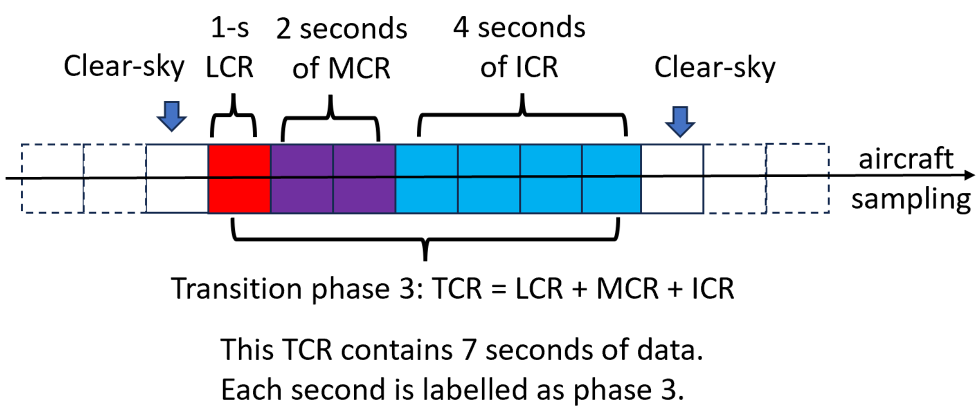

Figure 2. A schematic diagram that illustrates the identification of a total cloud region (TCR) sample, with 1 second of LCR (red), 2 seconds of MCR (purple), and 4 seconds of ICR (blue) embedded inside this TCR. All 7 seconds of samples inside this TCR are used in the following analysis of cloud properties.

To address the comment of "*What is the likelihood that a short cloud segment is incorrectly classified as liquid (phase 1) due to the low measurement volume missing ice crystals?*", we plotted the scatterplots of ice water content (IWC) versus liquid water content (LWC), as well as ice crystal number concentration (Nice) versus supercooled liquid droplet number concentration (Nliq), for the conditions when both ice and liquid are observed (shown below as **Figure R1**). We added this discussion in Section 3.1: "To investigate the possibility of misclassifying MCR as LCR due to the relatively lower number concentrations of ice particles compared with supercooled liquid droplets in a one-second sampling volume, distributions of mass and number concentrations of ice crystals are examined against those of supercooled liquid droplets (not shown). When liquid and ice coexist, the majority of the 1-second samples have both IWC > 0.01 g m^{-3} and LWC > 0.01 g m^{-3}. In addition, the mass concentrations and number concentrations of ice and liquid are positively correlated with each other. This indicates that when ice and liquid coexist, most likely both types of hydrometeors have significant mass and number concentrations. Thus, it is less likely that the smaller sampling volume for ice crystals would lead to a misclassification of MCR as LCR. It is possible though, that some pure ICR pockets with very low number concentrations of ice crystals may be missing."

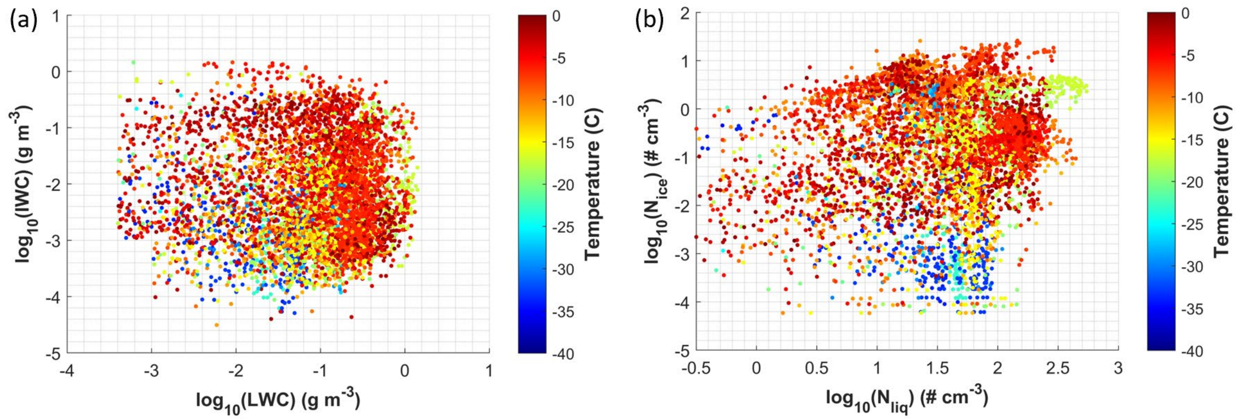

Figure R1. Distributions of (a) IWC with respect to IWC, and (b) Nice with respect to Nliq, all in log10-scale, plotted for the samples with coexistence of ice crystals and supercooled liquid droplets. The scatterplot dots are color coded by temperature.

To address the comment of "*How do the results depend on the length of the segments?*", we plotted the distributions of four phases similar to the revised Figure 4, but restricted to different length scales of TCR (**new supplemental Figure S3**, shown below). We added these comments in the text in Section 3.3: "The impact of length scales of TCR on the phase distributions is examined in supplemental Figure S3. TCR samples are separated into four scales – 0.1 – km, 1 – 10 km, 10 – 100 km, and > 100 km. The dependence on temperature for the distributions of four phases is consistently seen for various scales, e.g., phase 1 has more samples at higher temperatures, while phase 4 has more samples at lower temperatures. Comparing the shorter (Figure S3 a and b) and longer (c and d) TCR samples, the shorter ones have more samples in phase 1 (i.e., pure liquid phase), while the longer ones have more phases 2 and 3. This result indicates that the coexistence of ice and liquid occurs more frequently in clouds with larger spatial extent, such as stratocumulus and stratus clouds."

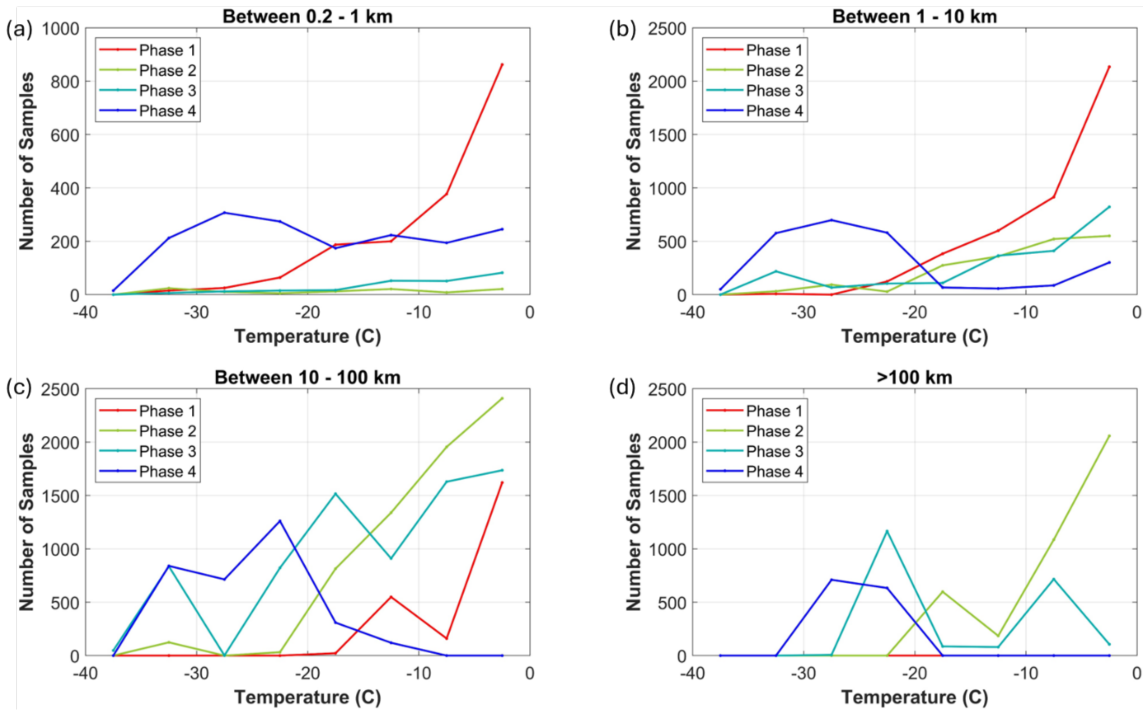

Figure S3. Similar to Figure 4 a, distributions of 1-Hz samples in four phases at various temperatures, separated by the length scales of TCR samples. E ach second within the TCR is counted as a sample.

Minor:

Abstract: Specify the dataset used and the types of clouds investigated.

We added more information to the abstract: "Using this method, we examine the relationship between the macrophysical and microphysical properties of Southern Ocean mixed-phase clouds at -40 to 0°C (e.g., stratiform and cumuliform clouds) based on the in-situ aircraft-based observations during the US National Science Foundation Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study (SOCRATES) flight campaign."

Line 180: Figure S1 is crucial for understanding the approach and should be moved to the main manuscript.

We revised this figure, which becomes the new Figure 2 as copied in our response above.

Line 184: The introduction of M1, M2, M3 is confusing and unnecessary as it only appears in Table 1.

We removed the terms of "M1, M2 and M3" and revised the text: "Within each TCR, the spatial ratios of LCR, MCR, and ICR relative to TCR are calculated. The definitions of each phase are based on these spatial ratios as described in Table 1. The number of one-second samples and the number of cloud segments for four phases are summarized."

The revised Table 1 is copied below.

Table 1. Definitions of four phases of mixed-phase clouds based on ratios of lengths of LCR, MCR, and ICR over the length of TCR within a consecutive cloud segment, i.e., $\frac{L_{LCR}}{L_{TCR}}$, $\frac{L_{MCR}}{L_{TCR}}$, and $\frac{L_{ICR}}{L_{TCR}}$, respectively.

Phase	Description	Number of 1-second samples	Number of TCR segments	Spatial Ratio of LCR	Spatial Ratio of MCR	Spatial Ratio of ICR
1	Only LCR	8243	1163	$\frac{L_{LCR}}{L_{TCR}} = 1$	$\frac{L_{MCR}}{L_{TCR}} = 0$	$\frac{L_{ICR}}{L_{TCR}} = 0$
2	MCR appears	12557 (LCR: 11096, MCR: 1461)	142	$0 \leq \frac{L_{LCR}}{L_{TCR}} < 1$	$0 < \frac{L_{MCR}}{L_{TCR}} \leq 1$	$\frac{L_{ICR}}{L_{TCR}} = 0$
3	Pure ICR must appear	11988 (LCR: 3478, MCR: 2973, ICR: 5537)	249	$0 \leq \frac{L_{LCR}}{L_{TCR}} < 1$	$0 \leq \frac{L_{MCR}}{L_{TCR}} < 1$	$0 < \frac{L_{ICR}}{L_{TCR}} < 1$
4	Only ICR	8646	1193	$\frac{L_{LCR}}{L_{TCR}} = 0$	$\frac{L_{MCR}}{L_{TCR}} = 0$	$\frac{L_{ICR}}{L_{TCR}} = 1$

Line 215: It should be a second-by-second analysis. Could an analysis of larger intervals (e.g., 10 seconds) provide a better understanding of the cloud phases?

We can see why the previous writing led to a misunderstanding. We indeed are using every second within a TCR as individual samples. We revised that sentence to clarify the difference between this work and previous studies: "Nevertheless, this method provides a statistical separation of the cloud phases and allows a more focused analysis of the coexistence of supercooled liquid water and ice crystals that cannot be achieved if a one-second sample is analyzed without the context of its surrounding conditions, for instance, if a one-second LCR is part of a pure liquid cloud segment, or is surround by MCR or ICR." Regarding the comment about intervals, we found that the longer TCR segments are generally associated with larger gaps between cloud segments, so the analysis restricting to larger gaps lead to a similar result as restricting to larger cloud segments, as discussed in Figure S3.

Line 225: "The lengths of cloud segments vary..."

We revised it to "The lengths of TCR segments vary…"

Line 225: After the sampling statistic, I expected the number of samples, not a time.

We thank the reviewer for catching the typo. That sentence has been revised: "The lengths of TCR segments 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 60 km…"

Line 236 – 238: A mixed-phase cloud (MPC) consists of supercooled droplets and ice crystals.

We revised this sentence: "For macrophysical properties of mixed-phase clouds, we focus on investigating the lengths of cloud segments and the spatial fraction of a cloud segment containing ice, which is defined as mixed spatial ratio and ice spatial ratio."

Which spatial fraction describes the macrophysical properties of MPCs? How do LCR, ICR, MCR, and TCR represent macrophysical properties, and why aren't they used in the analysis?

The spatial fraction containing ice for phase 2 is defined as mixed spatial ratio. The spatial fraction containing ice for phase 3 is defined as ice spatial ratio. These two spatial ratios are analyzed in Figures 7 – 10 in the revised manuscript. The length of cloud segments is analyzed in Figure 4 panels c and d of the revised manuscript.

Line 240 – 245: Is it correct that the mixed spatial ratio equals the spatial ratio of MCR (M3), but the ice spatial ratio differs from the spatial ratio of ICR? If yes, try to find clearer names and perhaps add the definition of mixed spatial ratio and ice spatial ratio to Table 1.

Yes, the reviewer is correct that "*mixed spatial ratio equals the spatial ratio of MCR (M3), but the ice spatial ratio differs from the spatial ratio of ICR*", because phase 3 may contain both MCR and ICR. Because the original terms of M1, M2 and M3 caused confusion, we removed their name, and directly refer to them as length of LCR, MCR and ICR relative to the length of TCR (as described in Table 1 caption). In addition, the discussions of mixed spatial ratio and ice spatial ratios do not appear until we analyze them in Figure 7 of the revised manuscript, thus we moved the definitions of these two terms into Section 3.6, right before we introduced Figure 7.

Line 270 – 272: What is the percentage of observations over 1.25 m/s in phase 2 and 3? Is the difference significant? I suggest moving Figure 4 i-p to the appendix and Figure S4 b-d to the main manuscript.

We revised the new Figure 5, and moved the original Figure 4 i – p to be the new supplemental Figure S4.

The percentage of σ_w values of one-second samples greater than 1.25 m/s in phases 2 and 3 are very small, which is 0.412% and 0.660% of the total samples of each phase, respectively. In addition, we added a new supplemental Table S2 to illustrate that phases 2 and 3 have higher frequencies of larger σ_w values than phases 1 and 4.

Table S2. Number of one-second samples of σ_w (i.e., standard deviation of vertical velocity) in four phases at various ranges. Percentages relative to the total number of σ_w samples of each phase are shown in parentheses.

Phase number	**All σ_w values**	**$\sigma_w \geq 0.5$ m/s**	**$\sigma_w \geq 1$ m/s**	**$\sigma_w \geq 1.25$ m/s**
Phase 1	4549	621 (13.7%)	15 (0.330%)	0
Phase 2	8730	1360 (15.6%)	174 (1.99%)	36 (0.41%)
Phase 3	8638	1491 (17.3%)	251 (2.91%)	57 (0.66%)
Phase 4	7814	176 (2.25%)	0	0

We revised the text in Section 3.4 describing the new Figure 5, Figure S4 and Table S2: "Probability density functions (PDFs) of RH_i, RH_{liq}, vertical velocity, and σ_w are further examined in Figure 5 i – l. The peak frequencies of RH_{liq} are seen at liquid saturation for phases 1 – 3, consistent with the findings in Figure 5 a – d. The PDFs of vertical velocity show higher frequencies of updrafts for phases 2 and 3 compared with phases 1 and 4. In addition, PDFs of σ_w show higher frequencies of large σ_w values in phases 2 and 3 than phases 1 and 4. The number of 1-Hz σ_w samples at various ranges (i.e., ≥ 0.5 m/s, ≥ 1 m/s, and ≥ 1.25 m/s) and their percentages relative to the total samples in each phase are shown in supplemental Table S2. That analysis also shows higher percentages of larger σ_w values in phases 2 and 3 compared with phases 1 and 4. Similarly, the distributions of σ_w as a function of temperature in supplemental Figure S4 show more samples above 1 m/s across a wide range of temperatures from -36°C to 0°C in phases 2 and 3 than phases 1 and 4."

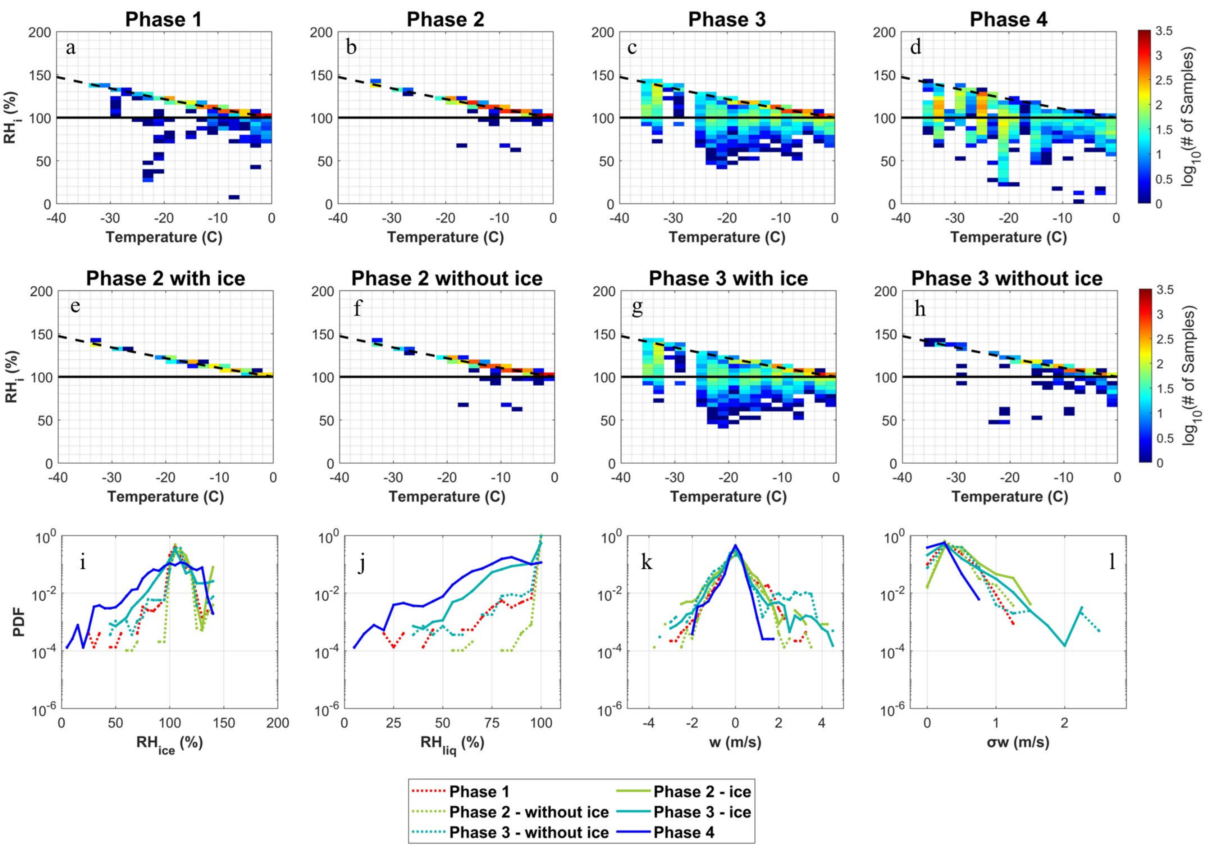

Figure 5. (a-h) Distributions of RH_i as a function of temperature. The PDFs of (i) RH_i, (j) RH_{liq}, (k) vertical velocity (w) and (l) σ_w of various phases. Dashed lines in (a) – (h) indicate liquid saturation.

Line 295 – 296: Which phase are you referring to? I suggest plotting all temperatures of the size distribution of this phase in one plot to emphasize the differences. As differences in temperature are not further discussed, I suggest moving the size distribution of the temperature interval to the appendix and showing only the size distribution of all temperatures in the main manuscript.

We revised this figure. The new Figure 6 now shows the entire temperature range between -40 and 0°C, while the separated temperature ranges are shown as the new supplemental Figure S5.

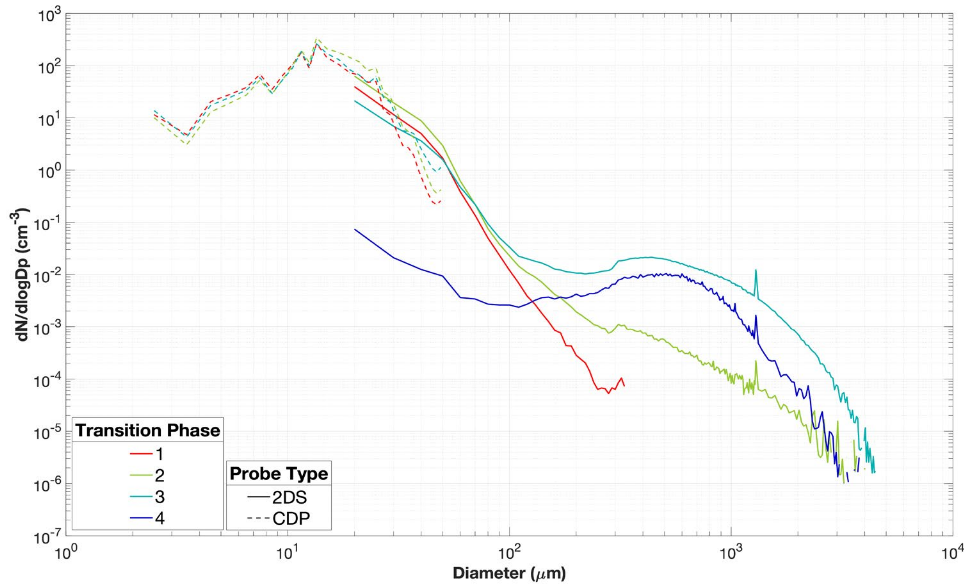

Figure 6. Particle size distribution of the four phases for mixed-phase clouds separated by probe types. The entire dataset at the temperature range of -40°C to 0°C is shown. Phase 4 only shows 2DS measurements because ice particles measured by CDP are excluded from the analysis.

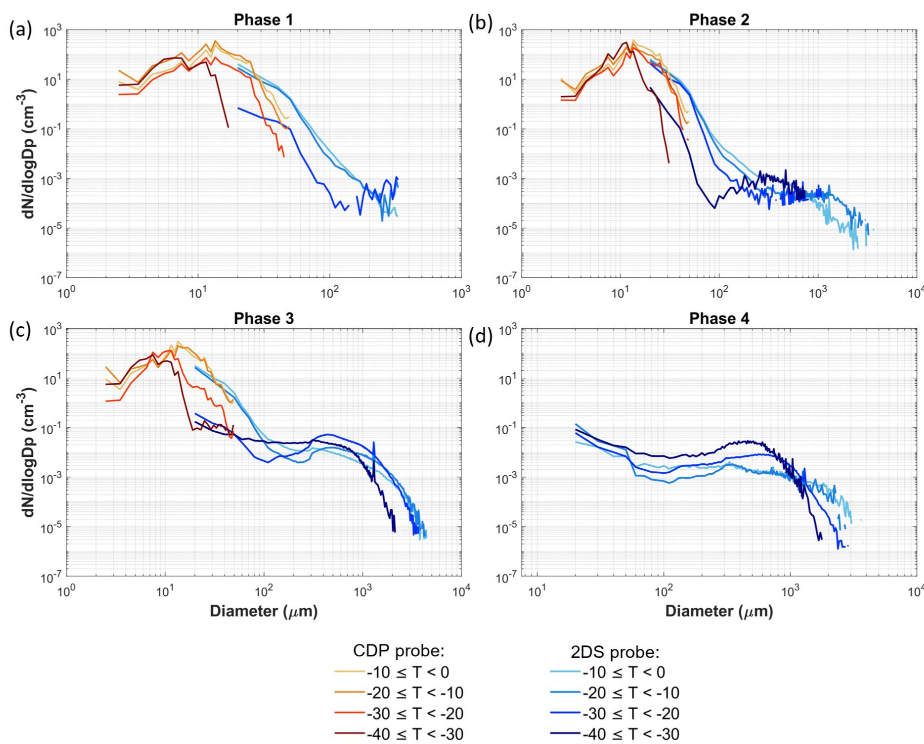

Figure S5. Particle size distribution separated by four phases and various temperature ranges. Four temperature bins between -40°C to 0°C are shown in each panel. Phase 4 only shows 2DS measurements because ice particles measured by CDP are excluded from the analysis.

Line 305 – 306: What is the fraction of observations with ice particle number fraction > 0.1 in Figure 6b? Why was the linear regression calculated on the mean of each ice spatial ratio bin (which weighted each bin equally despite very different numbers of observations in each bin) and not based on individual observations? Please add more information on the calculation of the linear regression.

The original Figure 6 b is now Figure 7 b in the revised manuscript. We added this in the discussion in Section 3.6: "After the corrections, out of 2866 seconds of samples analyzed in Figure 7 b, 172 seconds (i.e., 6.00%) show IPNF > 0.1."

We clarified the reason of conducting linear regression for the mean microphysical properties, and also added a new supplemental Figure S6 to illustrate the differences if the linear regressions are applied directly to individual seconds of samples. The discussion is added to Section 3.6: "The linear regression analysis is applied to the average values of microphysical properties in each spatial ratio bin, in order to assign an equal weight to each bin of mixed or ice spatial ratio. When directly applying the linear regressions analysis to individual seconds of IPNF (as shown in supplemental Figure S6), similar slope values are seen compared with Figure 7, but the bins of mixed spatial ratio and ice spatial ratio have uneven distributions of samples."

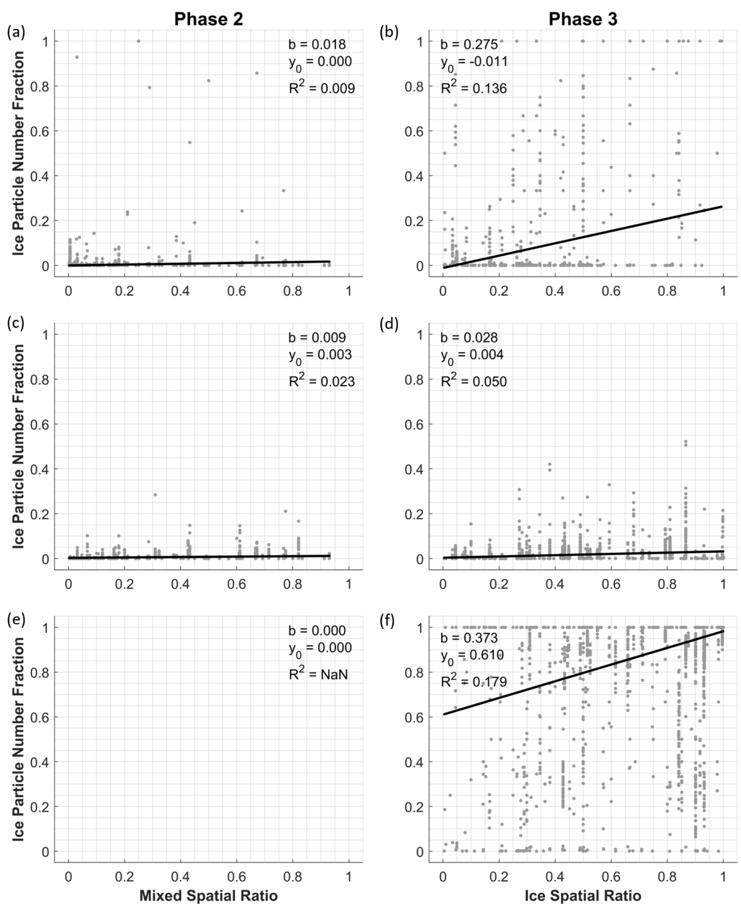

Figure S6. Similar to Figure 7, but applying the linear regressions directly to individual seconds of samples.

Line 315 – 320: I have difficulty following the argument. Are you referring to "ice crystals gradually dominating the total particle population" as high ice particle number fraction? How can "a particular TCR" be identified in the multiple subfigures? What is the spatial extent of the entire cloud segment?

We revised this sentence to clarify our point: "This means that while ice crystals gradually dominate the total particle population (i.e., IPNF increases) in cloud segments, the spatial fraction containing ice particles (i.e., MCR+ICR) also approaches 1 from a macroscopic perspective."

Line 323 – 324: How can "ICR appear" when phase 3 always has some ICR?

We deleted that phrase in the revised sentence: "On the other hand, in phase 3, ice crystals start to become the dominant particles by number concentration ~~when ICR appears,~~ and supercooled liquid droplets become less dominant."

Line 325: Please describe in more detail how you derived that "they experience similar rates of phase changes from liquid to ice" based on measurements of individual states of cloud microphysics?

We can see that the original comment is a little far reaching. Thus we deleted that discussion about the rate of phase change.

Line 336: How do you conclude that generating cells contain lower ice particle number fractions? What is the uncertainty of the generating cells measurements?

We added the explanation in the text in Section 3.6: "Previously, Wang et al. (2020) used airborne remote sensing measurements from the SOCRATES campaign to identify generating cells of ice crystals. Based on the definition from American Meteorological Society (2013), generating cells are defined as cloud-top regions with high radar reflectivity, which often produce fall streaks of falling hydrometeors. Out of the 16 cases of generating cells detected by Wang et al. (2020), all 16 cases contain supercooled liquid droplets. The average LWC and Nliq inside generating cells were found to be greater than those outside the generating cells. In addition, larger ice particles and higher Nice were seen in the generating cells, associated with the updrafts inside the cells. These reported generating cells are also analyzed in Figure 7, with the average IPNF values shown in each mixed and ice spatial ratio bin. The generating cells associated with LCR and MCR contain lower IPNF (Figure 7 a – d). This is because when generating cells are associated with high concentrations of supercooled liquid droplets, Nice may be lower than Nliq, which leads to the lower IPNF. But when the generating cells are associated with ICR, significantly higher IPNF (close to 1) are seen for most ice spatial ratio bins (Figure 7 f). This result suggests that not all regions within the generating cells experience significant phase change from liquid to ice, unless the ice-containing regions become dominated by ice."

Line 348-349: What do you mean by "... similar rate of increase between ice crystals embedded among supercooled liquid droplets..."?

We can see that the original discussion is quite confusing. We deleted that comment and focused on comparing the slope values in various phases.

Line 352: Should be Figure 7i.

Thanks for catching the typo. Yes, it should be Figure 7 i.

Line 357 – 358: What effect would the formation and growth of ice particles have, and could the depletion of the liquid phase also play a significant role?

We can see that the original comment about "formation and growth of ice particles" is unnecessary and causes confusion, so we deleted that part of the sentence. The revised sentence is: "As ice crystals grow into pure ice segments (i.e., ICR), liquid phase starts to rapidly evolve into ice phase~~, suggesting that the formation and growth of ice particles become more significant when pure ice segments appear~~."

Line 405: Why should SIP be stronger in phase 3 when phase 2 has more large droplets (according to Fig. 5)?

Secondary ice production (SIP) is generally identified when Nice is much higher than number concentrations of ice nucleating particles. In other words, the very high Nice values are usually associated with SIP. Even though phase 2 has higher Nliq than phase 3, phase 3 also has higher Nice than phase 2 (as shown in the particle size distribution in revised Figure 6). Thus, it is more likely that phase 3 contains more SIP events than phase 2.

Line 447-448: Please be precise if you are referring to MCR/ICR or phase 2/3.

We can see why the original comment is confusing. We revised it to: "… the method presented in this work allows one to compare the cloud segments when ice crystals are surrounded by supercooled liquid water in MCR with those when pure ICR starts to appear."

Line 468- 476: Move this paragraph to the definition of the phases.

Thanks for the suggestion. We moved the paragraph to the end of Section 3.1, after defining the four phases.

Line 480 – 484: Are you suggesting that once a small pocket of pure ice crystals appears in a cloud (phase 3), the rate of change from liquid to the ice phase accelerates for the whole cloud?

We revised the comment indicating a causal relationship to a correlation: "This study illustrates that the rates of phase change are also correlated with the existence of pure ice segments (Figures 7 and 8), not only with the mixed spatial ratio or ice spatial ratio which reflects the spatial fraction of ice-containing regions. Future model parameterization is recommended to quantify the varying rates of phase change throughout a cloud's lifetime by considering two main factors – the type of phases (especially phase 2 versus phase 3) and the spatial fraction of ice-containing region."