Review of "Conditions favorable for secondary ice production in Arctic mixed-phase clouds" by Pasquier et al.

1 Overview

The paper presents an important in-situ observation of secondary ice production in Arctic clouds from a tethered balloon with the help of a high-resolution imaging probe HOLIMO. The HOLIMO provided photographic quality images of cloud particles, which, in the majority of cases, allowed for unambiguous identification of particles habit and their phase. The in-situ observations were complemented by remote sensing observations, which allowed for accurate positioning of the HOLIMO measurements with respect to cloud boundaries. Ground based measurements of INPs provided a significant contribution to the value and quality of the collected data set. One of the major outcomes of this study is lowering the threshold concentration of ice particles separating primary and secondary ice production. This is different from the past observations when SIP was identified as an explosive increase in the number concentration of ice particles exceeding hundreds and thousands per liter. In the present study, the threshold concentration was reduced to the order of 0.1-10 per liter. The paper provides an important contribution to the understanding of the role of SIP in ice formation and, undoubtedly, it should be published in ACP. The paper is well written, and I do not have any significant comments on this work. There are a few minor comments which are worth addressing prior to publication.

Recommendation: The paper should be published in ACP after minor revisions.

We would like to thank Dr. Korolev for his encouraging remarks and insightful comments for improving the manuscript. We have addressed his comments point-by-point below in italic font and have included the changes in the text and their line numbers in the revised manuscript.
We would like to point out that additional validation checks on the holographic cloud microphysical data were done to improve the data quality. Holograms with too many bright pixels, which may have inhibited the correct detection of cloud particles, were not considered in the data analysis (approximately 5 % of the holograms). In addition, the frequency distributions of the spatial position of the particle in the detection volume were analyzed. The removal of the cluster of artifacts reduced the concentration of water droplets with D<40 µm. For this reason, the exact numbers in the revised manuscript may differ from the original manuscript and the Fig. 3 and Figs. 6-12, as well as Table 1 were updated, while the interpretation and conclusions remain unchanged by this clean-up.

2 Comments

1. Line 204 and throughout the text: "drizzle drops (defined with diameter larger than 64 µm)" Following commonly accepted definition (e.g., Glossary of Meteorology), drizzle is defined as drops in the size range 100 µm<D<500µm. The authors may consider using the term "supercooled large droplets" (SLD) instead. SLD is defined as droplets with D>50 µmm

at T<0C.

Thank you for this comment. We have replaced the term drizzle drops with supercooled large droplets (SLD) throughout the manuscripts, but amended its definition according to our measurement size bins as droplets > 64 µm at temperature below 0 °C.

2. Line 124-126: "In addition, thanks to the low true air speed of HOLIMO on the tethered balloon system and the adequate tower tips, the shattering of ice crystals in the sample volume is minimized." Reduction of the sampling speed will undoubtedly reduce the effect of shattering of ice due to decreasing the kinetic energy of particles impact. However, the HOLIMO inlet axis is unlikely to be always perfectly aligned with the local airflow, and particles with anisoaxial trajectories are likely to be frequently present during sampling. This is specifically relevant to the cases with the turbulent environment. Particle fall speed will also contribute to the deviation of the particle trajectories from the inlet flow. Such particles will impact with the HOLIMO inlet walls at a speed of ~1-2m/s, and they may get fragmented and contaminate measurements shattering artifacts. Fragmentation of freefalling (1-2m/s) particles on impact with a solid surface of observed by Vardiman (JAS, 1978). In this regard, it would be reasonable to make a disclaimer when talking about the effect of shattering.

Thank you for raising this point. The version of HOLIMO used on the HoloBalloon platform does not have an inlet (see Ramelli et al. (2020) and Pasquier et al. (2022)) and the imaging arms are oriented horizontally (see Figure 1 below). Therefore, the shattering of falling ice crystals on the inlet can be excluded from this study. Moreover, we analyzed the frequency distribution of the particles in the 3D sample volume and could not identify clusters of particles in the sample volume, which could be associated with shattered particles, similar to particles with small inter-arrival times. Finally, the kytoon and the payload are automatically orienting in the wind direction with the wind vane. To make this clearer in the text we have now added the following on lines 127-129: "In addition, HOLIMO's open-path configuration and antishattering tips (as recommended in Korolev et al. (2013)) as well as the automatic orientation of the kytoon and the payload in the wind direction mitigate the shattering of falling ice crystals in the sample volume."

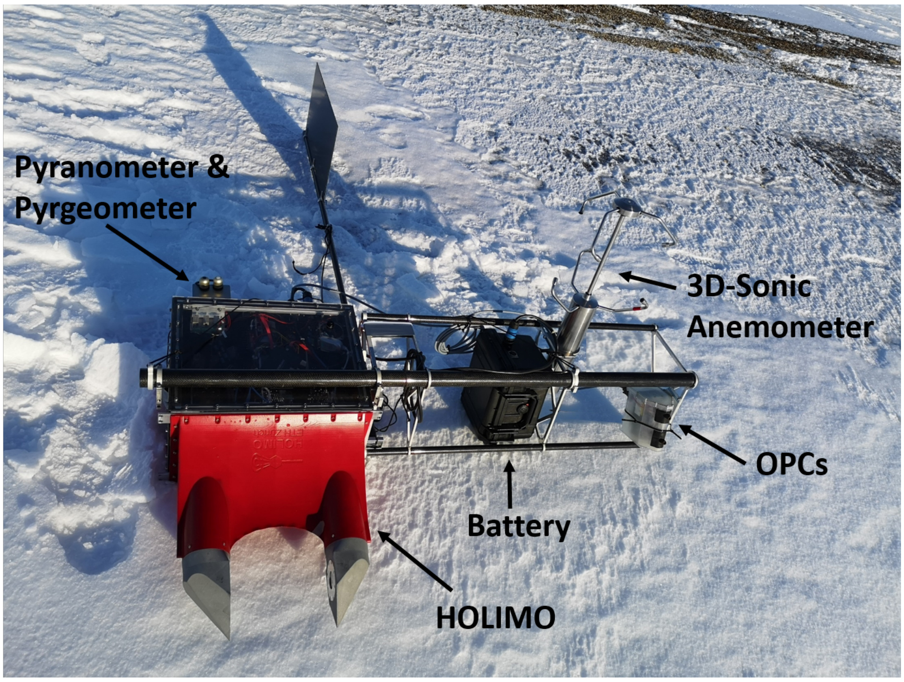

Figure 1: Payload hanging below HoloBalloon with description of the different instrumental parts.

3. Line 158-159: "Non-pristine crystals cannot have formed from vapor deposition growth, and could originate from breakups during impact with the instrument payload or from rime falling from the tethered balloon." The first part of this statement should be corrected. For example, non-pristine ice particles can be formed during water vapor deposition growth on polycrystalline frozen droplets or developed after formation dislocation in the ice crystal lattice of pristine particles.

We fully agree with you. As such we have now rephrased the sentence to state: "We exclude non-pristine ice crystals from the SIP analysis as their habits and thus, the environment in which they grew in cannot unambiguously be defined. This also removes the potential for any falling ice or rime from the balloon to be misclassified as SIP." on line 161-163 of the revised manuscript.

4. Line 290: "Indeed, particles resembling broken branches were observed (highlighted with the dark brown box in Figure 7a)." Could you elaborate why the images of the boxed particles were identified as broken branches. Particle having such shape could be formed without fragmentation.

Thank you for pointing this out. We fully agree and rather meant to give examples of the fragile looking nature of the needles growing on the recirculation particles that could break off and lead to additional SIP. As this is already stated in the previous sentence, we have now removed this sentence from the text and the brown box in Figure 7a.

5. Line 320: "The CDNCs measured by HOLIMO was generally below 1 cm^{-3} except at 13:10 UTC or between 13:45 and 14:15 UTC, when increases in CDNC were observed (Fig. 8b). These comparatively large CDNCs (> 10 cm^{-3}) are observed when HoloBalloon was in

the transition region from low to high radar reflectivity (i.e. in the embedded supercooled liquid layer). It suggests that in this region water saturation was sustained and promoted the formation and growth of cloud droplets, while below, the environment was subsaturated with respect to water and the cloud droplets were evaporating." I have serious concerns regarding the existence of sustainable liquid clouds in the boundary layer with droplets D<40μm and number concentration <1cm^{-3} (e.g., as shown in Fig.8b). Such clouds have a high level of instability and may exist in a relatively turbulent environment only for a limited time since any vertical motion will result in activation of CCNs or complete evaporation of droplets. Therefore, the interpretation of observations of cloud segments with N<0.1cm^{-3} as liquid or mixed-phase causes a question of whether the phase of particles with D<40 μm was identified correctly, or the images of these particles are a result of some non-recognized artifacts. In relation to Fig 8, I would like to add that the rapid increase of the radar reflectivity from -10dBZ to ~10dBZ at approximately 1km altitude is indicative of a presence of a liquid layer there. Such layers usually result in the enhanced growth of seeded from above ice crystals and an increase in radar reflectivity. On the other hand, Arctic clouds are frequently decoupled. I am wondering if the cloud base identified from the ceilometer measurements, as shown in Fig.8a, corresponds to the lower layer. This may result in a perception that the HOLIMO measurements were performed inside a liquid/mixed-phase cloud layer, whereas the measurements in fact were sampled in between cloud layers. Could you also comment on this?

Thanks for raising this point. With respect to the comment about artifacts potentially influencing the identification of the droplets D<40 μm. We would like to point out that after cleaning-up the dataset as mentioned above, the CDNC is reduced between the peaks at 13:10 and between 13:45 and 14:15 UTC. We agree with the reviewer that the regions with CDNC < 1cm^{-3} cannot be considered as mixed-phase or liquid clouds. While validating the dataset, we ensured that the detected cloud droplets are not generally wrongly artifacts classified as cloud droplets. However, as we do not differentiate between cloud droplets and ice crystals below 25 μm, smaller (sublimating) ice crystals are also classified as cloud droplets. Therefore we cannot tell if the detected particles below 25 μm are cloud droplets or ice crystals. We thus agree that in the regions where these low CDNCs are observed, HoloBalloon is likely measuring below the liquid layer. Meanwhile at 13:10 UTC or between 13:45 and 14:15 UTC when the higher CDNCs are observed, HoloBalloon is measuring inside of the liquid layer. This is consistent with the ceilometer observations in Fig. 8a where the track of HoloBalloon is above the detected cloud base. Unfortunately, the cloud base detection is obscured during heavier precipitation and therefore, it is difficult to always determine where the lowest liquid cloud base is. This makes identifying the presence of multiple liquid layers especially challenging. However, as the high CDNCs were only observed when HoloBalloon was at its highest altitudes, it is unlikely that several liquid layers were present between the ground and maximum height of HoloBalloon. Regardless, we intended for this section to state that HoloBalloon is measuring in the mixed phase/liquid layer during the high CDNC periods (e.g. CDNC>10 cm^{-3}) and that it is below this layer when the lower concentrations are observed. We have now reformulated this section to make this clearer on lines 325-329 by stating: "The CDNCs measured *by HOLIMO only reached concentrations higher than 10 cm^{-3} at 13:10 UTC and between 13:45 and 14:15 UTC (Fig. 8b). These higher CDNCs (> 10 cm^{-3}) are observed when HoloBalloon was in the transition region from low to high radar reflectivity (i.e. in the embedded supercooled liquid layer). It suggests that in this region water saturation was sustained and promoted the formation and growth of cloud droplets, while below, in the regions with low CDNCs, the environment was subsaturated with respect to water and the cloud droplets were evaporating."*

6. Line 341-342: "On the contrary, some ice crystals showed broken features, as highlighted by the blue frames in Figure 9a. As the ICNCs were large (up to 55 L^{-1}) collisions between ice crystals have likely occurred." I would be conservative regarding the interpretation of the boxed images in Fig.9a as fragments. Images of ice particles with under- or non-developed corners or branches can be found in Nakaya (1954) or Bentley and Humphreys (1931), respectively. Could you comment or provide strong evidence supporting the fragmented status of some specific particles.

Thank you for pointing this out. First, we have now updated the text to read that the example images are framed in black boxes, not blue, as is shown in the Figure. Second, you are correct, we cannot definitively state that the underdeveloped corners pointed to, are due to fragmenting. To account for this, we have now weakened the statement on lines 346-348 of the revised manuscript to read: "On the contrary, some ice crystals contained underdeveloped corners (highlighted by the black frames in Figure 9a.), which could be a result of recent ice-ice collisions. As the ICNCs were large (up to 78 L^{-1}) collisions between ice crystals likely occurred."

7. Line 342-344: "In addition, ice-ice collisions is believed to be most efficient at colder temperature (Takahashi et al., 1995) such as observed on this day. Therefore, we deduce that the ice-ice collisions were again the most likely active SIP mechanism in the low-level feeder cloud." This statement is based on the observations of ice particles that appear as fragments and association of these observations with ice-ice collision SIP mechanism. This induces the following questions: (a) Could the observed fragments be a result of anisoaxial ice particles impact with the HOLIMO walls? (b) Could the images of ice particles identified as fragments belong to intact particles? (c) Could the observed SIP particles originate from other SIP mechanisms?

Thank you for these pertinent questions. (a) As discussed in comment 2, we do not believe that shattering on the probe arms is a likely possibility as HOLIMO has no inlet and antishattering tips. Moreover, we examined the data for potential contamination from rime falling from the rope or from the kytoon in the measurement volume and removed the possibly contaminated holograms. (b) As discussed in the previous comment 6, we cannot be sure that the underdeveloped corners in the example images shown in Fig. 9a are due to collisions and therefore rephrased these sentence as mentioned in the answer of comment 6. However, the observation of small pristine ice crystals with concentrations up to 19 L^{-1} indicates that SIP was occurring. (c) Droplet shattering, and rime splintering can be ruled out due to the lack of SLD and temperatures far below the HM temperature range. We would like to refer here to lines 343-346 of the revised manuscript: "...the observed *temperature (-24 ° to -18°C) was far below the temperature range of rime splintering (-8°C to -3°C). Furthermore, no large droplets necessary for the droplet shattering process were observed. Therefore, the rime-splintering and the droplet shattering processes are unlikely to have played a significant role as SIP mechanisms in the observed cloud." One other possibility may be the occurrence of the thermal shock process (Korolev and Leisner, 2020), but because of the low CDNC this process which requires the collision between cloud droplets and ice crystals is unlikely to play a significant role in this cloud. Therefore, we believe that ice-ice collision is the most likely active SIP process. We have elaborated on this hypothesis on lines 350-354 of the revised manuscript: "We propose that the large ice crystals sedimenting from the seeder cloud are rapidly growing at lower altitude in the ice supersaturated regions. They could create secondary ice particles by colliding with other ice crystals in the low-level feeder cloud. This hypothesis is in agreement with the recent study by Georgakaki et al. (2022) associating the occurrence of the ice–ice collision mechanism with the occurrence of precipitating seeder–feeder events."*

References

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