Reviewer comments on the manuscript “Secondary ice production within shallow, mixed-phase clouds in cold air outbreaks over the Labrador Sea” by Biggart et al.

The manuscript reports in-situ measurements from the M-Phase field campaign over the Labrador Sea, aimed at understanding cloud formation in marine cold air outbreaks (CAOs). The authors observe that ice crystal counts within clouds often exceed the concentration of ice nucleating particles (INPs) measured outside the clouds, suggesting secondary ice production processes being active. Their conclusions are based on in-situ data, including ice crystal shapes from CPI and 2D-S probes, size measurements of ice crystals and cloud droplets, and cloud microphysical properties such as temperature profiles measured during the ascent and descent flight sections and updraft and downdraft velocities recorded during straight-leg flights. However, the absence of remote sensing data (such as ground-based doppler radar to measure vertical velocities) and comprehensive in-cloud microphysics measurements limits the ability to definitively identify secondary ice production mechanisms. This gap raises questions about the conclusions regarding SIP processes presented in the manuscript.

Many recent in-situ studies have shown a discrepancy between ice crystal number concentration (ICNC) and ice nucleating particle (INP) concentrations, particularly in shallow stratiform Arctic clouds (Li et al., 2021; Luke et al., 2021; Pasquier et al., 2022; Sotiropoulou et al., 2020). Naturally, the immediate interpretation of this discrepancy is that secondary ice production mechanisms are active. However, key questions remain unanswered: what specific mechanisms are active under certain conditions, and how can these mechanisms sustain steady ice crystal production over days despite low updrafts, minimal water vapor supersaturation, and the apparent lack of INPs active above -10°C? Answering these questions is at least as important as delivering yet another evidence of SIP in the shallow mixed-phase clouds.

On a less critical note, the manuscript is clearly written and provides a concise overview of the M-Phase field campaign and its main results. I am confident that the campaign data would provide a solid basis for numerous case studies aiming at reproducing the observed ICNC and potential SIP mechanisms. It is unfortunate that the manuscript describing the observational datasets from the M-Phase campaign (Clarke et al., 2025) is not yet accessible. Access to it would clarify some questions that arise while reading this manuscript.

Below are my specific comments, with original text of the manuscript cited in italic.

1. Page 2, line 75: “*Early laboratory experiments involving a rotating ice-covered metal rod, representing a large riming ice particle, in a chamber full of supercooled liquid droplets were all successful in generating secondary ice; the process was most efficient when temperatures between -3 and -8 °C coincided with high concentrations of droplets with diameter > 24 µm and < 12 µm, and a relative rimer-droplet impact velocity between 2 and 6 m s-1 (Hallet and Mossop, 1974; Mossop, 1978; Mossop, 1985b; Saunders and Hosseini, 2001)”.* This statement neglects the fact that even within the repeated experiments of exactly the same type (rotating icing rod) there was a wide spread of measured splinter production rates and these experiments repeatedly failed to provide any insight into the physical mechanism behind the rime-splintering (Korolev and Leisner, 2020).

2. The compliance of the cloud conditions observed in the M-Phase flights with the above-mentioned criteria (see my comment 1) for efficient riming-splintering SIP is the authors’ main argument for HM being the dominant SIP mechanism. However, the evidence for two of these conditions is actually not convincing: a) the droplets size distribution is not shown and the presence of droplets smaller than 12 µm in diameter is not documented; b) the largest updrafts measured during the straight-leg flights seldom exceed 2 m/s, making the arguments for HM questionable. These issues

have to be discussed in more details.

3. The ICNC measured in stratiform and convective clouds (figure 3b) both show significant enhancement of ice compared to numbers of INPs, indicating SIP activity. This enhancement is undoubtedly stronger in the convective clouds, but also clearly visible for stratiform cloud cases. Given a slow vertical velocity measured in the stratiform clouds (between -1.5 m/s and +1 m/s, Figure 4f), can rime-splintering SIP mechanism be held responsible for the observed enhancement of ICNC?

4. Page 3, lines 80 – 85: "*A more recent H-M experiment conducted by Seidel et al. (2024), using high-speed video microscopy and infrared thermography, found little evidence of efficient ice multiplication by rime splintering; however, this was a controversial result given differences in the experimental setup compared to the earlier studies, such as the representation of graupel particles (single, fixed particle in the Seidel et al. (2024) experiment) and the droplet size distributions used, with almost no droplets of diameter < 12 µm used by Seidel et al. (2024)*". Being different from earlier studies does not automatically mean delivering controversial results. Earlier laboratory studies NOT reproducing the Hallett-Mossop type of experiments (Dong and Hallett, 1989; Emersic and Connolly, 2017) found no evidence for SIP upon freezing of cloud droplets on a rimer surface. The study of Seidel et al. (2024) provided evidence that the underlying mechanism of rime-splintering is not spherically-symmetrical freezing of cloud droplets upon contact with riming ice crystal, whatever the droplet size distribution. With respect to the lack of rotation or lateral movement, no vigorous tumbling or rimer rotation or even turbulent-induced acceleration of a graupel particle in realistic cloud environment could account for lateral (centripetal) acceleration of 6g achieved in the Hallett-Mossop type of riming experiments (a rod sweeping the 30 cm diameter circular path with linear velocity of about 3 m/s).

5. Page 3, lines from 103: "*The ejected water freezes on the drop surface and seals the crack, progressively thickening the shell until a critical pressure is reached causing the drop to shatter, producing many small ice fragments (Korolev and Leisner, 2020). Previous studies suggest the frozen droplet fragmentation mechanism requires large, precipitation-sized drops and therefore favours strong updraft regions found in convective cloud systems*".
A. Recent finding of (Kleinheins et al., 2021) suggests that the cracking and associated SIP events can happen multiple times during the freezing of a single droplet (up to 15 times for a droplet of 300 µm in diameter). The growth of ice shell does not occur via freezing of water ejected from the crack, but via progressive inward growth of ice shell and is controlled by the balance between release of latent heat of crystallization and diffusion-evaporation-convective heat removal into surrounding air. B. Droplet fragmentation has been observed across a wide range of temperatures (between -5°C and -30°C) and droplet sizes, from less than 100 µm (Pander, 2015), to raindrops of up to 2 mm diameter (Pfeifer et al., 2025), so that precipitation-sized drop are not a requirement. Maybe droplet fragmentation could be considered an alternative SIP mechanism for shallow stratocumulus clouds.

6. Page 3, lines 114 and on: "*…mechanical breakup of ice particles was shown to favour hydrometeors of varying sizes with different fall velocities, to maximise collisional momentum, and ice crystals with fragile surface characteristics such as riming protuberances or dendritic branches grown by vapour deposition (Sullivan et al., 2017).*" The study of Sullivan et al. is a modelling study and cannot demonstrate the benefit of certain hydrometeors properties or cloud conditions on a particular SIP mechanism; it can only investigate the cloud response based on the numeric representation of the SIP process.

7. Page 6 line 202: "*A threshold number of 10 µm pixels is set in the OASIS software*,…" Please clarify this statement. What is the pixel size of the instrument?

8. Page 8 line 289: "*INP concentrations at higher temperatures (> -9 °C) have previously been assumed to continue to decrease log-linearly in similar mixed-phase CAO clouds (Järvinen et al., 2022); therefore, if we also adopt this assumption here, expected INP concentrations at the warmest In-cloud temperatures observed during M-Phase (~ 0 °C) would fall between* 10^{-2} *and* 10^{-3} L^{-1}". All off-line filter based INP measurements exhibit a steep drop of INP numbers between -10°C and -5°C, see e.g. Petters and Wright (2015). Log-linear extrapolation of the INP concentration trend into warm temperature would produce non-negligible number of INPs at 0°C. This assumption is not correct. On the other hand, this doesn't matter much because there are no reliable INP data points below -10°C.

9. Page 9, lines 315 to the end of the section. The CDP instrument is responsible for measuring droplet size distributions and thus providing the important evidence for the fulfillment of the HM requirements. Does CDP measure only droplets or also ice crystals? Is there any rejection algorithm allowing to filter out ice crystals? Showing a few size distributions for droplets would be very helpful.

10. Page 10, Figure 2. The GOES provides images every 10 min; only one GOES snapshot is provided per flight. How variable was the cloud field during a triangle flight? Was the transition line moving and if yes, how slow / fast? Why is the GOES imagery shown for the flight C332, which is not discussed in detail in the manuscript, but not shown for the flight C326, which is the ice-ice collision break-up flight?

11. Section 2.6, Splinter Production Rate (SPR) Calculation and discussion (Section 3.2.4). These sections give rise to several questions:

    a. If droplets smaller than 12 µm are required for rime-splintering SIP, why are they not accounted for in the calculations of the SPR, as it was done by Harris-Hobbs and Cooper (1987)?
    b. Is the concentration of rimers kept constant in this approach?
    c. The collection efficiency is calculated based on the terminal velocities of both rimers and droplets. Does this also determine collision velocity? Would it not be too small for efficient rime-splintering SIP? Why is the collision velocity not part of the splinter production rate formula?
    d. The CPI provides quite clear images of rimed ice crystals (Figures 7, 10, 16). Would that be possible to derive the fraction of rimed ice crystals and use it for the estimation of SPR?
    e. Can the mass of rime be estimated from the CPI images and how would that translate into the SPR using the Harris-Hobbs and Cooper relationship?
    f. Why is the SPR plotted against the mean ICNC (Figure 12), and not against the concentration of rimers (ice crystals larger than 300 µm)?
    g. Comparing the SPRs shown in Figure 12 with the SPRs from Harris-Hobbs and Cooper (1987)I find a very similar maximum values on the order 0.1 to 1 $L^{-1}s^{-1}$, which is surprising given the much stronger updrafts and ten-fold higher number concentration of droplets compared to this study. A direct comparison of both datasets would be interesting.

12. Case study 3 (flight C326), section 3.2.5 and 3.2.6. As in the case studies 1 and 2, the only "hard evidence" for SIP is the higher ICNC as compared to the numbers of INPs active at the temperature range where the ice crystals have been observed. However, the concentration of INPs at -20°C is strikingly equal to the ICNC observed at the same temperature (Figure 13). Could that be an indication that ice crystals have been actually formed as "primary ice" via immersion freezing and then got mixed into the cloud, with the concentrations strongly fluctuating in the alternating

updraft-downdraft cells?

13. Page 26 Line 850: "*This broad size distribution suggests that ice crystals of varying stages of growth were collocated during SIP 5, thus meeting the requirement for ice breakup that colliding particles have greatly varying vertical velocities (Korolev and Leisner, 2020).*" I don't see how the broadness of size distribution alone is sufficient to decide if ice-ice collision breakup is taking place. Collisions between freely settling compact ice crystals of 400 µm 800 µm do not carry enough kinetic energy to ensure fragmentation; and the information on the in-cloud updrafts is missing. The situation could change if one of the colliding partners would be a fragile snowflake as in the study of Grzegorczyk et al. (2023), but I don't see any diffusion-grown dendrites in the Figure 16a; all I see are rimed ice crystals.

14. Page 29 lines 916-917. "*SIP periods were predominantly observed in updrafts (80%), particularly low turbulence, weak updraft (< +0.5 ms-1) regions, but also frequently in moderate updrafts of up to ~ +2 ms-1 (Fig. 17b) ...*" This is misleading. All discussed SIP regions are not collocated with the regions where vertical velocities have been measured. On one hand, the authors demonstrate the enhancement of ICNC in the SIP regions as compared to the "background" ice crystal measurements; on the other hand, the mean values of updraft velocities measured outside of the SIP regions are used to identify the cloud conditions beneficial for SIP.

15. Page 29 lines 918-921. "*As previously discussed, updrafts provide a favorable environment for SIP, and particularly rime splintering, by supplying moisture to generate sufficient water supersaturation for the activation of aerosol into cloud droplets, which grow efficiently within updrafts to the necessary size (Dp > 24 µm) for riming and the initiation of the H-M process.*" This is very true, but not very helpful for the data analysis, since the supersaturation has not been reported in the manuscript. Besides, the droplet measurements show similar numbers of droplets larger than 24 µm in background and within SIP ranges, which, according to this argument, should imply sufficient updrafts everywhere.

Summarizing, I believe the authors too extensively rely on the established formal criteria for identifying the SIP mechanisms, such as the HM temperature range or collisional break-up temperature range. Recent studies have shown that such criteria do not have a solid physical basis and have to be revisited. The estimation of the splinter production rate leaves many questions open; a proper cloud model with full microphysics scheme is required to answer the question, whether any of the SIP mechanisms or combination of several mechanisms may explain the steady production of secondary ice.

I would strongly advise to reduce the focus on providing "evidence" for one or the other SIP mechanism. The experimental data is the most valuable outcome of this campaign; complemented with detailed droplet size and water vapor content measurements (if available) it could become a foundation for several numeric case studies, where new hypotheses, physical mechanisms and parameterization can be tested.

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