Review for: Conditions favorable for secondary ice production in Arctic mixed-phase clouds by Julie T. Pasquier

1 Summary

Pasquier et al. investigate the conditions that favor secondary ice production (SIP) in Arctic clouds, observed during NASCENT campaign, and examine the possible underlying mechanisms. For their investigations they use in-situ cloud microphysical measurements derived by a holographic cloud imager on a tethered balloon system and ground-based remote-sensing and ice nucleating particle (INP) measurements. Their analysis focuses on a six-day period which revealed the occurrence of SIP for about 40% of the time, while very high SIP events (with ICNCs > 10 L-1) were identified in 3.5% of the analyzed data. The highest SIP efficiency was found at temperatures between -3o C and -5o C; interestingly, this was attributed to the drop-shattering mechanism rather than to the Hallett-Mossop process, which has been thought to be the dominant SIP mechanism at these temperatures. Ice-ice collisions is also identified as the second most important SIP mechanism, especially at colder temperatures down to -24°C. This is a very well-written paper and a very interesting study. Models fail to reproduce the micro- and macro- physical structure of Arctic clouds and the description of ice microphysical processes has long been known to be a main contributor to these errors. Also while SIP has been hypothesized to be responsible for the enhanced cloud ice number concentrations often observed in the pristine Arctic environment, where INP availability is limited, the exact SIP mechanisms remain unknown. In general, there are very few in-cloud ICNC datasets from the Arctic and also most of them are not combined with INP measurements, which makes it difficult to quantify the influence of primary ice production versus SIP. This highlights the importance of the present study and the analyzed datasets for understanding ice production in Arctic clouds. For this reason, I recommend this paper for publication after the comments below have been addressed.

We would like to thank the anonymous reviewer for taking the time to carefully review our manuscript and provide insightful feedback that has improved the quality of our manuscript. Each of the comments are addressed point-by-point in bold font and all of the changes in the text and their associated line numbers are posted below.
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 analysed. 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 Fig. 6-12 were updated, as well as Table 1 were updated, while the interpretation and conclusions remain unchanged by this clean-up.

2 Comments:

Line 63-64: maybe discuss a bit how ice shape is expected to influence SIP (with references)
Thank you for this comment. We have reformulated the sentences on lines 65-67 of the revised manuscript: "In particular, large rimed ice crystals were found to increase the rate of splinters ejected during rime-splintering (Hallett and Mossop, 1974) and ice-ice collision (Vardiman, 1978). Particles with complex shapes are more likely to produce fragments during sublimation (Bacon et al., 1998)."

Line 105: since measurements were collected during four months in total, why only such a small sample of six days is presented here? Please explain
During the first campaign in October-November 2019, more flights were performed with HoloBalloon. However, the data analysis of holographic data is computationally expensive and time consuming, thus we limited the analysis to measurements days that we consider the most interesting in this study. Additionally, the limited reachable flight altitude (1000 m) of the kytoon and the harsh Arctic conditions frequently experienced in spring 2020 unfortunately limited the collected sample during the second campaign in March-April. We hope that in the future more robust sampling systems for these conditions will be developed.

Line 111-114: are particles below 25 micro re-examined manually or are they treated as droplets in the analysis? In the case they are treated as droplets, can you estimate the magnitude of SIP underestimation? Fragments generated by drop-shattering can about 10 micro (Phillips et al., 2018)), while these can be even smaller for the other two SIP processes.The same question concerns misclassified ice crystals with circular shape.
Due to the 3 micron pixel size of HOLIMO, determining the shape of anything smaller than 25 microns is extremely difficult (Ramelli et al., 2020). Therefore, all particles smaller than 25 microns are classified as cloud droplets. Consequently some small ice crystals may be misclassified as cloud droplets. However, as these small ice crystals are expected to grow to at least 25 microns in relatively short time periods, e.g. within 100 seconds for columns at the warmest temperatures (Korolev et al., 2020), and as these particles have very slow fall velocities, the concentration of secondary ice particles quantified in this study is likely representative of the mean cloud state. At the same time, any circular ice crystals larger than 25 microns, are likely frozen droplets that have not become faceted yet and these would indeed be classified as cloud droplets. Although these droplets may have contributed to the production of secondary ice due to droplet freezing and shattering (e.g., Lauber et al., 2018) they are not technically secondary ice particles themselves. This means that any frozen droplets that have not become faceted and are still classified as cloud droplets should not be considered secondary ice and therefore, their classification as cloud droplets is not influencing the secondary ice concentrations. As mentioned in the text, any cloud particle classified as an ice crystal by the neural network (Touloupas et al. (2020)) and is larger than 25

microns is then hand-labeled and confirmed as ice or not.

Line 114-115: could you provide more details on the criteria (characteristics) used to classify to ice particles as recirculated or aged? How do you separate these two categories?

Thank you for raising this point. As described in Section 3.2, recirculation particles were growing in different preferential temperature growth regimes (plates or columnar). This gives them more than one primary habit, e.g. capped columns with columns growing out of the plate corners. In contrast, aged ice crystals are rimed, aggregates or have irregular shapes and thus their form was likely shaped by interacting with other particles. One manuscript was submitted to GRL describing in detail the formation of the recirculation particles (Pasquier et al., 2022b). To make the difference between aged and recirculation particles clearer, we have reformulated the sentence on line 115-118 of the revised manuscript to : "All ice crystals were manually classified into habits based on their 2D shape to plates, columns, frozen drops, recirculation particles showing evidences for growth in the plate and columnar growth regimes (see Section 3.2 and Pasquier et al. (2022b) for details), and aged particles that comprise rimed, aggregated, and irregular ice crystals."

Lines 125-126: what do you mean 'minimized'? Could you provide an estimate for how frequently shattering occurs? You should use the inter-arrival time algorithm (Korolev and Field, 2015) to identify shattering artifacts and exclude them from the analysis.

Thank you for this comment. We totally agree that using an inter-arrival time algorithm is a really nice technique for eliminating the shattered particles in 2D-optical array probes. However, unlike the quasi-continuously measurements of 2D-optical array probes, holographic instruments only capture an image at a fixed frame rate e.g. 6 Hz. From a single image, the properties of all the particles (up to 1000) inside the sample volume are retrieved. This means that there is no inter-arrival time between particles as all of the particles in the sample volume are captured simultaneously. Nevertheless, to reduce any potential impact of shattering, the reconstructed sample volume is selected such that the volume within 2 cm of the tips is excluded from the analysis. Additionally, we analyzed the frequency distribution of the particles in the 3D sample volume and could not identified clusters of particles in the sample volume, which could be associated with shattered particles, similar to particles with small inter-arrival times. Together with the fact that our measurements are taken at low wind speeds and without an inlet that particles can shatter on, we are convinced that shattering is minimized.

Lines 153-154: There is something I don't understand about this method. Why pristine ice crystals with size < 106 microns cannot be newly-formed primary ice crystals? Why these should solely be associated with SIP? Please explain.

Indeed, we fully agree that pristine ice crystals at these small sizes can also be formed via primary ice nucleation. However, when comparing the measured ice nucleating particle concentrations to the concentration of small ice crystals, we find that the small ice crystal concentrations are higher by orders of magnitude. Therefore, you are correct and some of the small of the pristine ice could be primary ice, but the fraction of these crystals is very small relative to those that must have formed *via secondary ice processes.*

Lines 180-181: provide reference

Thank you for pointing this out. We have now added citations to Li et al. (2022) and Pasquier et al. (2022a) who present concurrent INP measurements that show an exponential relationship between INP concentration and temperature.

Lines 193-195: I am not convinced that the profiles derived on 8/11 and 11/11 are well- mixed. A θ-gradient of 0.5 C is often used as criterion for decoupling (Sotiropoulou et al., 2014; Gierens et al., 2020). Please use one of the proposed methods in the literature to ensure cloud-surface coupling.

Thank you for this comment. We calculated the difference in potential temperature between the surface level and at the height halfway to the liquid base height, following the technique introduced by Gierens et al. (2020). The difference in potential temperature amounted to 0.65 K on 8.11.2019 and 0.63 K on 11.11.2019. If this difference is slightly above the 0.5 threshold used in the literature, we think that it is still correct to state that 'no strong decoupling case was observed'.

Lines 238-239: you do not provide any information on updraft velocity for November 10 to support this statement. I suggest to provide a time-height cross-section for this parameter (at least in the appendix or as supplementary material).

Thank you for pointing this out. We were not giving the updraft velocities here, as we think that the wind speed and direction information shown in Figs. 3b and A1 are sufficient to prove that the dynamics were weak. Indeed, we can see from these Figures that the wind speed was below 5 m s^{-1} and that no wind shear was prevailing in the sampled cloud. We have however added information about the updraft speed on lines 237-238 of the revised manuscript: "The dynamic was weak within this cloud, as the horizontal and vertical wind speeds did not exceed 5 m s^{-1} (Fig. 3b) and 1 m s^{-1}, respectively."

Section 3.2: While this section focuses on the investigation of the high SIP event in the afternoon of 11/11, it is worth including a short discussion for the possible drivers of the weaker SIP before 18:00.

Thank you for this comment. We agree that it is interesting to discuss potential SIP mechanism for the weaker SIP before 18:00 UTC. We have now added the information on lines 276-278: "Ice crystals observed before 18:00 UTC are largely aged particles, whereas ice crystals observed during SIP periods starting from 18:10 UTC were frozen drops, recirculated particles..." and on lines 279-281: "The presence of aged particles together with cloud droplets smaller than 12 µm and larger than 24 µm before 18:00 UTC suggests that the rime-splintering process could be responsible for the $ICNC_{pr<106\,\mu m}$ below 5 L^{-1}. "

Lines 332-333: However, peaks in the columnar ice concentrations before 13:00, which are of similar magnitude as the one observed round 13:00-13:15, are not associated with CDNCs

increases. What is the reason behind their formation?

Thank you for raising this point. The peaks in $ICNC_{pr<106\,\mu m}$ *before 13:00 UTC are of similar magnitude as the one at 13:10-15 UTC, however columns and plates contribute to approximately the same extend to the increase, whereas at 13:10 and 14:00 UTC the columns clearly dominate the increase in* $ICNC_{pr<106\,\mu m}$*.*

Line 333: There seems to be an almost constant white shading at sizes between 7-8 micro in both Figures 6 and 8. Is this some kind of artifact?

Thank you for this comment. Yes, this was an artifact caused by the choice of bin sizes of the cloud droplet size distribution. We have now corrected this artifact in Figures 6 and 8.

Line 342: It worths discussing here a bit more about the ice-ice collision process. Is it the same mechanism here as proposed by Geogakaki et al. (2022) for seeder-feeder events? They suggest that ice particles falling from the upper cloud collide with ice particles within the lower mixed-phase layer, resulting in mechanical break-up. And if this the case, why the process does not take place earlier, since the seeding-feeding system is observed from approximately 12:00 to 14:00?

Thank you for this comment. First, we want to make clear that SIP was occurring during the entire flight, e.g. $ICNC_{pr<106\,\mu m}$ *amounted up to 7* L^{-1} *at around 12:20 UTC (see Figure 9b). Second, we can not say with complete certitude that it was the same mechanism as in Georgakaki et al. (2022) occurring, but it is very likely that the particles falling from the upper cloud were contributing to the formation of secondary ice crystal by ice-ice collision. Therefore, we have now added the following on lines 350-352 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."*

Line 344: please provide the reference of Georgakaki et al. (2022) here, who reached the same conclusion about seeder-feeder cases.

Thank you for pointing out the agreement with this study. We have now added the sentence on lines 353-354 of the revised manuscript: "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."

Line 344: do you exclude the possible contribution of sublimation break-up? Deshmukh et al. (2022) suggest that as precipitation particles falling from the seeder cloud into a sub-saturated environment may experience sublimation break-up. Then, as the new fragments enter the saturated conditions of the feeder cloud, they can further grow through vapor deposition and enhance ICNCs (see their schematic in Fig. 14).

Thank you for this comment. We agree with the argumentation of Deshmukh et al. (2022) that precipitation particles falling from the seeder cloud into a sub-saturated environment may experience sublimation break-up and enhance the ICNC in a low-level feeder cloud. However, these sublimating *ice crystals must still be large enough to overcome the updraft and sediment in the lower cloud and these ice crystals likely do not exhibits pristine shapes. As in this study we use the concentration of small pristine ice crystals to evaluate local SIP, ice crystals having experienced sublimation break-up during sedimentation would not be detected as SIP particles. We want to focus on the SIP occurring close to the measurement location in this study, and thus for consistency opted to exclude a discussion on all of the possible SIP process that could have been occurring in other parts of the atmosphere.*

Line 361: Could the correlation between ice-snow concentrations be due to the fact that SIP particles will eventually grow to snowflakes? This means that increases in snow concentration would follow the increases in the number smaller ice particle.

Thank you for raising this point. In general you are correct that the snowflake-SIP particles connection suffers from the chicken-egg problem: the SIP particles growing to larger sizes could be used in the correlation. To prevent this, we use only snow crystals larger than 327 µm, as we state on lines 366-369 of the revised manuscript: "The analysis of the influence of ice crystals on SIP is delicate because it is possible that the larger ice crystals are secondary ice crystals having grown to larger sizes than the threshold used (106 µm). To overcome this issue, we discuss only the connection between SIP and ice crystals larger than 327 µm, and refer to these as snow crystals." Concerning the time lag between SIP particles and snow crystal concentration increase, one important fact to keep in mind, is that the ice crystal only stay associated with their environment of origin for a certain time period. Therefore, larger snow crystals are likely to have experienced horizontal and/or vertical advection or turbulent diffusion and to not all originate from the same cloud region. Therefore it is inappropriate to make conclusion about peaks in snow crystal concentrations following peaks in SIP.

Line 367-368: Again, the contribution of sublimation break-up should be investigated here (see comment above).

Thank you, as answered above, with our approach we only investigate SIP occurring close to the measurements location and therefore not the SIP at higher altitude.

Line 385: How sensitive are the results to the choice of this CDNC threshold?

Thank you for this comment. We calculated the OEF using CDNC threshold between 3 and 10 cm $^{-3}$. *The results presented in Table1 below show that the frequency of occurrence is sensitive to the threshold used, but the OEF are not strongly sensitive to the CDNC threshold used. Indeed, for* SIP_{all}, SIP_{low}, SIP_{mod} *the OEF are lower than 1 for all thresholds and for* SIP_{high} *the OEF are higher than 1 for all thresholds.*

Table 1: Frequency of occurrence and OEF of the hydrometeor types cloud droplets with concentrations CDNC from 3, 5 and 8 cm $^{-3}$ as threshold during all measurements (N_{all}), SIP_{all}, SIP_{low}, SIP_{mod}, and SIP_{high}.

		N_{all}	SIP_{no}	SIP_{all}	SIP_{low}	SIP_{mod}	SIP_{high}
CDNC threshold=3 cm $^{-3}$	F (%)	47.15	50.36	42.05	45.61	34.32	58.82
	OEF			0.83	0.91	0.68	1.17
CDNC threshold=5 cm $^{-3}$	F (%)	33.32	35.88	29.26	31.71	22.79	45.88
	OEF			0.82	0.88	0.64	1.28
CDNC threshold=10 cm $^{-3}$	F (%)	20.65	23.29	16.47	18.05	10.99	32.94
	OEF			0.71	0.77	0.47	1.41

Lines 386-387: This comment concerns the small OEF for cloud droplets. The presence of cloud drops is expected to be important for SIP within the Hallet-Mossop temperature zone, but not outside of it. If you calculate the OEF only for the H-M zone, does the factor changes significantly?

Thank you for this comment, this is an interesting point. Yes, if we calculate the OEF only the Hallett-Mossop temperature range, the OEF for the cloud droplet increase by a factor of 2 to 3 (see Table 2 below). However, one need to keep in mind that by taking only the HM temperature range into account, one is mostly looking at the two cases on 11 and 12 November, when high SIP events were occurring, but most likely because of droplet shattering and not of rime-splintering, as discussed in the manuscript.

Table 2: Frequency of occurrence and OEF of the hydrometeor types cloud droplets in at all temperature ranges and only in the H-M temperature range.

		N_{all}	SIP_{no}	SIP_{all}	SIP_{low}	SIP_{mod}	SIP_{high}
CDNC at all temperatures	F (%)	33.32	35.88	29.26	31.71	22.79	45.88
	OEF			0.82	0.88	0.64	1.28
CDNC at H-M temperature	F (%)	31.16	25.27	35.54	39.28	28.33	42.6900
	OEF			1.41	1.55	1.12	1.69

Lines 400: updraft speeds are hardly discussed in this manuscript. I suggest to add a figure for this variable and discuss this more thoroughly in relevance to SIP occurrence and drizzle formation.

Thank you for this comment. We were investigating the influence of updraft and wind shear on SIP formation, as it is evident that updrafts are undeniably important for the formation of drizzle drops and large aged ice crystals that were found to be related to SIP. For this investigation we used the updrafts derived from the Doppler spectra as discussed in the supplementary material, as well as the updraft from the wind lidar (see Figure 1 below). However, we found no direct connection between updraft or wind shear and SIP. E.g., there was no evidence for an increase in updraft or *wind shear in regions with SIP. For example, on 11 November 2019, the high SIP regions were located in regions with moderate updrafts (1 $^{-1}$ to 2 m s^{-1}) and above the wind shear layer. On 12 November 2019 however, the high SIP regions were located in low updraft regions (<1 m s^{-1}) below the wind shear layer (see Figure 1 below).*

A likely reason for the lack of connection between SIP and updraft or turbulence, is the insufficient quality and resolution of the wind measurements. Both techniques (wind lidar and Doppler spectra) are not resolving updraft fluctuations or turbulence on the meter scale due to limitations in temporal and vertical resolution of the measurement. Additionally, the signal obtained from the wind lidar is attenuated by the presence of hydrometeors and wind speed measurements are lacking in the interior of the clouds or during strong precipitation events. The radar Doppler spectra -based technique relies on the slower edge of the Doppler spectrum to be representative of particles that can act as tracers for air motion, and it is influenced by the hydrometeor population and amount of turbulence. For these reason, we have omitted the discussion of the impact of wind and turbulence on SIP.

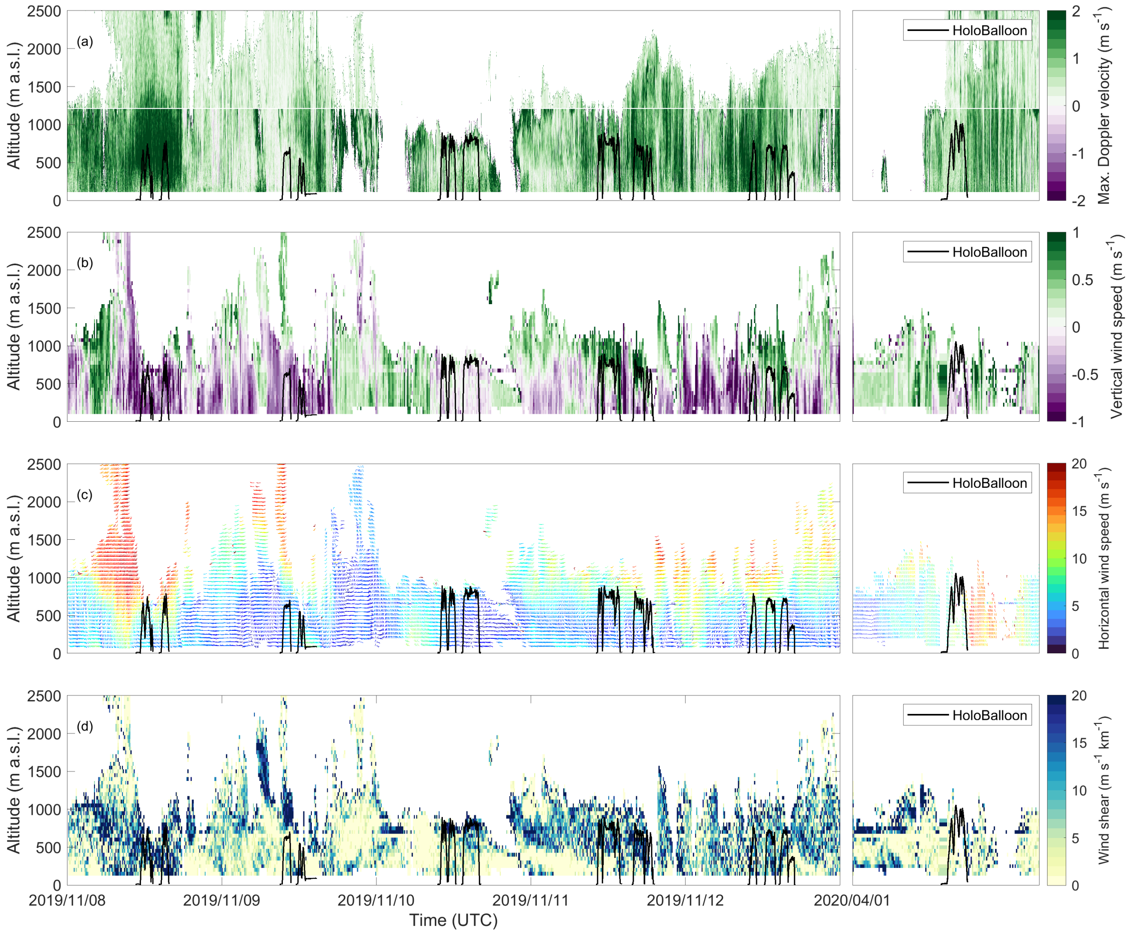

Figure 1: (a) Updraft velocities from the maximal Doppler velocity derived from the cloud radar Doppler spectra. The white line at 1200 m and the attenuated signal above is caused by a change in measurement settings of the cloud radar at this altitude. (b) Updraft velocities measured by the wind lidar. Note the different colorbar limit in (a) and (b). (c) Horizontal wind speed and direction measured by the wind lidar. (d) Wind shear derived from the wind lidar horizontal wind.

Line 433: Please add the reference of Luke et al (2021) here. They were the first to show that high SIP events are associated with the presence of large drops in Arctic clouds. It is worth trying to relate your analysis to their findings, derived with remote-sensing methods.
Thank you for this comment. We fully agree with you and have added the statement: "This would be in agreement with laboratory studies showing that a large number of splinters (>10) can be produced from the freezing of a single drop (Lauber et al., 2018; Korolev and Leisner, 2020) as well as with recent remote sensing studies showing that high SIP events are associated with the presence of large drops in Arctic clouds (Luke et al., 2021). " on lines 440-443 of the revised manuscript

Line 450: Could you infer a minimum INPC that is necessary to initiate SIP from your measurements? Or at least an INPC level below SIP is never expected to occur.
Thank you for this comment. It would be great to give a minimum INPC that is necessary to initiate SIP. Unfortunately, it results from this study that a unique threshold cannot be given, but that this threshold

depends on other parameters such as the atmospheric dynamics or the presence of supercooled drizzle drops. Indeed, as the ice crystal formation was limited by the INPC on 10 November 2019, the INPC was not lower than on the other days, but the dynamic was likely inhibiting the initiation of SIP.

Line 461: This is a statement that is not clearly supported by the analyzed data. The connection of updraft velocities, CCN and drizzle concentrations should be shown explicitly in the figures.
Thank you for this comment. It is true that we don't show in figures showing explicitly the connection between updraft velocities, CCN and drizzle. The reason is that such a discussion would require a study on its own and is beyond the scope of this paper, which focuses on SIP. However, past studies have related CCN and updrafts to the formation of drizzle drops and a publication using data from the NASCENT campaign is currently in preparation for publication on this topic (Motos et al., in prep). To make more clear that we suggest that the formation of drizzle drops is linked to the low CCN and sufficient updraft but without proving this in the paper, we changed the sentence on line 469-470 of the revised manuscript to: "We suggest that the presence of SLD itself is related to the strong updrafts and low CCN concentrations observed in the clean Arctic environment.".

Lines 465-466: if the contribution of sublimation break-up cannot be excluded with the existing data, maybe the possibility of having more mechanisms activated should be addressed here.
Thank you for this suggestion. As discussed in the comment above, we only discuss the prevalence of SIP close of the measurement location and therefore we don't discuss the prevalence of sublimation breakup in particles precipitating from the seeder cloud.

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