## Response to the reviewer comments by Dr. P. Connolly

The authors would like to thank Dr. P. Connolly for giving his thoughtful feedback to our manuscript. We are very thankful for pointing our attention to the paper by Harris-Hobbs and Cooper (1987) and for providing the Python code enabling fruitful discussion of our results. To keep our response concise, we do not include here the full review provided by Dr. Connolly, which can be found at https://doi.org/10.5194/egusphere-2023-2891-RC2, but structure our response in such a way that it addresses the Dr. Connolly's review point by point. The excerpts from the reviewers' comments and criticism are highlighted red and placed in quotation marks. The changes in the manuscript will be indicated by violet color in the revised version of the manuscript.

1. The main criticism expressed by Dr. Connolly is that the experimental conditions in our study would not allow us to observe significant secondary ice production, as reported in the Hallett-Mossop type experiments. In particular, the droplets smaller than 13 µm, that are prerequisite for riming-splintering SIP (Mossop, 1985), were absent in our experiments except for one case corresponding to the droplet generating regime MGD2 and associated droplet size distribution DSD2. In support of his criticism, Dr. Connolly provided a parameterization of SIP rates based on the paper by Harris-Hobbs and Cooper (1987) and realized in the publicly available Python code (see his review). As confirmed by Dr. Connolly in the off-line private communication, there was a mistake in the original version of his code associated with the double integral in his equation (1) which led to the negligible SIP rate according to HH&C parameterization applied to the experimental conditions of our experiments. After correcting his code and introducing further refinements of the parameterization scheme (e. g., usage of diameter instead of the radius and the reduced collision efficiency between the rimer and small droplets according to Löffler and Muhr, 1972), Dr. Connolly concludes that in case of DSD2, the HH&C parameterization predicts the formation of 3 to 5 secondary ice particles with an accreted rime mass of 0.1 to 0.2 mg; while we didn't observe any secondary ice in this experiment, the number is statistically too insignificant to be considered as a proof of the absence of secondary ice production. Therefore, his main conclusion formulated in his review still holds:

"So while I am very supportive of the new set-up for studies of the RS mechanism, I think there is a major flaw in the paper to state that the measurements mean there is no evidence of the RS mechanism".

We have reproduced the calculation of Dr. Connolly and fully agree with the outcome of his numerical analysis. If the underlying physical mechanism of the HH&C parameterization is valid, and the parameterization is universally applicable to all cases of SIP involving collision of a rimer and supercooled droplets, chances of no secondary ice observation in our experimental setup would be high. However, it was not our intention to copy the experimental setup of Hallett and Mossop, with which we might even have been able to see a high number of secondarily produced ice particles. Instead, we aimed to investigate the underlying physical mechanism of potential SIP during droplet-rimer collisions under realistic conditions, especially with respect to the realistic size of a rimer particle.

We thus have to question the validity of the physical mechanism and the applicability of HH&C parameterization to all riming experiments including those that were not conducted in the H&M-type experimental setups.

The parameterization from Harris-Hobbs and Cooper (1987) is based on the observations by Mossop (1978a,b) that the SIP rate was highest when droplets smaller than 13 µm and larger than 24 µm in diameter were present in the droplet size distribution. Later, an explanation was suggested by hypothesizing that larger droplets freezing on top of smaller ones would develop spherical ice shells thus enabling pressure rise and splintering (Fig. 8b in the preprint, e.g., Choularton et al., 1978;

Choularton et al., 1980). The only direct experimental support for this hypothesis was provided by Choularton et al. (1980) where a photograph of *"…droplet of 35μm diameter, showing evidence for the disruption of a protuberance, collected at a speed of 1.5 ms and temperature -7°C*" is shown in their Figure 2c. Note however, that the droplets in this experiment were accreted on a **frost coated needle**; no accretion of smaller droplets has been reported there. The direct applicability of this evidence to the results of H&M-type experiments is, therefore, questionable.

Based on our observations, the underlying assumption of spherical freezing of larger droplets lacks physical evidence. As we state in the manuscript (page 9, line 195-202), we clearly observe all droplets spreading on smooth or rough ice surface instead of freezing spherically at T > -10°C. Our explanation of this observation is that the time required for a droplet to freeze is too long compared to the characteristic spreading time, so that the spreading cannot be arrested upon impact or midway into the freezing process. This observation, as discussed in our manuscript (page 15, line 249-252), was supported by previous studies of Dong and Hallett (1989) and Emersic and Connolly (2017), which have shown that the droplets tend to spread upon impact on smooth and rough ice surfaces at temperature above −10 °C. To the best of our knowledge, a negative control test excluding droplets smaller than 13 μm in diameter has never been performed in the HM-type experiments (e.g., Hallett and Mossop, 1974; Mossop 1976, Mossop 1978a,b; Heymsfield and Mossop, 1984; Mossop 1985; Saunders and Hosseini, 2001).

We changed and added a sentence [page 17, line 317] in the manuscript: *This is lower than the concentration ratios from 0.1 to 2.0 (or higher) used in HM-experiments, in which the efficiency of rime-splintering was found to correlate with the accretion rate of droplets smaller than 12 μm and larger than 24 μm in diameter. Different droplet size distributions were tested in the HM-type experiments (e.g. Mossop and Hallett, 1974; Mossop, 1976, 1978a, b; Heymsfield and Mossop, 1984; Mossop, 1985a; Saunders and Hosseini, 2001), but to our knowledge, a negative control test excluding droplets smaller than around 12 μm in diameter was not conducted.*

On a side note, Mossop wrote in his 1976 paper, that based on his observations, *"the formation of an ice shell around the periphery of an accreted droplet"* can be excluded [page 55, 8i]. Further, he states "*Two lines of attack appear promising. The production of splinters either from needle-like growths or by evaporation."* [page 56].

This brings us to the conclusion, that the correlation between the presence of small droplets and the high SIP rates in **the HM-type experiments** has to be based on a different physical mechanism than a hypothetical spherical freezing of larger droplets. From this follows, that the HH&C parameterization is applicable only to the experiments that closely reproduce the HM-type SIP experimental settings and cannot be used for interpretation (and even much less as an explanation) of our negative results. The detailed discussion of what mechanism it could be is beyond the scope of this response and will be addressed in the follow-up study.

We add the summary of the above discussion to the "Discussion" section of the revised manuscript [page 17, line 327-339]:

*Harris-Hobbs and Cooper (1987) presented a parameterization (HHC) relating the SI production rates and the droplet size distribution featuring droplets smaller than 13 μm and larger than 24 μm (flat DSD). This parameterization is based on the results of HM-type experiments in support of the hypothetical mechanism of spherical freezing of larger droplets landing on top of smaller ones (Choularton et al., 1978; Choularton et al., 1980). Since small droplets were present in only one of our experiments at -5°C, the HHC parameterization predicts no SIP or just a few particles for DSD2, which we, however, have not observed (see the reviewer comment by P. Connolly and our response). While we don't doubt the true nature of the correlation between the flat shape of the droplet size distribution* *and high SIP rates observed in the HM-type experiments, we have found no evidence supporting the underlying physical mechanism. We thus conclude, that the correlation between the presence of small droplets and the high SIP rates in the HM-type experiments has to be based on a different physical mechanism, rather than a hypothetical spherical freezing of larger droplets landing on top of smaller droplets. Thus, the HHC parameterization is applicable only to the experiments that closely reproduce the HM-type SIP experimental settings and cannot be used for interpretation and even much less as an explanation of our negative results.*

2. Technical issues

The reviewer made us aware of some issues regarding the droplet size distributions (Table 1). To fit our droplet size distributions (DSD1-4), we used the following lognormal-fit function:

$$\frac{1}{N}\frac{dN}{dD} = \frac{1}{\sqrt{2\pi}\sigma}\exp\left[-\frac{(\ln(D)-\mu)^2}{2\sigma^2}\right] = \frac{1}{\sqrt{2\pi}\,\ln(\sigma_g)}\exp\left[-\frac{\ln\left(\frac{D}{D_g}\right)^2}{2\,\ln(\sigma_g)^2}\right]$$

with the geometric mean diameter $D_g$ and geometric standard deviation factor $\sigma_g$. As the reviewer noticed, Table 1 in the paper should contain the unitless geometric standard deviation factor together with the geometric mean (median) droplet diameter. Misleadingly, we gave the standard deviation around the arithmetic mean. Table 1 will be corrected accordingly.

| | μ | σ | $D_g$ [μm] | $\sigma_g$ [] |
|---|---|---|---|---|
| DSD1 | 2.90 | 0.08 | 18.39 | 1.09 |
| DSD2 | 3.04 | 0.24 | 22.18 | 1.27 |
| DSD3 | 3.22, 3.46 | 0.08, 0.10 | 25.26, 31.8 | 1.08, 1.10 |
| DSD4 | 3.37 | 0.26 | 31.00 | 1.29 |

Further, we will mention the relative fraction of the two modes of DSD3, which is 0.7 for mode1 and 0.3 for mode2. Also, the y-label of Figure B1 is changed accordingly.

References:

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