Review of the manuscript: "Measurement Report: Aircraft observations of secondary ice production in convective clouds over the Tibetan Plateau" by Long et al.

General Assessment

This manuscript presents aircraft observations of enhanced ice particle concentrations in convective clouds over the Tibetan Plateau and investigates the potential role of secondary ice production (SIP).

The manuscript places substantial emphasis on identifying specific SIP mechanisms, namely Hallett-Mossop (HM), freezing droplet fragmentation (DS), and ice-ice collisional breakup (BR). In my opinion, the available observations do not support such attribution. While the observations may be consistent with the occurrence of SIP, much of the interpretation regarding the specific SIP mechanisms is speculative and extends beyond what can be inferred from the presented measurements.

The principal weakness of the study is that it attempts to infer SIP mechanisms from correlations between measured ice concentrations and local environmental variables without adequately accounting for cloud dynamics, particle age, transport history, and observational limitations. In addition, several conclusions rely heavily on uncertain identification of liquid drops from optical-array imagery and on a SIP metric that is neither novel nor free from significant uncertainties.

I believe the manuscript could make a useful contribution if it were refocused on observational evidence of enhanced ice concentrations consistent with SIP and if the claims regarding specific SIP mechanisms were substantially reduced.

Recommendation: Reject with encouragement to resubmit

The manuscript in its present form does not meet the standards of Atmospheric Chemistry and Physics and cannot be considered suitable for publication. The central objective, namely identifying and quantifying specific SIP mechanisms, is not achievable with the presented observations. If that objective is removed, much of the paper would require restructuring, including Sections 3.3, 4, and the Conclusions. In addition, several key analyses (e.g., Figure 16, drizzle identification, interpretation of the SIP ratio/IMF, and attribution of cold-temperature ice enhancement to local SIP) would need to be reconsidered from the ground up.

The paper contains numerous minor issues which I did not include in my review. The major concerns outlined below are not isolated issues that can be addressed through routine revision. They affect the conceptual framework of the study and the interpretation of the principal results. Addressing these concerns would require substantial reanalysis of the observations, reconsideration of the methodology, and major restructuring of the manuscript. Consequently, the required changes go beyond the scope of a conventional major revision.

Nevertheless, the observational dataset itself is highly valuable. I would encourage the authors to develop a substantially revised manuscript focused on the observational characteristics of the studied convective clouds, including their microphysical structure and dynamical evolution. Such a paper could focus on the detailed analysis of the 6 July 2014 case and/or provide an observational synthesis of all seven TIPEX-III flights. A manuscript emphasizing robust observations rather than attributing specific SIP mechanisms would likely make a more significant and defensible contribution to the literature.

Major Comments

1. Identification of specific SIP mechanisms is not supported by the observations

A major portion of the manuscript is devoted to distinguishing among HM, DS, and BR mechanisms using associations between elevated ice concentrations and four environmental parameters: temperature, droplet concentration, droplet size, and graupel occurrence.

A fundamental limitation of this approach is that no attempt is made to determine the age of sampled ice particles. As discussed by Korolev et al. (2020), secondary ice particles may be transported substantial distances from their source regions and lose their spatial correlation with the environmental conditions under which they formed. Consequently, the local environment at the aircraft sampling location may not represent the environment where SIP actually occurred.

This limitation is inherent to Eulerian airborne sampling and affects most in situ studies of SIP. Airborne measurements can reliably identify enhanced ice concentrations that may be interpreted as evidence of SIP. However, direct observation of the SIP process itself is generally not possible from aircraft moving at speeds of 100 m s^{-1} or greater.

Therefore, attributing enhanced ice concentrations to specific SIP mechanisms on the basis of local measurements of graupel, droplet size, droplet concentration, and temperature is not justified. I strongly recommend removing, or substantially reducing, the discussion of SIP type throughout the manuscript and focusing instead on observational evidence of enhanced ice concentrations.

2. Enhanced ice concentrations observed below −8°C do not demonstrate active SIP at those temperatures

One of the central conclusions of the manuscript is that SIP remains active over a broad temperature range extending from approximately −3°C to −16°C and that multiple SIP mechanisms operate throughout this range.

However, the manuscript appears to assume that enhanced ice concentrations observed at colder temperatures were generated locally. This interpretation is not unique and may not be the most likely explanation.

An alternative explanation is that large concentrations of secondary ice particles formed at warmer cloud levels were subsequently transported upward by convective updrafts. Such vertical transport can readily produce enhanced ice concentrations at temperatures below −8°C without requiring active local SIP at those temperatures.

Korolev et al. (2020, 2024) discussed this issue extensively and demonstrated that enhanced concentrations of secondary ice particles can extend from regions immediately above the melting layer to substantially colder cloud levels. The presence of enhanced ice concentrations at cold temperatures does not necessarily imply that SIP was initiated there. Rather, these particles may simply have been transported from warmer levels where SIP was active. The central conclusion of that study was that airborne observations can only identify active SIP when very young ice particles are observed and cannot reliably infer SIP source regions from aged ice populations alone.

The present study does not distinguish between: (a) local SIP occurring at temperatures T< −8°C, and (2) secondary ice produced at warmer temperatures and transported upward. The available observations do not appear capable of separating these two scenarios. Therefore, the statement that SIP is active throughout the entire temperature range down to −16°C is not supported by the presented evidence.

The observations demonstrate that enhanced ice concentrations were observed down to −16°C. They do not demonstrate that secondary ice production was actively occurring throughout this temperature range.

3. The introduction of the SIP ratio is overstated and its limitations require greater discussion

The manuscript introduces the quantity

$$\text{SIP ratio} = \frac{N_{ice}}{N_{INP}}$$

as a primary metric for characterizing SIP.

The presentation of this metric creates the impression that it represents a new approach. However, this is not the case.

Comparison of ice particle concentration (N_{ice}) with ice-nucleating particle concentration (N_{INP}) has been used for decades as a basic approach for identifying secondary ice production (Field et al., 2017). More recently, Wieder et al. (2022) formalized this ratio as the **Ice Multiplication Factor (IMF).** Since then, the cloud physics community has used the IMF to characterize SIP events in remote sensing and in situ observations.

Therefore, the "SIP ratio" introduced here is not a new metric but effectively equivalent to the IMF described by Wieder et al. (2022). The manuscript should avoid presenting the SIP ratio as a novel contribution. There is also no need to rename the conventional names established in the cloud physics community. For consistency I will keep using the conventional IMF abbreviation instead of SIP ratio in the rest of my review.

The interpretation of this quantity also requires greater caution. In this study, $N_{INP}$is not measured directly but estimated using the DeMott et al. (2010) parameterization. Such estimates are subject to potentially large uncertainties because primary nucleation may occur on INPs transported by convective updrafts or entrained from above cloud top, whereas aerosol measurements and parameterizations may not accurately represent the INP population responsible for primary ice formation.

Another concern is that Figure 16 includes many observations with IFM values substantially below one. Assuming that $N_{ice}$and N_{INP}characterize the same cloud volume, one would generally expect $N_{ice}/N_{INP} \geq 1$, because each activated INP can generate at most a primary ice particle before SIP occurs. The occurrence of numerous cases with $N_{ice}/N_{INP} \ll 1$raises questions about the representativeness of the estimated INP concentrations and emphasizes limitations of the IMF methodology in the present study.

The uncertainty of the INP estimates should be quantified and discussed in much greater detail.

4. Interpretation of Figure 16 may be affected by self-correlation

Section 3.3 relies on the relationships shown in Figure 16 to describe parameters controlling SIP. However, some of these correlations may be partly mathematical rather than physical.

The IMF is defined as

$$\frac{N_{ice}}{N_{INP}} = \frac{N_{irreg} + N_{round}}{N_{INP}}$$

where both irregular and round particle concentrations are components of the numerator.

Figure 16b shows the IMF (SIP ratio as a function of N_{irreg}. Therefore, N_{irreg} appears on both axes. Likewise, Figure 16a relates SIP ratio to N_{round}, although N_{round} is already contained within the SIP-ratio definition.

As a result, the reported strong correlations may reflect mathematical coupling among the variables rather than independent physical relationships. For a narrow temperature interval, N_{INP} is expected to vary only weakly and can be considered as a scaling coefficient, and much of the apparent relationship may therefore arise from variations in the ratio N_{round}/N_{irreg}. The latter is shown in Fig.16d.

Consequently, the positive correlations between N_{irreg}, N_{round}, and IFM shown in Figure 16 cannot be viewed as independent evidence that drizzle concentration, large droplet concentration, or ice concentration controls SIP intensity.

The authors should either:

- perform an analysis using independent variables,
- quantify the effect of mathematical self-correlation, or
- substantially temper the interpretation of Figure 16.

Without such analysis, Figure 16 provides limited evidence for the proposed SIP mechanisms.

5. Identification of drizzle and large drops is highly uncertain

Interpretation of SIP type largely depends on identifying drizzle and large liquid drops using 2DS imagery. Thermodynamic phase classification is primarily based on particle roundness.

This approach introduces significant uncertainties.

First, 2DS images with 10 µm pixel resolution offer limited shape information for particles smaller than 20 pixels. Compact, non-spherical particles represented by 10-20 pixels can easily appear circular (Korolev et al., 2017).

Second, diffraction effects can cause out-of-focus images of compact ice particles to resemble out-of-focus spherical patterns, leading to potential misidentification as liquid drops.

Third, a spherical appearance does not necessarily indicate a liquid phase, as frozen drops and compact ice particles may also appear circular.

As a result, inferred concentrations of drizzle and large drops may be significantly affected by the presence of ice particles.

This issue is especially relevant for Figure 16a, b, and d. Reported drizzle concentrations above 100 L^{-1} at temperatures near -15°C and −20°C (Fig. 16a, d) are unusually high and uncommon at that low temperatures and raise concerns about the phase-classification methodology. Because SIP interpretation depends on identifying drizzle and large drops, the associated classification uncertainty should be quantified.

6. Evidence for freezing droplet fragmentation is inconclusive

The manuscript interprets CPI images presented in Figure 9 as evidence for frozen-drop fragments produced by active droplet shattering.

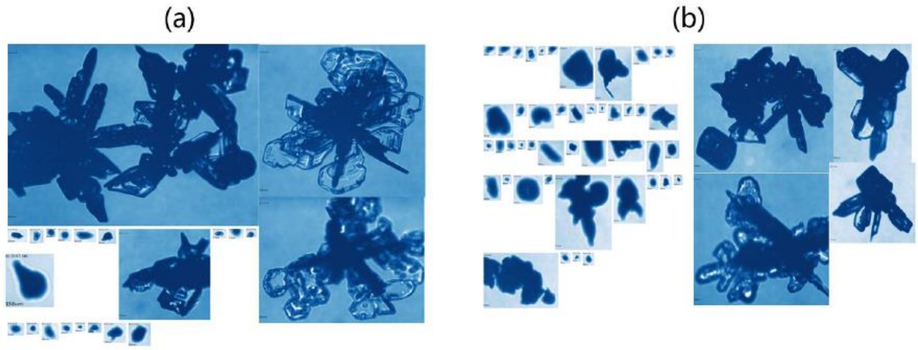

Figure 9: Example CPI images of panel (a) case 1 and panel (b) case 2.

Visual inspection of the images does not reveal clear examples of characteristic frozen-drop fragments. Previous studies have typically relied on identifiable fragmentation products to infer active freezing droplet breakup SIP mechanism.

The evidence presented in Figure 9 is insufficient to support the conclusion that freezing droplet fragmentation was active; thus, this interpretation remains speculative.

The discussion should be significantly toned down.

7. Attribution to collisional breakup is speculative

The evidence supporting the collisional-breakup mechanism is even less convincing.

Currently, no observational technique can determine whether a fragmented ice particle sampled by an airborne probe originated from collisional breakup in the cloud or from shattering upon interaction with the probe itself.

Therefore, attributing enhanced ice concentrations to active collisional breakup remains speculative. While this mechanism cannot be excluded, the presented observations do not provide direct evidence that it occurred.

8. Several conclusions overstate novelty and significance

Several conclusions need revision.

Conclusions 2 and 3 largely repeat results that have been reported in previous SIP studies and should be discussed in the context of the broader literature.

The statement:

"… our results demonstrate that the effective SIP temperature range over the TP extends to −16 ℃ through the coexistence of HM, DS, and BR mechanisms, with the SIP

ratio reaching up to 1000, substantially higher than the enhancement factors in other regions"

is not supported for several reasons:

1. The coexistence of HM, DS, and BR mechanisms has not been demonstrated. On top of that, the coexistence of these mechanisms was discussed in previous observational and modeling studies.

2. Enhanced ice concentrations observed at −16°C do not imply locally active SIP at −16°C.

3. Observations of SIP with IMF values exceeding 1000 have already been reported by Ladino et al. (2017), Yang et al. (2020), Järvinen et al. (2022), and Wieder et al. (2022).

Therefore, neither the mechanism attribution nor the claimed uniqueness of the observed SIP ratios (IMF) is justified.

9. Insufficient meteorological context

Although the manuscript states that the observations were collected within a mesoscale convective cloud system, the meteorological context is not adequately described.

Important missing information includes:

- cloud-top altitude,
- cloud-base altitude,
- melting-layer altitude,
- cloud-top temperature,
- cloud-base temperature,
- cloud lifetime,
- storm evolutionary stage (developing, mature, decaying).

This information is essential for interpreting cloud microphysics and may be obtained from radar and satellite observations.

10. Exclusion of HVPS measurements

It is unclear why HVPS observations were not incorporated into the particle size distributions shown in Figures 10 and 15.

Because the largest hydrometeors often play an important role in SIP-related processes, omission of HVPS measurements results in incomplete characterization of particle size spectra. The authors should either include the HVPS measurements or clearly justify their exclusion.

11. Literature review is incomplete

Over the last several decades, numerous in situ and remote-sensing studies have documented SIP events in a variety of cloud systems and climatic regions.

Table 2 contains only a limited subset of these observations and appears strongly biased toward Northern Hemisphere studies. To place the present work into a broader context, I recommend a substantially more comprehensive review of previous SIP observations and expansion of Table 2.

Overall Recommendation

The airborne observations presented in this manuscript are potentially valuable and may provide useful evidence of enhanced ice concentrations consistent with secondary ice production in convective clouds over the Tibetan Plateau. However, the manuscript currently overinterprets the observations and attributes enhanced ice concentrations to specific SIP mechanisms without sufficient supporting evidence.

The most significant issues are:

- the unsupported identification of HM, DS, and BR mechanisms;
- the neglect of vertical transport as an alternative explanation for enhanced ice concentrations at cold temperatures;
- the uncertain identification of drizzle and large liquid drops;
- the overstated novelty of the IMF (SIP ratio), which is essentially equivalent to the Ice Multiplication Factor introduced by Wieder et al. (2022);
- the limitations of the IMF methodology when applied using parameterized rather than directly measured INP concentrations;
- the potential mathematical self-correlation in Figure 16.

A publishable paper could emerge from this study if the authors focus on robust observational evidence for enhanced ice concentrations and substantially reduce unsupported claims regarding the identification and relative importance of specific SIP mechanisms.

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