

We thank our reviewer Andy Heymsfield for this very thorough and constructive feedback. The incorporated suggestions significantly improved the quality of the manuscript. In the following, we address each comment and point to the according changes in our manuscript. The reviewer comments are displayed in italics, while the responses are given below each comment with the according changes in the manuscript in blue.

## General comments

1. *This is obvious but several cases that support your conclusions would have been desirable.*

We agree that several cases would have been desirable for a more conclusive analysis. However, the meteorological conditions (predominant wind direction in Klosters from southern/western direction) and instrumental issues did not allow for more observations under similar conditions during the RACLETS campaign. However, we plan to investigate this process in future campaigns and test the parameterization for other case studies.

2. *Many studies based on in-situ aircraft observations, especially those in tropical regions, have sampled updraft regions (some of them with weak vertical motions) with comparable size drops that have not identified secondary ice particles (SIP) from particle probes, including holographic imagers and the cloud particle imager (CPI) in this temperature range.*

Thank you for this comment, which led to a more thorough literature research. However, we could not find any of the mentioned studies, which showed comparable droplet sizes but no SIP. On the contrary, all studies with similar conditions support our findings and a discussion of these studies was added to the manuscript (page 19, line 8-18):

### **“4.4 Other case studies with similar observations**

Several other studies observed an increase in the concentration of small ice crystals at the presence of large supercooled drops in clouds (e.g., Stith et al., 2004; Lawson et al., 2015; Keppas et al., 2017; Korolev et al., 2020; Lloyd et al., 2020). Updrafts were made responsible for the origin of these large droplets in all of the studies. The measurements were taken mostly at temperatures lower than during our case study, where newly formed ice crystals grow into columns. The recirculation process through the melting layer described above (Fig. 7) is therefore also expected to play a role for droplet fragmentation in higher regions of the cloud if the updrafts are strong enough to lift drizzle drops high enough until they freeze. Besides droplet fragmentation, the rime-splintering process is expected to be active in the temperature regime between -3 °C to -8 °C, which makes the assignment of the observed secondary ICNC to a specific process more difficult. However, images of deformed frozen drops in all of the above mentioned studies strongly support that droplet fragmentation was active and should be accounted for besides the rime-splintering process.”

3. *It seems entirely possible that snow that could fall through the updraft into the melting layer partially melted and created fragments that would have been carried up into the 0 to -3C temperature range and been incorrectly identified as SIP.*

It is possible that ice crystals only partially melted and created fragments before they were carried up into our measurement regime, which is also a SIP process. We added the following paragraph to our manuscript to argue why we think this has a negligible effect (page 13, line 29 to page 14, line 2):

“Ice crystals can partially melt and create fragments, which can be lifted back into the cloud. However, the ice crystals have to be sufficiently small to be lifted, while at the same time, small ice crystals are likely to completely melt before re-entering the cloud. Furthermore, the breakup rates of this process depend on temperature and humidity and largely on the initial shape of the ice crystals (Korolev and Leisner, 2020). Oraltay and Hallett (1989) observed no sublimation breakup for columnar and plate-like crystals and breakup during melting only at relative humidities below 90%. The shapes of ice crystals in our study are mostly solid particles that have columnar and plate-like shapes (Fig. 5) and the relative humidity on the gondola never dropped below 95% (Fig. 3d). Taking all these considerations into account, we assume that ice fragmentation during melting has a negligible effect on the SIP.”

4. *Given my comments below about ice crystal growth rates and terminal velocity, it seems unlikely that the droplets and SIP plates would have resided in this temperature range long enough to have grown to 60 microns and larger.*

The responses are given to the following comments.

5. *Although obvious, how applicable are the laboratory experiments of fragmentation applicable to natural clouds?*

The applicability of the laboratory experiments to natural clouds is very hard to assess at this point in time due to the lack of direct measurements inside natural clouds. The laboratory measurements are therefore only used as the best estimate available. We hope that the parameters can better be constrained with future measurements. They are easy to be adapted accordingly in our parametrization. We added the following paragraph to the manuscript to discuss this shortcoming (page 17, lines 20-23):

“The first main caveat is that the parametrization was derived solely from laboratory measurements. However, the direct observation of SIP by droplet fragmentation is basically impossible as the process happens on a millisecond timescale (Lauber et al., 2018) and the secondary ice splinters can be smaller than 10  $\mu\text{m}$  (Korolev et al., 2020). Therefore, the laboratory measurements are the best estimate available.”

## More detailed comments

1. Page 10, 11. *Is it possible that the small plates are a result of partially melted ice that fell through the melting layer and then partially melted ice fragments were carried up brought the melting layer by the 0.6 m/s updraft?*

See answer to general comment 3.

2. 1, line 7: *small plates at -3C? According to Fukuta and Takayashi (1999), the basic crystal habit is thick plates (>-4.0°C). I recommend providing that reference at this point in the article.*

According to Fukuta and Takahashi (1999), the basic ice crystal habits at temperatures above -4°C are indeed thick plates. However, they do not show any measurements above temperatures of -3°C. In our study, we observed thin plates at temperatures between 0°C and -2.7°C, which can be seen on the particle images of plates in Fig. 5 in the manuscript. Furthermore, Bailey und Hallett (2009) show a habit diagram of ice crystals including thin plates at temperatures warmer than -3°C. Therefore, we argue that the mass dimensional relationship for plates by Mitchell et. al. (1990) is applicable to our case.

3. 1, line 11: *high temperatures > slightly sub-0C temperatures.*

The text was adapted accordingly.

4. 2, 5: *"water vapor pressure" to "relative humidity?"*

If the water vapor pressure is reduced, the relative humidity is also reduced at a given temperature. Therefore, the two terms are used interchangeably in this sentence. We prefer to describe it with water vapor pressure, as it is more consistent with the rest of the description.

5. 2, 12: *"exist" to "can exist".*

The text was adapted accordingly.

6. 2, 13: *I don't think it's necessarily the primary ice that causes SIP.*

We adapted the sentence to also account for secondary ice produced by secondary ice (page 2, line 13-15):

*"The resulting so-called primary ice can create additional ice crystals (secondary ice), which again can fragment and produce more secondary ice by any kind of fragmentation referred to as secondary ice production (SIP) (e.g., Field et al., 2017)."*

7. 11, line 6. You've calculated how long it takes to grow plates of up to 93 microns diameter at temperatures 0 to -3C. The linear growth rate is extremely slow, because the plates are "thick". The Mitchell et al. (1990) mass dimensional relationship for plates is therefore not applicable. What would the growth rate be if the ratio of the diameter to thickness is 1.0?. Please refer to Figure 10 of Fukuta and Takahashi, who give the appropriate axial dimensions. And their terminal velocity, which will govern how long they stay in the 0 to -3C temperature range before being lofted to higher altitudes and lower temperatures.

See answer to detailed comment 2.

8. 13, 20: The ice number concentration at temperatures below -12C or so are not too much higher than the IN concentration. Also, one does not see evidence from in-situ measurements that there are copious numbers of small plate-like ice crystals at temperatures below -12C that would suggest a vibrant SIP process.

Droplet fragmentation requires the presence of large droplets (>~40µm). In the absence of such large droplets, we do not expect droplet fragmentation to take place. In this study, we do not have measurements of cloud particles below temperatures of -12°C (our measurements were limited to a temperature range of 0°C to -2.5°C). Other measurements at such low temperatures with the presence of large droplets showed indeed a high concentration of small plates (e.g., Lawson et al. 2015, Korolev et al. 2020).

9. 15, 13: "larger" to "higher"

The text was adapted accordingly.

10. 16, 5-6. Terminal velocity can be readily calculated for all ice crystal sizes, based on their shape from the holographic images.

There are only equations available for specific ice crystal habits but not for irregular shaped ice crystals. To have a more accurate calculation of the relative velocities of ice crystals and cloud droplets, we now divide the crystals into plates and lump graupel and use given parametrizations to calculate the fall velocity of each ice crystal. This had a noticeable change only in the collision rate of the largest observed droplet (see Fig. 8 manuscript) by a factor of about 0.5. It had a minor effect on the splinter generation rate. See page 16, line 20-30 and all changes in the calculations hereafter:

"To calculate the fall velocities of the ice crystals, we divide them into plates and lump graupel. The former includes the classes plates and unidentified, while the later includes all other ice crystals (see section 2.2 for a more detailed description of the classes). The fall velocity of plates was calculated with the following equation from Pruppacher and Klett (2010) (converted to SI base units) using the maximum dimension of plates  $L_{\text{pla}}$ :

$$v(L_{\text{pla}}) \approx 156\text{m}^{0.14}\text{s}^{-1} \cdot L_{\text{pla}}^{0.86}. \quad (6)$$

To derive the fall speed of lump graupel with a maximum dimension of  $L_{\text{gra}}$ , we use the equation provided by Locatelli and Hobbs (1974) (again, converted to SI base units):

$$v(L_{\text{gra}}) \approx 124 \text{m}^{0.34} \text{s}^{-1} \cdot L_{\text{gra}}^{0.66}. \quad (7)$$

This yields a splinter generation rate of  $0.06 \text{ L}^{-1} \text{ min}^{-1} \pm 0.02 \text{ L}^{-1} \text{ min}^{-1}$  of secondary ice, which is about one order of magnitude below the estimated production rate of secondary ice of  $0.24 \text{ L}^{-1} \text{ min}^{-1} \pm 0.09 \text{ L}^{-1} \text{ min}^{-1}$  derived from the observations.”

11. 16, 25: *Is it even reasonable to assume that 40 micron droplets all freeze and produce splinters? There's no evidence for this from in-situ aircraft measurements.*

The parametrization does not assume that all 40  $\mu\text{m}$  droplets freeze and produce splinters. It provides a probability for both to happen. A 100  $\mu\text{m}$  droplet for example will only freeze with a probability of 27% during the time it is being lifted through the complete measurement volume and when it freezes it fragments with a likelihood of 18% (see eq. (4) in the manuscript). (A 100  $\mu\text{m}$  droplet has a fall velocity of about  $0.4 \text{ms}^{-1}$  and will be lifted with  $0.2 \text{ms}^{-1}$  when an updraft of  $0.6 \text{ms}^{-1}$  is present. To be lifted up 490 m it will thus take 2450 s and the collision rate of a 100  $\mu\text{m}$  droplet is  $f_{\text{col}}(100 \mu\text{m}) = 1.1 \text{e-}4 \text{s}^{-1}$ , see eq. (3) in the manuscript).

## Additional remarks

We would like to point out that we did some essential changes, which are not all part of the responses. They are addressed in the following:

1. We removed Figure 7, which showed a histogram of the sizes of all observed droplets larger than 40  $\mu\text{m}$  as all droplets are now shown in Fig. 8 of the manuscript, where the values of the different parameters are plotted.
2. The observed secondary ice production rate is given as a number with uncertainties instead of a range as this is easier to interpret (page 13, lines 8-9):

“Taking all named uncertainties into account, the rate of secondary ice production during our case study is  $0.24 \text{ L}^{-1} \text{ min}^{-1} \pm 0.09 \text{ L}^{-1} \text{ min}^{-1}$ .”

This change slightly influenced all calculations, which included the observed secondary ice production rate in section 4.3.1.

3. The total splinter generation rate is given per volume and time instead of only per time as this makes its interpretation easier. Furthermore, we no longer consider size bins for the ice crystals but take all ice crystals into account to have more accurate calculations. This changed the equations in section 4.3.
4. The collision rate given in eq. (3) was off by a factor of 0.5 in the old version of the manuscript. This change led to different results and slightly different interpretations. Changes can be found in the abstract, section 4.3.1 and the summary. The following lines of the manuscript were adapted:

Page 1, lines 12-15:

“Based on previous measurements, we estimate that a droplet of 200  $\mu\text{m}$  in diameter produces 18 secondary ice crystals when it fragments upon freezing. The application of the parametrization to our measurements suggests that the actual number of splinters produced by a fragmenting droplet may be up to an order of magnitude higher.”

Page 20, lines 10-13:

“Applying the presented parametrization to our measurements could not explain the estimated concentration of secondary ice and the number of splinters produced per fragmenting droplet has to be higher, i.e., a droplet of 200  $\mu\text{m}$  in diameter has to produce  $99 \pm 62$  splinters upon fragmentation. This number can be reduced to  $44 \pm 26$  if we assume that all droplets larger than 40  $\mu\text{m}$  fragment when they freeze.”

All changes are marked in the final version of the manuscript.

## References

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