Review of "In-situ Observation of Riming in Mixed-Phase Clouds using the PHIPS probe" by Waitz et al.

Overview

The paper presents an interesting study of in-situ observations of rimed ice. This study is essential in view of large errors in cloud models and numerical weather prediction simulations related to the overproduction of graupel. The authors performed a large amount of work manually analyzing over 5000 ice particle images collected in mixed-phase clouds. The results of particle images classification were correlated against bulk microphysical and state parameters. The obtained results could be used for the validation of cloud microphysical schemes in cloud simulations. Besides the merits of this work, I found that the text of the paper requires improvements focusing on clarity of formulations and accurate statements. In my opinion, this could be easily fixed. My biggest concern is the quality of measurements of humidity and vertical gust velocity used in this work. I strongly recommend contacting the data managers of the ACLOUD, SOCRATES, IMPACTS projects and asking them to redirect you to the researchers responsible for the collection and postprocessing of these measurements for further consultation. In my opinion, the paper undoubtedly deserves publication in ACP. However, keeping in mind the issue with the data quality, I do not have a choice other than recommend major revisions.

Recommendation: The paper can be published in ACP after major revisions and addressing the comments listed below.

Major comments

- 1. Page 3, 3rd para: The description of initiation of riming is incomplete. It may also start from the freezing of precipitation size drops, e.g., D>100um. Such frozen drops, due to their relatively large fall velocity are efficiently collect cloud droplets with 10<D<40um. It is also worth mentioning that large drops may work collectors of slower falling ice particles (e.g., ice lollies).
- 2. Page 4: The definition of the riming efficiency is concerning: "The riming efficiency of an ice particle is a function of (i) its collection efficiency and (ii) the number of supercooled droplets, integrated over (iii) the time the ice particle spends in the cloud and during precipitation." I could guess that the authors meant "riming rate" rather than "riming efficiency." However, item (iii) assumes integration of the riming rate over time, which yields the mass of the rimed particle. Therefore, item (iii) cannot be used in the definition of the riming rate. When defining the rate of riming, it would be more strict to consider collection kernel in item (i), and droplet size distribution in item (ii). The entire statement should be reconsidered.
- 3. Lines 156-159: "...data-set were visually classified into seven habit classes: (i) plate-like particles (single plates, sectored plates, skeleton plates, and side planes), (ii) columnar particles (solid columns, hollow columns, and sheaths), (iii) needles, (iv) frozen droplets, (v) bullet rosettes, (vi) graupel, and (vii) irregular particle. In addition to the habits, the particles were assigned the attributes (i) aggregate, (ii) rimed or (iii) pristine." This definition is directly related to the objective of this paper, and therefore, it is worth adding some extra text elaborating on it. It appears that the attribute "non-rimed" is missing here. It is not clear whether aggregates and pristine ice can be rimed or non-rimed. In other words, are "rimed aggregates" or "rimed pristine ice" subcategories of the attribute "rimed" (ii) ice. Into which category will a rimed aggregate fall? To which habit class will aggregates of one column and one plate belong? Should "aggregate" be a habit class rather than an attribute?

- 4. Lines 167-172: "Particles were classified regarding their surface riming degree (SRD) as (i) unrimed (SRD = 0%, no visible riming), (ii) slightly rimed (SRD < 25%, a few scattered droplets on the particle's surface), (iii) moderately rimed ($25\% \leq SRD \leq 50\%$, up to half of the particle's surface is covered by droplets), (iv) heavily rimed (50% < SRD ≤100%, most or all of the particle's surface is covered by rime) as well as (v) graupel (SRD>>100%, the whole particle surface is covered by multiple layers of rime, so that the structure of the underlying particle is no longer recognizable)." As in the previous comment, this definition requires additional explanations. What is the tolerance of determining SRD = 0% ? For example, if an ice particle has one or two frozen droplets on its surface, which category would it fall? Or what is the SRD for the column shown in Fig.3c? What is the probability of misidentification of ice particles as "SRD = 0%, no visible riming"? Could you assess such probability? The criterion "SRD>>100%" looks confusing for the two following reasons. First, the definition of surface riming fraction as SRD=(area of rime)/(surface area of the facetted ice particle prior riming) limits max SRD by 100%. The case "SRD>>100%" would be relevant to the definition of the rime fraction as (mass of rime)/(mass of ice particle prior riming). Second, in the frame of your SRD definition, how would rimed ice particles with, e.g., 100%<SRD<500% be classified? Note, such particles do not fall in the category SRD>>100%? It is not clear how was SRD identified for particle images with non-transparent sections formed due to refraction. Any explanations in this regard would be useful.
- Page 6. Along with the description of the particle probes, could you also provide the description of the humidity sensor, temperature sensor, gust velocity probes, radars employed on the Polar-6, Gulfstream-V and P3. Description of the accuracy of measurements (especially the Doppler velocity) would be useful here.
- 6. Page 8, 1st para: Could you explain why did you limit your consideration by the cases with T>-17C? Could you also indicate the ranges of LWC (min and max values) and Doppler velocity? What is the averaging time of LWC, T, V_{Dopl}, etc.? Based on the title of the paper, I believe that all measurements were performed when LWC> LWC_{min}. It would be relevant to indicate it in this section.
- 7. Lines 184-185: "The riming rate is a function of the relative flux of available droplets and hence droplet number concentration and relative velocity with respect to the ice particle." This is a misleading statement. For example, following this statement, the riming rate of an ice particle will be higher for the cloud consisting of 1um droplets compared to the cloud with 10um droplets, assuming that both clouds have the same LWC. In fact, the 1um droplet will follow the airstream and flow around the ice particle without accretion, whereas 10um droplets will have a high rate of impact with the ice particle due to its larger mass. Therefore, the above statement should consider droplet size distribution instead of the droplet number concentration.

The averaging of the Doppler velocity (V_{Dopl}) over the vertical column is concerning. The vertical Doppler velocity may vary in the vertical direction. Therefore, the averaged along the vertical direction Doppler velocity may be biased compared to that on the flight level. There are many documented examples of the variability of V_{Dopl} in vertical direction available from the literature. Could you please comment on the vertical variability of the measured Doppler velocity? It would be useful to see some examples.

8. Humidity measurements. The results of the RH measurements presented in Figs.9, S4f, S6f, S7f are questionable. As shown in these diagrams and stated on page 17, the average value of RH_{ice} is

centered at ~100%. However, in mixed-phase clouds, water vapor pressure is close to saturation over liquid water, i.e., RH_{liq}=100%. This had been shown both theoretically (Korolev and Mazin, JAS, 2003) and experimentally (Korolev and Isaac, JAS, 2006). After leveling the flight at ~4300m (~12:42:30) the measurements were performed at T=~-5C to -7C. At that temperature saturation water vapor pressure over liquid water would be equivalent to RH_{ice}~105% to 107%. However, as shown in Fig.9 between ~12:44:30 and 12:49 relative humidity over ice was always <5-7% within mixed-phase cloud segments. On the other hand, at ~12:42:30 and 12:44 RH_{ice} peaked up to ~150% and 125%, respectively. Such relative humidity over ice would be equivalent to supersaturation over liquid of ~40% and 18%, respectively. These are unrealistically high supersaturation over liquid water, which do not occur in the free atmosphere. Such behavior of RH_{ice} is suggestive that the humidity measurements had a major issue.

- 9. In situ vertical velocity measurements. As it is seen from Fig.9 (4th diagram from the top) that during descent, the vertical gust velocity (V_z) changed from -15m/s (at the beginning of the diagram) to 0m/s (12:42:30), when the flight was leveled. Such behavior is highly suspicious. It looks that the vertical component of the aircraft speed was not subtracted from the gust velocity measurements. After leveling the aircraft (>12:42:30) the measurements of V_z look reasonable. I am wondering if the inclusion of the time segments with the faulty V_z measurements resulted in a big difference between the fraction rimed ice versus the Doppler velocity (Figs.S4d,S6d,S7d) and versus ambient vertical velocity (Figs.S4h,S6h,S7h)?
- 10. Epitaxial growth: Line 400: "It is further possible that older rime grows on the expense of recently accreted droplets that partly evaporate due to latent heat during the freezing process." It is unlikely that droplets freezing on the surface of the ice particle may have any significant contribution to the epitaxial growth of the pre-existing rime. During freezing, the temperature of the droplet may temporarily increase to 0C and generate within its vicinity (<2D) a region of high supersaturation, which may potentially enhance a diffusional growth of the rime within its direct neighborhood. The endurance of the enhanced supersaturation is limited by the droplet freezing time, which will be decreased by the heat transfer into the ice crystal. For a 20um diameter droplet, the freezing time is estimated to be less than 50ms. Observation of sparse or single rime regrown into facetted crystals (e.g., Fig.9 (Slightly Epitaxial Riming), Fig.S8(CTA1)) on the surface of collector ice particles does not support the proposed hypothesis due to the absence of freezing droplets around, which may source water vapor for the pre-existing rime. The growth of the rime can be simply explained by the deposition of water vapor available from the supersaturated over ice environment, which is always available in mixed-phase clouds.

Minor comments

- 1. Lines 25-26: "*Mixed-phase clouds, …, play a major role in the life cycle of clouds…*" This sentence sounds confusing. It is worth rewording it. You may consider mentioning e.g., the hydrological cycle.
- 2. Line 32: "main growth modes" is worth replacing by "main ice growth modes".
- 3. Line 33-35: "Riming can be divided into two (not always easily distinguishable) sub-topics: riming of small ice particles (diameter $D \cong 100 1000 \ \mu m$) in clouds and riming of large ($1000 \le D \le 5000 \ \mu m$) precipitating ice, graupel and snow particles." This statement is disconnected from the rest of the paragraph. A reader would anticipate the following elaboration of this statement and explanation of division in two subranges. What is the relevance of this statement to the objective of the paper? It is not used in the following text.

- 4. Line 40: "...until gravitational settling becomes efficient." This statement is unnecessary here. Droplets will freeze on the surface of ice particle after enhancement of their gravitational settling as well.
- Line 71: "...discriminate between rimed and irregular particles". Consider replacement by "...discriminate between rimed and non-rimed particles". Note rimed particles fall in the category of irregular ice particles.
- 6. Line 159: "In addition to the habits, the particles were assigned the attributes (i) aggregate, (ii) rimed or (iii) pristine." This definition is directly related to the objective of this paper, and therefore, it is worth adding time text elaborating on it. It appears that the category "non-rimed" is missing here. It is not clear whether aggregates and pristine ice can be rimed or non-rimed.
- 7. Line 186-188: "Further, it is dependent on the collision probability (and hence the cross sections of ice particles and droplets) as well as on the collection efficiency, i.e. the probability that a colliding droplet sticks as rime." This statement has to be modified. Note that collision efficiency is a product of collection efficiency and coalescence efficiency. When talking about the dependence on "the cross-sections of ice particles and droplets", you should also mention ice particle density and its orientation. In general, it is simpler to talk about the collection kernel rather than the parameters affecting it.
- 8. Line 260, 278, 327: "coder temperatures" replace by "lower temperatures" (jargon: temperature cannot be cold or warm).
- 9. Line 340: "warmer temperatures" replace by "higher temperatures".
- 10. Line 263: "orientation within the cloud" replace by "actual orientation with respect to horizon"
- 11. Line 323: "...*investigating the orientation of the freezing of rimed droplets...*" change to "investigating the orientation of crystallographic axes of the freezing of rimed droplets.
- 12. Line 373: "shown in Fig. 9b". Labeling "a" and "b" are missing in Fig.9.
- 13. Line 374: "were not classified since they were identified as potential shattering fragments smaller than $D=100 \ \mu m$." What was the criterion for identification of shattering artifacts?

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