Supporting Information for

**Secondary ice production in summer clouds over the Antarctic coast:
an underappreciated process in atmospheric models**

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Supplementary Text S1 to S4

Supplementary Figures S1 to S4

Text S1: Additional ICNC statistics

While mean statistics and maximum values are discussed in the main text in detail, additional statistical metrics are presented in Fig. S1: median ICNCs and the 25th, 75th and 95th percentiles are shown as a function of temperature. FRAG1 is in closest agreement with median observations, while TAKAH substantially overestimates median values; all the other simulations result in substantial underestimates (Fig. S1a). However, all simulations underestimate the 25th percentiles (Fig. S1b), with FRAG1 and TAKAH being closer to observations. In Fig. S1c, FRAG1siz is in good agreement with the observed 75th percentiles, while FRAG1 and TAKAH overestimate these values; the rest of the simulations generally underestimate measurements. Finally, the 95th percentile in FRAG1 and TAKAH is higher than in observations, while CNTRL, PHIL0.2 and PHIL0.3 result in substantially underestimated values; PHIL0.4, FRAG1siz and TAKAHsiz are generally closer to observations (Fig. S1d).

While mean and median ICNC observations are of similar magnitude, suggesting that this parameter follows a normal distribution, in all simulations except CNTRL, PHIL0.2 and PHIL0.3, mean and median results can differ by order of magnitude, suggesting positively skewed distributions. Thus the better agreement of median results between FRAG1 and measurements, compared to the rest of the simulations, is because the end right tail of their ICNC distribution is unrealistically long, including extreme values (ICNCs > 6000 L⁻¹).

Text S2: Cloud Radiative Forcing

The surface cloud radiative forcing (CRF) is calculated as in Young et al. (2019), who used the methodology of Ramanathan et al. (1989) along with the modifications of Vavrus (2006) to exclude the influence of the high surface albedo at high latitudes. The difference between all-sky (as), thus with clouds, and clear-sky (cs) net longwave (LW) and shortwave (SW) radiation is used to calculate the net radiative effect at the surface:

$$CRF_{LW} = LWD_{as} - LWU_{as} - LWD_{cs} + LWU_{cs}$$

$$CRF_{SW} = SWD_{as} - SWU_{cs}$$

$$CRF = CRF_{LW} + CRF_{SW}$$

The sum of CRF_{LW} and CRF_{SW} anomalies for all sensitivity simulations, presented in Fig. S2, is calculated by considering the differences between each simulation and CNTRL. Red (blue) color indicates that more (less) SW radiation reaches the surface than in CNTRL, suggesting the presence of less (more) liquid-dominated clouds. Activating BR and thus enhancing

ICNCs can result in significant CRF biases over the ocean surface, in agreement with Young et al. (2019). Considering the maximum/minimum CRF biases in simulations PHIL0.4 (-78/73 Wm⁻²), FRAG1siz (-45/75 Wm⁻²) and TAKAHsiz (-52/86 Wm⁻²) which improve the representation of the observed ICNCs, our results indicate that including a BR description in climate models can have a significant impact in the projections of the future climate.

Text S3: Interactions between collisional break-up and Hallet-Mossop

To investigate the interactions between BR and H-M, we compare simulations in which the H-M efficiency is either enhanced or turned off. Following the methodology of Young et al. (2019), we remove all liquid thresholds from the H-M description, allowing for the process to become active over the whole droplet spectrum. This modification is applied to CNTRL, PHIL0.3 and PHIL0.4 set-up, resulting in three additional sensitivity tests: CNTRL_NOTHRES, PHIL0.3_NOTHRES and PHIL0.4_NOTHRES, respectively. Furthermore, another three simulations are performed in which H-M is completely deactivated: CNTRL_NOHM, PHIL0.3_NOHM and PHIL0.4_NOHM.

Increasing H-M efficiency has almost no impact on CNTRL_NOTHRES and PHIL0.3_NOTHRES (Fig. S3a). A slight increase in the mean values is noted in PHIL0.4_NOTHRES at lower temperatures, compared to PHIL0.4, while results at higher temperatures remain almost unaffected. Differences in the 95th percentile (N_{isg95}) are only observed between PHIL0.4 and PHIL0.4_NOTHRES, while CNTRL, PHIL0.3, CNTRL_NOTHRES and PHIL0.3_NOTHRES produce similar values (Fig. S3b). Deactivating the H-M process has no impact on the mean ICNCs in all three set-ups (Fig. S3a), while differences in N_{isg95} become obvious only for $\Psi=0.4$ but still remain small throughout the whole temperature range (Fig. S3b).

The results of this section indicate that although the examined clouds fall within the H-M temperature range, this process is fairly ineffective. In all sensitivity tests the H-M process is very weak and cannot explain the observed concentrations, if BR is not included. The BR mechanism, on the other hand, appears much more efficient and results in improved N_{isg} , independently of whether H-M is activated or not.

Text S4: Interactions between primary and secondary ice production

To examine how the uncertainty in parameterizations for primary ice affects SIP efficiency, we perform two sets of simulations by dividing or multiplying the efficiency of all primary ice production mechanisms (immersion freezing, contact freezing and

deposition/condensation-freezing nucleation) by a factor of 10. Specifically, the first set includes CNTRL_INP0.1, PHIL0.3_INP0.1 and PHIL0.4_INP0.1, while the second one consists of CNTRL_INP10, PHIL0.3_INP10 and PHIL0.4_INP10.

Decreasing primary ice production by a factor of 10 inhibits BR multiplication. Note that while maximum N_{isg} in PHIL0.3_INP0.1 and PHIL0.4_INP0.1 is 8.8 L^{-1} and 11 L^{-1} respectively, these runs produce lower mean values (Fig. S4a) and N_{isg95} (Fig. S4b) than CNTRL_INP0.1 with maximum N_{isg} 5.7 L^{-1} . This is partly due to the fact that a larger number of values in CNTRL_INP0.1 fall below the 0.005 L^{-1} threshold and is not included in the presented mean statistics. Nevertheless, it is obvious that the fewer primary ice crystals in all these sensitivity tests result in decreased frequency of ice-ice particle collisions, which is not sufficient to initiate BR multiplication. Increasing primary ice production by an order of magnitude in CNTRL_INP10 still results in underestimated ice concentrations than the observed, providing additional evidence for the significant role of SIP in these conditions. The increased concentration of primary ice crystals enhances BR efficiency in PHIL0.3_INP10 compared to PHIL0.3, however the produced mean concentrations still are lower than the observed; N_{isg95} in PHIL0.3_INP10 only slightly exceeds unity (Fig. S4b). In contrast, PHIL0.4_INP10 produces similar mean ICNCs with PHIL0.4, while N_{isg95} differs by a factor of ~ 3 throughout the examined temperature range (Fig. S4b). This suggests that increasing concentrations of available ice particles tend to decrease the efficiency of BR mechanism, hence possible overestimations in primary ice production are likely offset by decreasing production of secondary ice. In summary, the above results indicate that BR is not very sensitive to uncertainties in primary ice production, as long as there are enough primary ice crystals to initiate this process.

References:

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Figures:

Figure S1: Total ice number concentrations (ice+snow+graupel, N_{ice}) as a function of temperature for the case study (grey) and the eight model simulations. Median values, the 25th, 75th and 95th percentile are shown in panels (a), (b), (c) and (d), respectively (see Text S1). Modeled ice properties are calculated for particles $> 80 \mu\text{m}$ and for $N_{ice} > 0.005 \text{ L}^{-1}$ within the lowest 1.5 km a.s.l. Limits of y-axes differ between all panels.

Figure S2: Map of daily mean Cloud Radiative Forcing (CRF) anomalies at the surface averaged over the 1-km domain for the seven sensitivity simulations presented in the main text (see Text S2).

Figure S3: Total ice number concentrations (N_{ice}) for particles $> 80 \mu\text{m}$ as a function of temperature for our case study (grey) and the nine sensitivity simulations with varying treatment of the H-M process (see Text S3). Mean values and the 95th percentile are shown in panels (a) and (b), respectively. Limits of y-axes differ between panels (a) and (b).

Figure S4: Same as in Fig. S3 but for sensitivity simulations with varying INP conditions (see Text S4). Limits of y-axes differ between panels (a) and (b).