**Review of "The Transition from Supercooled Liquid Water to Ice Crystals in Mixed-phase Clouds based on Airborne In-situ Observations" by F.V. Maciel and M. Diao et al. (2022)**

**Overview**

This study is focused on the microphysical characterization of mixed-phase environments and on linking it to the various stages of phase transition. The data explored here was collected in-situ during the SOCRATES field campaigns over the Southern Ocean. Identification of the phase transition stage is based on the assessment of the presence ice, liquid and mixed-phase cloud segments coexisting in the same cloud. Depending on the combination of these three thermodynamic states, the clouds were separated into four categories: (1) liquid, (2) mixed-phase ⋀ liquid; (3) mixed-phase ⋀ (liquid ⋁ ice ⋁ (liquid ⋀ ice)); (4) ice. Aerosol concentration, in-cloud dynamics and atmospheric state conditions were quantified for each of these four categories. The applied method enabled conclusions regarding the effect of aerosols, atmospheric state and cloud dynamics parameters of the evolution of mixed-phase clouds. This paper is interesting and deserves attention, however, I have concerns about the general approach, data quality and clarity of presentation. I would also recommend a more thorough acknowledgement of past studies on mixed-phase clouds.

**Recommendation**: I regret to say that, in my opinion, the paper is not suitable for publication in ACP in its present form. I would recommend rewriting the manuscript addressing the comments below and resubmitting the paper.

**Major comments**

*Methodology and basic assumptions*

1. The proposed method is based on a preconception that, during their lifetime, mixed-phase clouds pass through the stages (1)=>(2)=>(3)=>(4) as described in the paper, i.e., the cloud is initiated as liquid under supercooled conditions; then it experiences nucleation of ice and turns into mixed-phase; after that some section of the mixed-phase cloud glaciates and turns into ice, and in the final stage, the entire cloud is glaciated. The conceptual diagram of this process is shown in Fig.2, and it is used as the basis for the following interpretation of the data. This kind of "classical" evolution of mixed-phase clouds was observed and documented over 35 years ago (e.g., Hobbs and Rangno, 1985). However, besides the classical progression of mixed-phase, there are two other routes of evolution of mixed-phase clouds. The first scenario is when, after nucleation of INPs and turning liquid cloud into mixed-phase, all ice particles precipitate out of the cloud, turning the mixed-phase back into liquid. In other words, the thermodynamic phase evolution of such cloud can be described by the diagram: liquid => mixed-phase => liquid (i.e. (1)=>(2)=>(1)). The imbalance between the water vapor supply and the bulk ice mass crystal growth, required for the maintentenance of mixed-phase clouds, was discussed in Rauber and Tokay (1991), Pinto (1998). An interesting aspect of maintenance of supercooled liquid clouds was discussed by Westbrook and Illingworth (2011). There is a fair amount of modelling attempts to find an explanation of maintenance of mixed-phase clouds through the balance of INPs and dynamic forcing (e.g., Avramov, A., et al. 2011; Fan et al. 2009, 2011; Smith et al., 2009; to name a few). The second mixed-phase evolution scenario is related

to the generation of mixed-phase clouds in a pre-existing ice cloud due to dynamic forcing, which can presented by a diagram ice=>mixed-phase (i.e. (4)=>(2)). Note, that Fig.2 considers stage (4) as a final stage, whereas in the second scenario, (4) is an initial stage. The theoretical basis explaining such process was developed in Korolev and Mazin, 2003; Korolev and Field, 2008, Field et al. 2014; Hill et al, 2014). These studies were supported by earlier observations of mixed-phase clouds embedded in pre-existing, deep ice clouds (e.g., Hogan et al., 2002; Field et al. 2004). To summarize the above, the direction of the evolution of a mixed-phase environment may differ from the classical consideration (as in Fig.2), which was assumed in this work. Since the present study does not contain evidence justifying the classical evolution ( (1)=>(2)=>(3)=>(4) ) of the sampled mixed-phase clouds, it would be relevant to rewrite the sections of the paper discussing the “transition phases” and make a disclaimer of two other scenarios of the mixed-phase evolution.

2. As follows from the explanation in section 3.1, this study considers two types of mixed phase clouds as “genuine” where ice particles and liquid droplets are spatially mixed on a small scale, and “conditionally” mixed clouds, where ice and liquid are spatially separated (see Korolev et al. 2017 (Fig.5-1); Korolev and Milbrandt, 2022 (Fig.1)). In conditionally mixed-phase clouds the WBF process is disabled due to spatial separation of ice and liquid. Thus, the clouds identified in this study as (3), may form a sequence of spatially adjacent cloud segments …-ice-liquid-ice-liquid-… Such clouds are thermodynamically stable, and their lifetime will be determined by processes other than the interaction between ice and liquid (e.g., WBF, riming). Therefore, the term “transition phase” is not directly applicable to the “conditionally” mixed clouds considered in this paper, and it is relevant only to “genuinely” mixed-phase clouds. On the same note, in the frame of classical consideration of the mixed-phase evolution, the ice stage (4) is stable; (in terms mixed-phase transformation, other types of instabilities are not considered). Therefore, the term “transition phase” should not be applied to stage (4). Having said that, the term “transition phase” should be reconsidered in this study and used cautiously and applied only to “genuinely” mixed clouds.

3. Since the direction of the evolution of mixed-phase environment may go backward, in contrast to the classical evolution, it makes sense to consider the microphysical properties of cloud thermodynamic states (1-4) without connection to the evolution of mixed-phase or to do so cautiously. This may also involve changing of the title of the paper.

*Data quality*

I have some concerns regarding the results of the particle size distributions (PSD), humidity and vertical wind measurements presented in this paper. The details are described below.

4. Measurements of DSD in liquid clouds (red lines):
   (a) The DSDs, measured by the 2DC in liquid clouds in all temperature subranges (including $-30<T<-20$C and $-40<T<-30$C), extend up to 3mm in diameter. These are exceptionally large raindrops for stratiform clouds at temperatures below -20C. To my best knowledge, I have never seen reports of observation of 2-3mm supercooled raindrops at $-40<T<-20$C.
   (b) As follows from Fig. 6, the concentration of supercooled drops with D>200um measured by 2DC in liquid clouds (red lines) appears to be higher than the

concentration of ice particles measured in mixed-phase clouds (light green lines) by 2DS. At temperatures -40<T<-20C (Figs.6c,d) the concentration of drops D>~2mm measured by 2DC is higher than the concentration of ice particles in ice clouds. Such behaviour appears to be anomalous.

(c) $D_{max}$ measured by 2DS and 2DC are expected to be approximately close to each other. This statement is well satisfied for PSDs in cloud types (2), (3) and (4) in Figs. 5 & 6. However, in liquid clouds (type (1)) there is a well pronounced difference between $D_{max}$ measured by the 2DS (~300um) and that measured by the 2DC (~3mm).

Items (a)-(c) are indicative that the SLD measurements by 2DC in liquid clouds are compromised.

5. The 2DS DSD in Fig.6a,c,d in the temperature subranges -10<T<-0C, -30<T<-20C and -40<T<-30C appear to be nearly the same. All three DSDs have the same $D_{max}$=~300um. Based on the past in-situ observations the concentration of SLD and $D_{max}$ is expected to decrease with the decrease of temperature due to an increasing of the probability of droplet freezing with the decrease of T and increase of their D. Both the absence of the temperature dependence of 2DS DSDs and observations of SLDs with D~200-300um below -30C are highly questionable.
6. The particles counted by CDP in ice cloud (type (4)) are most likely artifacts related to counting ice (e.g. Korolev et al. 2013), and therefore, their contribution to ice should be excluded.
7. The diagrams in Fig.7 show observations of LWC in liquid clouds as low as $10^{-6}$ g/m$^3$ (b), and IWC in ice clouds as low as $10^{-5.5}$ g/m$^3$ (h). Such low LWC and IWC values are below the minimum threshold, which can be measured from aircraft at 1s-averaging time by the particle probes employed in this study (e.g. Baumgardner et al. 2017).
8. Both theoretical and observational studies (Korolev and Mazin, 2003; Korolev and Isaac, 2006) showed $RH_{liq}$ in mixed phase clouds is close to 100%. Due to the short time of phase relaxation (typically 0.1-10s) in liquid and mixed-phase clouds, the evaporating droplets will rapidly bring the system of "droplets-water vapor" to quasi-equilibrium and saturate the environment. In this regard, the observations at -25C in liquid and mixed-phase clouds (with no ice) of $RH_{liq}$ ~88%, 82% and 75%, respectively (Fig.10b), is suggestive of large biases in $RH_{liq}$ measurements. The low accuracy of $RH_{liq}$ does not allow for the conclusions made in the paper about the relationships between humidity and microphysical parameters of cloud type (1)-(4).
9. Numerous in-situ observations (including those, cited in the present study e.g., Wang et al. 2020) showed that in stratiform clouds the distribution of the vertical wind is centered close to zero. A visual assessment of the diagram in Fig.10c suggests systematic biases of the vertical wind with an average speed of ~-0.2m/s or lower. For mixed-phase clouds (type 2) at -25C the biases in vertical wind reached -0.5m/s and ~-0.9m/s. Clouds subsiding at such speed are expected to evaporate within a relatively short time due to adiabatic heating. Thus, for LWC(0)=0.1g/m$^3$ at -10C, a cloud parcel descending at 0.2m/s will evaporate within 8 minutes.

*Clarity or presentation*

There are several items that require an explanation or need a more detailed description.

10. What is the definition of a cloud employed in this study? E.g., LWC>X, or N>Y or something else?

11. In section 3.1, I had a hard time understanding what the total cloud region (TCR) is. Is it a cloud separated from other clouds by a clear sky segment? Or is it an entire cloud domain sampled during a field campaign? If it is the latter, was a cloud free environment included in the statistics? If TCR refers to separate clouds, then how was the calculation of cloud statistics performed? i.e., were TCRs normalized on their spatial extension?
12. Definition of mixed-phase clouds:
    (a) The definition of mixed-phase based on LWC (or IWC) mass fraction LWC/TWC (or IWC/TWC) has been used in the cloud physics community for approximately thirty years. It is worth acknowledging this in the paper.
    (b) The second definition of mixed-phase, based on particle concentrations, is $N_{liq}/(N_{liq}+N_{ice})$, where $N_{liq}$ and and $N_{ice}$ are the concentrations of droplets and ice particles, respectively. Since, for most clouds, $N_{liq}$ is typically larger than $N_{ice}$ by 3 to 5 orders of magnitude, with a very few exceptions the ratio $N_{liq}/(N_{liq}+N_{ice})\cong 1$. Therefore, since $N_{liq}/(N_{liq}+N_{ice})>0.9$, the majority of clouds should fall in the category of liquid clouds. This is clearly inconsistent with the results shown in the diagram on Fig.4b. This contradiction requires an explanation.
    (c) The spatial ratio is defined as Length(cloud type)/Length(total). In this regard, the statement on line 165: " *…for each TCR, ice spatial ratio is calculated as length of (ICR+MCR) / length of TCR*" sounds contradictory to this definition. If the definition of the spatial fraction is different from that stated above, then a more detailed explanation is required. Also, note that the spatial ratio was used for characterisation of mixed-phase clouds in Korolev et al. (2017, Fig.5-13a).
13. It would be beneficial for this work to discuss the effect of the WBF process and glaciation on the thermodynamic state of mixed-phase clouds. I found no mention of the glaciation process. The WBF was mentioned only once at the end of the paper.
14. The rapid increase of occurrence of ice clouds in the temperature range -15C to -20C was observed by other research groups (e.g., Wallace and Hobbs 1975; Moss and Johnson 1994; and others), which is worth acknowledging here.
15. It is worth indicating sampling statistics (cloud length) for each cloud type in Table 1.

*Concluding remarks*

Given the amount of work invested in this study, I would encourage the authors to rewrite the paper accounting the above comments. I did not consider any minor comments since they are eclipsed by the major issues of this work. My biggest concern is related to the data quality issues. Fixing other issues is just a matter of time.

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