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.

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.

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”.

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?

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.

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.

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.

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.

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.

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?

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.

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.

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.

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.

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%, $dM_{ice}/dt=0$.

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 $\lambda_{ice}\cong0$, whereas in ice clouds $\lambda_{ice}\cong1$. 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, $\lambda_{ice}\cong0$. 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 $\lambda_{ice}\cong0$, whereas in ICR $\lambda_{ice}\cong1$. For the sake of argument, consider a case with $\lambda_{ice}\cong0.4$. 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 $N_{ice} = \lambda_{ice} N_{liq} /(1- \lambda_{ice}) \cong 67\ cm^{-3}$. 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 $N_{ice} = 100L^{-1}$ (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).

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.

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.

Concluding remarks

As in my previous comment, I did not consider any minor questions since they overlapped with the major issues of this work. My biggest concern remains the data quality of this paper.

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