Secondary ice production processes in wintertime alpine mixed-phase clouds

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17 Abstract

18 Observations of orographic mixed-phase clouds (MPCs) have long shown that measured ice 19 crystal number concentrations (ICNCs) can exceed the concentration of ice nucleating particles 20 by orders of magnitude. Additionally, model simulations of alpine clouds are frequently found to underestimate the amount of ice compared with observations. Surface-based blowing snow, 21 22 hoar frost and secondary ice production processes have been suggested as potential causes, but 23 their relative importance and persistence remains highly uncertain. Here we study ice 24 production mechanisms in wintertime orographic MPCs observed during the Cloud and 25 Aerosol Characterization Experiment (CLACE) 2014 campaign at the Jungfraujoch site in the 26 Swiss Alps with the Weather Research and Forecasting model (WRF). Simulations suggest 27 that droplet shattering is not a significant source of ice crystals at this specific location - but 28 break-up upon collisions between ice particles is quite active, elevating the predicted ICNCs 29 by up to 3 orders of magnitude, which is consistent with observations. The initiation of the ice-30 ice collisional break-up mechanism is primarily associated with the occurrence of seeder-feeder 31 events from higher precipitating cloud layers. The enhanced aggregation of snowflakes is found 32 to drive secondary ice formation in the simulated clouds, the role of which is strengthened 33 when the large hydrometeors interact with the primary ice crystals formed in the feeder cloud. 34 Including a constant source of cloud ice crystals from blowing snow, through the action of the 35 break-up mechanism, can episodically enhance ICNCs. Increases in secondary ice fragment generation can be counterbalanced by enhanced orographic precipitation, which seems to 36 37 prevent explosive multiplication and cloud dissipation. These findings highlight the importance 38 of secondary ice and "seeding" mechanisms - primarily falling ice from above and to a lesser

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degree blowing ice from the surface – which frequently enhance primary ice and determine the
 phase state and properties of MPCs.

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42 1. Introduction

43 Understanding orographic precipitation is one of the most critical aspects of weather 44 forecasting in mountainous regions (Roe, 2005; Rotunno and Houze, 2007; Chow et al., 2013). 45 Orographic clouds are often mixed-phase clouds (MPCs), containing simultaneously supercooled liquid water droplets and ice crystals (Lloyd et al., 2015; Lohmann et al., 2016; 46 47 Henneberg et al., 2017). MPCs are persistent in complex mountainous terrain, because the high 48 updraft velocity conditions generate supercooled liquid droplets faster than can be depleted by 49 ice production mechanisms (Korolev and Isaac, 2003; Lohmann et al., 2016). In mid- and high-50 latitude environments almost all precipitation originates from the ice phase (Field and 51 Heymsfield, 2015; Mülmenstädt et al., 2015), highlightingemphasizing the 52 importancenecessity of correctly simulating the amount and distribution of both liquid water 53 and ice (i.e., the liquid-ice phase partitioning) in MPCs (Korolev et al., 2017).

54 Our understanding of MPCs remains incomplete owing to the numerous and highly 55 nonlinear cloud microphysical pathways driving their properties and evolution (Morrison et al., 56 2012). Due to the lower equilibrium water vapor pressure over ice crystals than over liquid 57 water, MPCs tend to glaciate over time through the Wegener-Bergeron-Findeisen (WBF) 58 process, which is the rapid ice crystal growth at the expense of the surrounding evaporating 59 cloud droplets (Bergeron, 1935; Findeisen, 1938). Another process that Ice crystals falling from a high-level seeder cloud into a lower-level cloud (external "seeder-feeder" event) or a lower-60 lying part of the same cloud (in-cloud "seeder-feeder" event) can trigger cloud glaciation and 61 62 has been shown to enhance precipitation over mountains is the seeder feeder mechanism (e.g., Roe, 2005). This mechanism has been observed in several field studies (e.g., Reinking et al., 63 64 2000; Purdy et al., 2005; Mott et al., 2014; Ramelli et al., 2021) and refers to ice crystals falling 65 from a high level seeder cloud into a lower level cloud (external seeder feeder event) or a 66 lower lying part. Analysis of the same cloud (in cloud seeder feeder event), where they act as 67 seeds for the glaciation of clouds. Satellite products covering satellite remote sensing over the 68 11-year period between April 2006 and October 2017 indicatedsuggest that seeding events are 69 widespread over Switzerland, occurring with a frequency of 31% of the total observations in 70 which cirrus clouds seed lower mixed-phase cloud layers (Proske et al., 2021). Despite these 71 two mechanisms that can readily destabilize an orographic cloud, a high frequency of MPCs

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have been reported under high updraft velocity conditions prevailing over the complex
 mountainous terrain (e.g., in the Swiss Alps), where supercooled liquid droplets are generated
 faster than depleted by depositional ice growth and riming, leading to persistent mixed phase
 conditions (Korolev and Isaac, 2003; Lohmann et al., 2016).

At temperatures between 0 °C and 38 °C, where mixed phase conditions can occur, 76 77 primary ice formation in clouds is catalyzed by the presence of insoluble aerosols that act as. 78 Primary ice formation in MPCs is catalyzed by the action of ice nucleating particles 79 (INPs) (e.g., Hoose and Möhler, 2012, Kanji et al., 2017). However, in-situ observations of 80 MPCs forming over mountain-top research stations or near mountain slopesin orographic 81 environments regularly reveal that there is a mismatch between the scarcity of primary INPs 82 and the measured ice crystal number concentrations (ICNCs) - the latter beingare several 83 orders of magnitude more abundant than INPs (Rogers and Vali, 1987; Geerts et al., 2015; 84 Lloyd et al., 2015; Beck et al., 2018; Lowenthal et al., 2019; Mignani et al., 2019). Model 85 simulations of alpine MPCs frequently fail to reproduce the elevated ICNCs dictated by 86 observations (Farrington et al., 2016; Henneberg et al., 2017; Dedekind et al., 2021).

87 The fact that inability of primary ice cannot explain to reproduce the observed ICNCs in 88 orographic MPCs has often been attributed to the influence of surface processes such as 89 theincluding lofting of snowflakes (i.e., "blowing snow;", Rogers and Vali, 1987; Geerts et al., 90 2015), detachment of surface hoar frost (Lloyd et al., 2015), turbulence near the mountain 91 surface or convergence of ice particles due to orographic lifting (Beck et al., 2018) and riming 92 on snow-covered surfaces (Rogers and Vali, 1987). The impact of blowing snow ice particles 93 (BIPS) has been studied thoroughly, either using observations collected in mountainous regions 94 (e.g., Lloyd et al., 2015; Beck et al., 2018; Lowenthal et al., 2019), remote sensing (e.g., Rogers 95 and Vali, 1987; Vali et al., 2012; Geerts et al., 2015) or detailed snow-cover models (e.g., 96 Lehning et al., 2006; Krinner et al., 2018) coupled with atmospheric models (e.g., Vionnet et 97 al., 2014; Sharma et al., 2021. The extent to which BIPS can affect ICNCs in MPCs remains 98 poorly understood. 99 Among these surface processes, the In-cloud secondary ice production (SIP) - or ice 100 multiplication - processes may also enhance ice production above the concentration of INPs

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 al., 2018; Lowenthal et al., 2019), and detailed surface snow models (e.g., Lehning et al., 2006;
 Vionnet et al., 2013, 2014) or through remote sensing techniques (e.g., Rogers and Vali, 1987;
 Vali et al., 2012; Geerts et al., 2015). BIPS are found to hover close to the surface provided

106 that the wind speed exceeds a threshold value, which varies between 4 and 13 ms⁻¹ (e.g., Déry 107 and Yau, 1999; Mahesh et al., 2003), depending on the snowpack properties and the prevailing 108 atmospheric conditions. The transport of BIPS is commonly separated into the saltation laver 109 and the turbulent suspension layer. The saltation layer is a shallow layer formed close to the 110 ground, where the transported ice particles are found to follow ballistic trajectories. Turbulent 111 eddies or upward gusts can then diffuse the saltated ice particles up to a height of several tens 112 of meters above the surface, into the suspension layer (e.g., Vali et al., 2012; Vionnet et al., 113 2014).

In cloud secondary ice production (SIP) processes may also enhance ice production after the initial primary ice nucleation events. Especially for orographic clouds, whose cloud top temperatures are not cold enough to activate sufficient INPs, ice multiplication through SIP might be particularly important. Over the past few decades, several SIP mechanisms have emerged in literature, a detailed review of which is provided by Field et al. (2017) and Korolev and Leisner-(_2020). We briefly review the three main SIPThree mechanisms below.

120 are thought to be responsible for most of the SIP. The rime splintering, also first, known 121 as the Hallett-Mossop (HM) process (Hallett and Mossop, 1974), is argued to be the most 122 efficient one in slightly supercooled clouds (i.e., temperatures warmer than 10 °C). The HM 123 process refers to the ejection of small secondary ice splinters after a supercooled droplet with 124 a diameter larger than $\sim 25 \,\mu m$ rimes onto a large ice particle at temperatures between -8 and -3 °C (Choularton et al., 1980; Heymsfield and Mossop, 1984). Although this is the only This 125 126 SIP mechanism is widely implemented in current microphysics schemesatmospheric models 127 (e.g., Beheng, 1987; Phillips et al., 2001; Morrison et al., 2005), recent modeling studies of 128 slightly supercooled polar clouds, have shown that it cannot sufficiently but cannot on its own 129 explain the enhanced ICNCs in remote environments (Young et al., 2019; Sotiropoulou et al., 130 2020, 2021a). Moreover, aircraft measurements have reported high ICNCs, especially for when 131 the conditions required for HM initiation are not fulfilled (e.g., Korolev et al., 2020).

132 A second process that is found to contribute to ice multiplication over a wider 133 temperature range is the collisional Collisional fracturing and breakup (BR), which involves the 134 fracturing) of delicate ice particles due to collisions with other ice particles (Vardiman, 1978; 135 Griggs and Choularton, 1986; Takahashi et al., 1995). Evidence for this process is provided 136 from several is another important SIP mechanism. Several field studies in the Arctic (Rangno 137 and Hobbs, 2001; Schwarzenboeck et al., 2009)-or in, the Alps (Mignani et al., 2019; Ramelli 138 et al., 2021) and from limited laboratory investigations (Vardiman 1978; Takahashi et al. 1995). 139 These 1995) all show the importance of BR. The latter two studies created athe basis for various Field Code Changed

140 numerical formulations of the BR mechanisma mechanistic description of BR (e.g., Phillips et 141 al., 2017a; Sullivan et al., 2018a; Sotiropoulou et al., 2020). Parameterizations of this 142 mechanism are BR have recently been implemented in small-scale-models (Fridlind et al., 2007; 143 Phillips et al., 2017a, b; Sotiropoulou et al., 2020, 2021b; Sullivan et al., 2018a; Yano and 144 Phillips, 2011; Yano et al., 2016), mesoscale models (Hoarau et al., 2018; Sullivan et al., 145 2018b; Qu et al., 2020; Sotiropoulou et al., 2021a; Dedekind et al., 2021) and global climate 146 models (Zhao and Liu, 2021a). These modeling studies followed several approaches to 147 implement the effect of BR. For instance, Hoarau et al. (2018) assumed a constant number of 148 fragments generated per collision in the Meso NH model, while Sullivan et al. (2018b) 149 implemented a temperature dependent relationship in the COSMO ART mesoscale model 150 based on the results of Takahashi et al. (1995). This simplified formulation was further 151 modified to account for the hydrometeor size scaling, which improved the representation of 152 ICNCs in alpine clouds (Dedekind et al., 2021). Sotiropoulou et al. (2020) and (2021a) 153 reproduced the observed ICNCs in polar clouds, by applying the physically-based 154 parameterization developed by Phillips et al. (2017a, b). At slightly colder temperatures 155 (between 12.5 °C and 7°C), however, BR was found to be generally weak over the Arctic 156 (Sotiropoulou et al., 2021b; Zhao et al., 2021), each with their own approach towards BR 157 description.

158 Droplet freezing and shattering (DS) during freezing is a third process that is frequently 159 suggested to explain the unexpected ice enhancement in clouds. This SIP mechanism that can 160 produce significant amounts of ice crystals. It occurs when a drizzle-sized droplet, with a size 161 drops (diameter larger than -exceeding 50 µm-collides) comes in contact with an ice particle 162 or INP, triggering its freezing after a. A solid ice shell is initially formed around the droplet 163 (e.g., Griggs and Choularton, 1983). As the freezing moves inward, the pressure starts to build 164 and the freezing droplet reacts by either, and as it thickens begins building up pressure that 165 leads to breakup in two halves, cracking, bubble burst or jetting (e.g., Keinert et al., 2020). 166 These processes may be accompanied by the ejection Ejection of small ice fragments may 167 occur, the number of which is yet poorly constrained as recent laboratory studies are showing 168 a large diversity of results yaries considerably (Lauber et al., 2018; Keinert et al., 2020; 169 Kleinheins et al., 2021; James et al., 2021). Individual experiments of freezing droplets 170 reported the maximum Experimentally, fragmentation rate maximizes at temperatures between 171 ~-10 and -15 °C (Leisner et al., 2014; Lauber et al., 2018; Keinert et al., 2020). DS is found 172 to be very efficientcan dominate in vigorous convective updrafts (Lawson et al., 2015; Phillips 173 et al., 2018; Korolev et al., 2020; Qu et al., 2020), while remote. Remote sensing observations

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174 indicate thatof warm Arctic MPCs suggest DS can be much more conducive to SIP in slightly 175 supercooled Arctic MPCs than the well-known HM process (Luke et al., 2021). This is in line 176 with singleSingle-column simulations performed by Zhao et al. (2021); support this, but 177 contradicts the findings of is in contrast with small-scale modeling studies suggesting that DS 178 is ineffective in polar regions (Fu et al., 2019; Sotiropoulou et al., 2020). Mesoscale model 179 simulations of winter alpine clouds formed at temperatures lower than -8 °C indicate that DS 180 is not contributing to the modeled ICNCsactive (Dedekind et al., 2021), while field 181 observations suggest the increasing efficiency of the mechanism at temperatures warmer than 182 -3 °C (Lauber et al., 2021).

183 In the orographic MPCs observedOrographic ICNCs in MPCs exceeded the predicted 184 INPs by 3 orders of magnitude, reaching up to ~1000 L^{-1} at -15 °C during the Cloud and 185 Aerosol Characterization Experiment (CLACE) 2014 campaign at the high altitude research 186 station of Jungfraujoch (JFJ) station in the Swiss Alps, the measured ICNCs exceeded the 187 predicted INPs by 3 orders of magnitude, reaching up to ~1000 L⁻¹ at temperatures around -15 188 [•]C (Lloyd et al. 2015). Whilst ice multiplication through Although the efficiency of BR and 189 DS mechanisms show a peak productionpeaks at around a similar the same temperature, Lloyd 190 et al. (2015) did not find evidence for their occurrence. Instead, they suggested that at periods 191 when there was a strong correlation between horizontal wind speed and observed ICNCs, ICNC 192 they suggested that BIPS is contributing to the latter, but the mechanism was incapable of 193 producing ICNCs higher than could not get ICNCs to exceed ~100 L^{-1} . In the absence of such 194 correlation, a flux of hoar frost crystals was considered responsible for the very high ice 195 concentration events (ICNCs > 100 L^{-1}), albeit without any direct evidence. Beck et al. (2018) 196 argued that the relationship between ICNCs and horizontal wind speed may not be a good 197 indicator for distinguishing between blowing snow and hoar frost. Their measurements 198 conducted at the Sonnblick Observatory in the Austrian Alps revealed the presence of several 199 hundred ice crystals of blowing snow per liter during cloud free conditions. In a cloudy 200 environment, though, such high contribution from BIPS was found only close to the surface, 201 with the concentrations dropping to several tens to 100 L⁴ at heights above ~10 m.

202From a modeling perspective, the causes of the surprisingly high ICNCs in orographic203MPCs formed during the CLACE 2014 campaign were explored in Farrington et al. (2016).204Since temperatures at JFJ are generally outside the HM temperature range (< 8°C), Farrington</td>205et al. (2016) used back trajectories analysis to investigate whether splinters produced at lower206altitudes through the HM process could be lifted to the summit of JFJ elevating the modeled207ICNCs. TheyFarrington et al. (2016) showed that the inclusion of the HM process upwind of

JFJ could not explain the measured concentrations of ice, while the addition of a surface fluxof hoar crystals provided the best agreement with observations.

210 Although surface-originated processes have been frequently invoked to explain the 211 disparity between ICNCs and INPs, the role of SIP processes - especially the BR and the DS 212 mechanism - has received muchfar less attention. In and is addressed in this study we. We 213 utilize the Weather Research and Forecasting model (WRF) to conduct simulations of two case 214 studies observed in winter during the CLACE 2014 campaign. Our primary objective is to 215 investigate if the implementation of two SIP parameterizations that account for the effect of 216 BR and DS can reduce the discrepancies between observed and simulated ICNCs. Additionally, 217 we aim to identify the conditions favoring the initiation of SIP in the orographic terrain and 218 explore the synergistic influence of SIP with wind-blown ice.

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220 **2. Methods**

221 2.1 CLACE instrumentation

222 CLACE is a long-established series of campaigns taking place for over two decades at the 223 mountain-top station of JFJ, located in the Bernese Alps, in Switzerland, at an altitude of ~3580 224 m above sea level (a.s.l.) (e.g., Choularton et al., 2008). The measurement area is very complex 225 and heterogeneous with distinct mountain peaks (Fig. 1), while JFJ is covered by clouds 226 approximately 40% of the time, offering an ideal location for microphysical observations 227 (Baltensperger et al., 1998). Owing to the local orography surrounding the site, the wind flow 228 is constrained to two directions (Ketterer et al., 2014). Under southeasterly (SE) wind 229 conditions, air masses are lifted along the moderate slope of the Aletsch Glacier, whereas under 230 northwesterly (NW) wind conditions the air is forced to rise faster along the steep north face 231 of the Alps, which is associated with persistent MPCs (Lohmann et al., 2016). A detailed 232 description of the in-situ and remote sensing measurements taken during January and February 233 2014 as part of the CLACE 2014 campaign is provided by Lloyd et al. (2015) and Grazioli et 234 al. (2015). Here we only offer a brief presentation of the datasets used in this study.

Shadowgraphs of cloud particles were produced by the two-dimensional stereo hydrometeor spectrometer (2D-S; Lawson et al., 2006), part of a three-view cloud particle imager (3V-CPI) instrument. The 2D-S products have been used to provide information on the number concentration and size distribution of particles in the size range of 10-1280 μm. Following Crosier et al. (2011), the raw data were further processed to determine betweendistinguish ice crystals andfrom droplets, and to remove. Removal of artefacts from 241 shattering events was also considered (Korolev et al., 2011)-, however analysis of the probe 242 imagery (Crosier et al., 2011) along with inter-arrival time histograms did not reveal the 243 presence of shattered particles, presumably because of the much lower velocity at which the 244 2D-S probe was aspirated (~15 ms⁻¹) compared to those during aircraft deployments (Lloyd et 245 al., 2015). An approximation of the ice water content (IWC) at JFJ could also be derived by 246 the 2D-S data using the Brown and Francis (1995) mass-diameter relationship with a factoran 247 uncertainty of up to 5 uncertaintytimes (Heymsfield et al., 2010). Additionally, the 248 quantification of the liquid water content (LWC) is based on the liquid droplet size distribution 249 data derived from a DMT cloud droplet probe (CDP; Lance et al., 2010) over the size range 250 between 2 and 50 µm. Typical meteorological Meteorological parameters (e.g., temperature, 251 relative humidity, wind speed and wind direction), that served as comparison to assess the 252 validity of the model, were provided by the weather station managed by MeteoSwiss at JFJ. 253 The instrumentation was set up on the roof terrace outside the Sphinx laboratory and used to 254 evaluate the model.

256 2.2 WRF simulations

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257 WRF-model, version 4.0.1, with augmented cloud microphysics to include the effects of all 258 SIP mechanisms (Sotiropoulou et al., 2021a) is used for non-hydrostatic cloud-resolving 259 simulations. The model has been run with three two-way nested domains (Fig. 1), with a 260 respective horizontal resolution of 12, 3 and 1 km. Two-way grid nesting is generally found to 261 improve the model performance in the inner domain (e.g., Harris and Durran, 2010), although 262 the sensitivity of the results to the applied nesting technique has been shown to be negligible 263 (not shown). The parent domain consists of 148×148 grid points centered over the JFJ station 264 (46.55°N, 7.98°E, shown with a black dot in Fig. 1), while the second and the third domain 265 include 241×241 and 304×304 grids, respectively. The Lambert conformal projection is 266 applied to all three domains, as it is well-suited for mid-latitudes. Here we adapted the so-called 267 "refined" vertical grid spacing proposed by Vignon et al. (2021), using 100 vertical eta levels 268up to a model top of 50 hPa (i.e., ~20 km). This set-up provides a refined vertical resolution of 269 ~100 m up to mid-troposphere at the expense of the coarsely resolved stratosphere. To 270 investigate the dynamical influence on the development of MPCs under the two distinct wind 271 regimes prevailing at JFJ (Section 2.1), we simulate two case studies, starting on 25 January 272 and 29 January 2014, 00:00 UTC, respectively. Both case studies are associated with the 273 passage of frontal systems over the region of interest, approaching the alpine slopes either from the NW (cold front) or the SE (warm front) direction, as shown by the vertically-integrated condensed water content (ICWC; sum of cloud droplets, rain, cloud ice, snow, and graupel) in Fig. 1. For both cases the simulation covers a 3-day period, with the first 24 hours being considered sufficient time for spin up. A 27-s time step was used in the parent domain and goes down to 9 s in the second domain and 3 s in the third domain. Note that achieving such small time steps in the innermost domain is essential to ensure numerical stability in non-hydrostatic simulations over a region with complex orography such as around JFJ.

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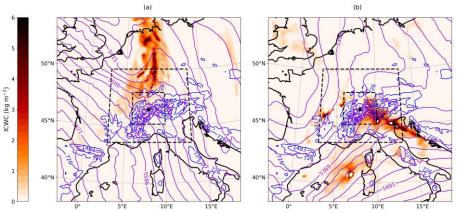


Figure 1. Map of synoptic conditions around JFJ station at (a) 00:00 UTC, 26 January 2014 and (b) 00:00 UTC, 30 January 2014, from the control simulation (12-km resolution domain). The purple (blue) contours show the 500 hPa geopotential height in m (the terrain heights in m). The color shading shows the vertically-integrated condensed water content (in kg m⁻²). The black dashed lines delimit the 3-km and 1-km resolution domains, while the black dot locates the JFJ station.

290 The ERA5 reanalysis The fifth generation of the European Centre for Medium-Range 291 Weather Forecasts (ECMWF) atmospheric reanalyses dataset (ERA5; Hersbach et al., 2020) is 292 used to initialize the model and provide the lateral forcing at the edge of the 12-km resolution 293 domain every 6 hours. Static fields at each model grid point come from default WRF pre-294 processing system datasets, with a resolution of 30" for both the topography and the 4 and 295 use'use fields. The MODIS-Land use categories are based dataset is used foron the Moderate 296 Resolution Imaging Spectroradiometer (MODIS) land-cover classification. Regarding the 297 physics options chosen to run WRF simulations, the Rapid Radiative Transfer Model for 298 general circulation models (RRTMG) radiation scheme is applied to parameterize both the 299 short-wave and long-wave radiative transfer. The vertical turbulent mixing is treated with the

Mellor-Yamada-Janjic (MYJ; Janjić, 2002) 1.5 order scheme, while surface options are modeled by the Noah land-surface model (Noah LSM; Chen and Dudhia, 2001). The Kain-Fritsch cumulus parameterization has been activated only in the outermost domain, as the resolution of the two nested domains is sufficient to reasonably resolve cumulus-type clouds at grid-scale.

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306 2.2.1 Microphysics scheme and primary ice production

The Morrison two-moment scheme (Morrison et al., 2005; hereafter M05) is used to parameterize the cloud microphysics, following the alpine cloud study of Farrington et al. (2016). The scheme includes double-moment representations of rain, cloud ice, snow and graupel species, while cloud droplets are treated with a single-moment approach and therefore the <u>cloud</u> droplet number concentration (N_d) must be prescribed. Here N_d is set to 100 cm⁻³, based on the mean N_d observed within the simulated temperature range (Lloyd et al., 2015).

313 Three primary ice production mechanisms through heterogeneous nucleation are 314 described in the default version of the M05 scheme, namely immersion freezing, contact 315 freezing, and deposition/condensation freezing nucleation. Immersion freezing of cloud 316 droplets and raindrops is described by the probabilistic approach of Bigg (1953). Contact 317 freezing is parameterized following Meyers et al. (1992). Finally, deposition and condensation 318 freezing is represented by the temperature-dependent equation derived by Rasmussen et al. 319 (2002) based on the in-situ measurements of Cooper (1986) collected from different locations 320 at different temperatures. Following Thompson et al. (2004), this parameterization is activated 321 either when there is saturation with respect to liquid water and the simulated temperatures are 322 below -8 °C or when the saturation ratio with respect to ice exceeds a value of 1.08. The 323 accuracy of these parameterizations in representing atmospheric INPs is debatable as they are 324 derived from very localized measurements over a limited temperature range. Nevertheless, 325 Farrington et al. (2016) argued that the deposition/ condensation freezing parameterization of Cooper (1986) can effectively explain INPs between the range 0.01 and 10 L⁻¹, which is 326 327 frequently observed during field campaigns at JFJ (Chou et al., 2011; Conen et al., 2015).

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329 2.2.2 Ice multiplication through rime splintering in the M05 scheme

330 Apart from primary ice production, the HM process is the only SIP mechanism included in the

- default version of the M05 scheme. This parameterization adapted from Reisner et al. (1998),
- based on the laboratory findings of Hallett and Mossop (1974), allows for -splinter production

333 after cloud- or rain- drops are collected by rimed snow particles or graupels graupel. The 334 efficiency of this process is zero outside the temperature range between -8 and -3 °C, while it 335 follows a linear temperature-dependent relationship in between. HM is not activated unless the rimed ice particles have masses larger than 0.1 g kg⁻¹ and cloud or rain mass mixing ratio 336 exceeds the value of 0.5 g kg⁻¹ or 0.1 g kg⁻¹, respectively. Since these conditions are rarely met 337 338 in natural MPCs, previous modeling studies had to artificially remove any thresholds to achieve 339 an enhanced efficiency of this process (Young et al., 2019; Atlas et al., 2020). In the current 340 study, however, the HM process is not effective, as the simulated temperatures at JFJ altitude 341 are below -8 °C (see Sect. 2.3).

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343 2.2.3 Ice multiplication through ice-ice collisions in the M05 scheme

344 In addition to the HM process, we have also included two parameterizations to represent the 345 BR mechanism. An extensive description of the implementation method is provided in 346 Sotiropoulou et al. (2021a) (see their Appendix B). Among the three ice particle types included 347 in the M05 scheme (i.e., cloud ice, snow, graupel), we assume that only the collisions between 348 cloud ice-snow, cloud ice-graupel, graupel-snow, snow-snow, and graupel-graupel can result 349 in ice multiplication. The first parameterization tested here follows the simplified methodology 350 proposed by Sullivan et al. (2018a), which is based on the laboratory work of Takahashi et al. 351 (1995). Their findings revealed a strong temperature dependence of the fragment numbers 352 generated per collision (N_{BR}) :

$$N_{BR} = 280 \left(T - T_{min}\right)^{1.2} e^{-(T - T_{min})/5},$$
(1)

where $T_{min} = 252 \ K$, is the minimum temperature for which BR occurs. Yet their experimental set-up was rather simplified involving only collisions between large hail-sized ice spheres with diameters of ~2 cm. Taking this into account, Sotiropoulou et al. (2021a) further scaled the temperature-dependent formulation for size:

$$N_{BR} = 280 \left(T - T_{min}\right)^{1.2} e^{-(T - T_{min})/5} \frac{D}{D_0},$$
(2)

where *D* is the size in meters of the particle that undergoes break-up and
$$D_0=0.02$$
 m is the size
of the hail-sized balls used in the experiments of Takahashi et al. (1995).

Phillips et al. (2017a) proposed a more physically-based formulation, developing an energy-based interpretation of the experimental results conducted by Vardiman (1978) and Takahashi et al. (1995). The initial collisional kinetic energy is considered as the governing constraint driving the BR process. Moreover, the predicted *N_{BR}* depends on the ice particle type and morphological habit and is a function of the temperature, particle size and rimed fraction.Here the generated fragments per collision are described as follows:

$$N_{BR} = aA \left(1 - exp \left\{ - \left[\frac{CK_0}{aA} \right]^{\gamma} \right\} \right), \tag{3}$$

where $K_0 = \frac{1}{2} \frac{m_1 m_2}{m_1 + m_2} (\Delta u_{n12})^2$ is the initial kinetic energy, in which m_1 and m_2 are the masses 365 of the colliding particles and $|\Delta u_{n12}| = \{(1.7 u_{n1} - u_{n2})^2 + 0.3 u_{n1} u_{n2}\}^{1/2}$ is the difference 366 in their terminal velocities. The correction term is proposed by Mizuno et al. (1990) and Reisner 367 368 et al. (1998) to account for underestimates when $u_{n1} \approx u_{n2}$. The parameter *a* in Eq. 3 is the surface area of the smaller ice particle (or the one with the lower density), defined as $a = \pi D^2$, 369 370 with D as in Eq. 2. A in Eq. 3 represents the number density of breakable asperities on the 371 colliding surfaces. For collisions that involve cloud ice and snow particles A is described as $A = 1.58 \times 10^{7} (1 + 100 \Psi^{2}) (1 + \frac{1.33 \times 10^{-4}}{D^{1.5}})$, where $\Psi < 0.5$ is the rimed fraction of the most 372 373 fragile ice particle. For graupel-graupel collisions A is given by a temperature-dependent equation as $A = \frac{a_0}{3} + \max\left(\frac{2a_0}{3} - \frac{a_0}{9}\right|T - 258|, 0)$, in which $a_0 = 3.78 \times 10^4 \left(1 + \frac{0.0079}{D^{1.5}}\right)$. C 374 375 is the asperity-fragility coefficient, which is empirically derived to account for different 376 collision types, while the exponent γ is equal to 0.3 for collisions between graupel-graupel and 377 is calculated as a function of the rimed fraction for collisions including cloud ice and snow. 378 The parameterization was developed based on particles with diameters 500 μ m < D < 5 mm, 379 however Phillips et al. (2017a) suggest that it can be used for particle sizes outside the 380 recommended range as long as the input variables to the scheme are set to the nearest limit of 381 the range. Finally, since N_{BR} was never observed to exceed 100 in the experiments of Vardiman 382 (1978), here we also use this value as an upper limit for all collision types (Phillips et al., 383 2017a). All predicted fragments emitted through BR are added to the cloud ice category.

384

385 2.2.4 Ice multiplication through droplet shattering in the M05 scheme

Two different parameterizations are implemented in the M05 scheme to investigate the potential efficiency of the DS mechanism in producing secondary ice splinters (N_{DS}). Phillips et al. (2018) proposed two possible modes of raindrop-ice collisions, that can initiate the freezing process. In the first mode, the freezing of the drop occurs either by collecting a small ice particle or through heterogeneous freezing. In the default M05 scheme, the product of the collisions between raindrops and cloud ice is considered to be graupel (snow) – if the rain mixing ratio is greater (lower) than 0.1 g kg⁻¹, following Reisner et al. (1998). Additionally, the heterogeneous freezing of big raindrops in the immersion mode follows Bigg's (1953) parameterization (Section 2.2.1). Here we consider that the product of these two processes can undergo shattering and generate numerous ice fragments, the number of which is parameterized after Phillips et al. (2018). The formulation is derived by fitting to a pooled dataset frommultiple laboratory studies and is given as<u>datasets to</u> a Lorentzian function of temperature and a polynomial expression of the drop size. More precisely, in the first mode of the formulation, the total number of fragments (*N*) generated per frozen drop are given by:

$$N = \Xi(D_r)\Omega(T) \left[\frac{\zeta \eta^2}{(T - T_0)^2 + \eta^2} + \beta T \right],\tag{4}$$

400 where T is the temperature (in K) and D_r is the size of the freezing raindrop (in mm). Note that 401 *N* is defined as the sum of the big fragments (N_B) and tiny splinters (N_T). Equation (4) applies 402 only to drop diameters less than 1.6 mm, which is the maximum observed experimentally. For 403 droplet sizes beyond this maximum value, *N* can be inferred by linear extrapolation. N_B is 404 described by another Lorentzian:

$$N_B = \min\left\{\Xi(D_r)\Omega(T)\left[\frac{\zeta_B\eta_B^2}{\left(T-T_{B,0}\right)^2+\eta_B^2}\right], N\right\}.$$
(5)

The factors $\Xi(D_r)$ and $\Omega(T)$ in Eq. (4) and (5) are cubic interpolation functions, preventing the onset of impeding DS for $D_r < 0.05$ mm and T > -3 °C. Furthermore, the parameters ζ , η , T₀, β , ζ_B , η_B , T_{B,0}, found in these relationships, are derived from previous laboratory studies and are analytically described in Phillips et al. (2018). Note that That the big fragments emitted (i.e., N_B) will be initiated in the model as graupel, snow or frozen drops, while it is only the tiny splinters ($N_T = N - N_B$) that are considered secondary ice (i.e., $N_{DS} = N_T$) and will be are passed to the cloud ice category.

412 The second mode of raindrop-ice collisions includes the accretion of raindrops on impact 413 with more massive ice particles, such as snow or graupel, the description of which in the M05 414 scheme is adapted from Ikawa and Saito (1991). This mode has been studied only once in the 415 laboratoryWhile there is only one dedicated laboratory study of this SIP mode (James et al., 416 2021), it was also indirectly investigated in the experimental study of Latham and Warwicker 417 (1980), who reported that the collision of supercooled raindrops with hailstones can potentially 418 stimulategenerate secondary ice. Since there was no quantitative observation of this mode, 419 Phillips et al. (2018) proposed an empirical, energy-based formulation to account for the tiny 420 splinters ejected after collisions between raindrops and large ice particles:

$$N_{DS} = 3\Phi(T) \times [1 - f(T)] \times max(DE - DE_{crit}, 0) , \qquad (6)$$

where $DE = \frac{K_0}{S_e}$, is the dimensionless energy given as the ratio of the initial kinetic energy (K_0 ; 421 described in 2.2.3) over the surface energy, which is expressed by the product $S_e = \gamma_{liq} \pi D_r^2$, 422 423 in which $\gamma_{liq}=0.073$ J m⁻², is the surface tension of liquid water. The critical value of DE used 424 in Eq. (6) for the onset of splashing upon impact is set to $DE_{crit} = 0.2$. The parameter f(T) =425 $-c_w T/L_f$, represents the initial frozen fraction of a supercooled drop during the first stage of 426 the freezing process, where $C_w = 4200 \text{ J kg}^{-1} \text{ K}^{-1}$, is the specific heat capacity of liquid water, 427 $L_f = 3.3 \times 10^5$ J kg⁻¹, is the specific latent heat of freezing, while T is the initial freezing 428 temperature (°C) of the raindrop. Finally, $\Phi(T) = \min[4f(T), 1]$ is an empirical fraction, which 429 represents the probability of any new drop in the splash products to contain a frost secondary ice particle. At temperatures ~ -10 °C this formulation yields $\Phi = 0.5$, meaning that the 430 431 probability of a secondary drop to contain ice is 50%. The first laboratory investigation of this 432 rather uncertain parameter as a function of temperature is provided in James et al. (2021)-433 provided the first laboratory study to constrain this parameter. Further details regarding the 434 derivation of the empirical parameters and the uncertainties underlying the mathematical 435 formulations are discussed in Phillips et al. (2018).

Following Sullivan et al. (2018a), the <u>The</u> second DS parameterization tested in this study was developed by Sullivan et al. (2018a) and is described as the product of a polynomial expression function of the freezing droplet size (Lawson et al., 2015 diameter (D_r in μ m), a shattering probability (p_{sh}) and a freezing probability (p_{fr}):

 $N_{DS} = 2.5 \times 10^{-11} \left(\frac{D_{\#}}{4} \right)^4 D_r^4 p_{sh} p_{fr}$

440 The diameter dependence describing the fragment numbers generated per fractured frozen 441 droplet is derived by nudging the liquid water and ice particle size distributions in one-442 dimensional cloud model simulations towards aircraft observations collected in tropical 443 cumulus clouds (Lawson et al., 2015). The psh is based upon droplet levitation experiments 444 shown in Leisner et al. (2014) and is represented by a temperature-dependent Gaussian 445 distribution, centered at ~ -15 °C. Note that p_{sh} is non-zero only for droplets with sizes greater 446 than 50 μ m. The p_{fr} is 0 for temperatures warmer than -3 °C and 1 if temperatures fall below – 447 6 °C, following the cubic interpolation function, $\Omega(T)$, adapted from Phillips et al. (2018). 448

449 2.3 Model validation

450 The control simulation (CNTRL), performed with the standard M05 scheme, sets the basis for 451 assessing the validity of the model against available meteorological observations. Temperature,

(7)

452 relative humidity, wind speed, and wind direction are obtained from the MeteoSwiss weather 453 station at JFJ. The comparison of each meteorological variable with the results from the nearest 454 model grid point of the CNTRL simulation is shown in Fig. 2. Note that the outputs are from the first atmospheric level of the innermost domain at ~10 m above ground level (a.g.l) (Fig. 455 456 1), while the first 24 hours of each simulation period are considered spin up time and are 457 therefore excluded from the present analysis. The mean modeled values and standard 458 deviations (std), along with the root mean square error (RMSE) and the index of agreement 459 (IoA) between model predictions and observational data are summarized in Table 1. IoA is 460 both a relative and a bounded measure (i.e., $0 \le IoA \le 1$) that describes phase errors between 461 predicted (P_i) and observed (O_i) time series (Willmott et al., 2012):

$$IoA = 1 - \left[\frac{\sum_{i=1}^{N} (P_i - O_i)^2}{\sum_{i=1}^{N} (|P_i'| - |O_i'|)^2}\right],$$
(8)

462 where $P'_i = P_i - \overline{O}$ and $O'_i = O_i - \overline{O}$, in which \overline{O} is the mean of the observed variable. 463

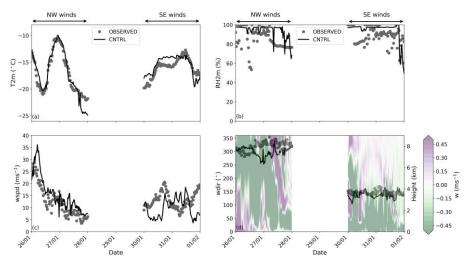


Figure 2. Time series of (a) temperature (T2m), (b) relative humidity with respect to liquid phase at 2m height (RH2m), (c) wind speed (wspd) and (d) wind direction (wdir). Grey circles indicate measurements collected between 26 January and 1 February 2014 at JFJ station, while modeled values from CNTRL simulation are shown with a black line. The semi-transparent contour plot is representing the vertical velocity (w) profile predicted by the CNTRL simulation. Each day starts at 00:00 UTC.

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464

Throughout the two case studies, the WRF simulations seem to closely follow the observed temperatures (Fig. 2a), which is also indicated by the high IoA in Table 1. The 474 synoptic situation occurring on 26 January, with a deep trough extending to western Europe 475 (Fig. 1), has been associated with intense snowfalls in the alpine regions (Panziera and Hoskins, 476 2008). The passage of the cold front was followed by a sharp temperature decrease, with the 477 simulated temperatures fluctuating between -10 and ~ -20 °C throughout the NW case (Fig. 478 2a). Under the influence of the warm front during the SE case, the modeled temperatures rose 479 from ~ -18 °C to ~ -14 °C and remained less variable until 30 January 12:00 UTC, with mean 480 values of ~ -15.5 °C (Table 1).

481

Table 1. Mean modeled values (± standard deviations), RMSE and IoA between the CNTRL
 simulation of WRF and measurements carried out by the MeteoSwiss station at JFJ.

Variable	Mean ± std		able Mean ± std F		RN	ISE	I	oA
	NW winds	SE winds	NW winds	SE winds	NW winds	SE winds		
T2m (°C)	-17.10 ± 4.36	-15.48 ± 1.75	1.40	1.33	0.97	0.84		
RH2m (%)	94.07 ± 7.02	94.24 ± 10.31	14.01	11.61	0.55	0.64		
wspd (ms ⁻¹)	15.57 ± 7.45	9.78 ± 3.94	4.85	6.75	0.88	0.22		

484

485 Fig. 2c and 2d reveal that the 1-km resolution domain can sufficiently capture the local 486 wind systems to a certain extent. During the NW flow, the horizontal wind speeds are 487 reproduced better by the CNTRL simulation (IoA=88%), whereas during the SE winds, the 488 simulated wind speed is frequently underestimated compared with observations (Fig. 2c). Such 489 deviations in the horizontal wind speed might be caused by the relatively coarse horizontal 490 resolution of the model, which prevents some small-scale and very local orographic structures 491 from being resolved. As discussed in Section 2.2, the observed winds at JFJ are channeled by 492 the orography to either NW or SE directions. The CNTRL simulation of WRF can satisfactorily 493 reproduce the wind direction in both cases, although the simulated values exhibit larger 494 fluctuations than the measured ones (Fig. 2d), presumably because of the surrounding 495 orography being less accurately represented in the model. This is particularly evident during 496 NW winds, when the simulated wind directions shift slightly to west directions compared to 497 observations. The positive vertical velocities, illustrated in the contour plot in Fig. 2d, result 498 from the orographically forced lifting of the airmasses over the local topography, and are not 499 related to convective instability in the lower atmospheric levels. The stronger updrafts 500 prevailing until the end of 26 January are associated with the steep ascent of the air parcels,

which can also contribute to the enhanced relative humidity (Fig. 2b). After the frontal passage, the vertical velocities at the lower levels are downward directed, with the vertical profile of potential temperature revealing that the atmosphere at JFJ is stabilized (not shown). The same vertical velocity pattern, with mainly downward motions, characterizes the stably stratified atmosphere after 30 January. Overall, Fig. 2 suggests that local meteorological conditions at JFJ are reasonably well represented by the model.

507

508 2.4 Model simulations

509 Given the good representation of the atmospheric conditions at JFJ, the CNTRL simulation of 510 WRF is further accompanied by <u>fourfive</u> sensitivity simulations, aiming to investigate the 511 contribution of BR and DS mechanisms. Here we also perform three additional sensitivity 512 experiments to explore the potential impact of blowing ice and the synergistic interaction with 513 SIP on the development of the simulated MPCs. A detailed list of the sensitivity experiments 514 is provided in Table 2.

515 The contribution of the DS mechanism is addressed in two sensitivity experiments, 516 DS_PHILL and DS_SULL, where the parameterizations of Phillips et al. (2018) and Sullivan 517 et al. (2018a) were applied, respectively (Section 2.2.4). Both sensitivity simulations yield 518 predictions that coincide with the CNTRL simulation (supplement Fig. S1) suggesting that the 519 DS mechanism is hardly ever activated, and fail to produce realistic total ice number 520 concentrations (N_{isg} ; cloud ice + snow + graupel). The absence of correlation between LWC 521 and N_{isg} fluctuations might also suggest the ineffectiveness of this mechanism under the 522 examined conditions. Note that the parameterized expressions used to describe the DS 523 mechanism involve a number of empirical and rather uncertain parameters, the value of which 524 could potentially influence the efficiency of the process in producing secondary ice fragments. 525 However, the sensitivity of our results to the choice of these parameters would be negligible, 526 as the low concentrations ($\lesssim 10^{-2}$ cm⁻³) of relatively small raindrops with mode diameters 527 below the threshold size of 50 µm seem to completely prevent the onset of the DS process 528 (supplement Fig. S2). The DS mechanism is therefore excluded from the following discussion. 529 This result is in line with the modeling study of Dedekind et al. (2021), who also 530 highlighted<u>reported</u> the inefficiency of this mechanism in wintertime alpine clouds.

531Two additional Three sensitivity simulations are also conducted to investigate if activating532the BR mechanism can account for the observed ICNCs.generation of secondarily formed ice

533 <u>particles through BR.</u> First, the <u>TAKAH simulation, adopts the</u> temperature-dependent formula

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534 of Takahashi et al. (1995) scaled with the size of the-particles that undergo fragmentation 535 (Sotiropoulou et al. 2021a) is tested in the TAKAH simulation. The PHILL simulation is then conducted to test the performance of the more advanced Phillips et al. (2017a) 536 537 parameterization. Note that the parameters involved in the parameterized expression of N_{BR} 538 (Eq. 3) concern the effect of ice habit and rimed fraction of the colliding). Applying Equation 539 (2) to collisions between all ice particles which is not explicitly resolved categories considered 540 in the M05 scheme. Regarding the ice habit, we assume spatial planar (except collisions between cloud-ice particles, based on the imagery presented in Lloyd et al. (2015) and; Section 541 542 2.2.3) inserts -Grazieli et al. (2015), which revealed the predominance of sectored plates and 543 oblate particles (probably columns or needles), along with some rimed hydrometeors, at 544 temperatures ~ 15 °C. Balloon-borne measurements taken in low level orographic MPCs 545 within seeder feeder events revealed the presence of a large fraction of graupel and rimed 546 particles (Ramelli et al., 2021). For this reason, during the NW and SE cases we consider rimed 547 fractions of 0.4 and 0.3 to account for heavily and moderately rimed ice particles, respectively. 548 A higher rimed fraction is prescribed for the NW-winds case study though, as the co-existence 549 of ice crystals and liquid droplets under the stronger updraft conditions are expected to 550 favor caveat to our approach. The laboratory results of Takahashi et al. (1995) suggest that, it 551 is mostly the collisions between rimed particles and graupel that are more conducive to SIP 552 through BR. Vardiman (1978) also reported that ice crystal growth through riming- However, 553 the is essential to boost fragmentation. Applying the Takahashi break-up scheme for unrimed 554 ice particles might, therefore, overestimate the number of secondary ice fragments. To test this 555 hypothesis, we performed TAKAHrim sensitivity of our results to the rimed fraction was not 556 found significantsimulation, where we enabled ice multiplication through BR only after 557 collisions between rimed cloud ice/ snow and graupel particles. To diagnose the presence of 558 rime on ice particles we used the amount of cloud droplets or raindrops accreted by snow and 559 cloud ice, which is predicted in the M05 scheme.

560

561 **Table 2.** List of sensitivity simulations conducted with the WRF-model.

Simulation	BR process	DS process	NBIPS (L-1)	•
CNTRL	off	off	0	+
DS_PHILL	off	Phillips et al., 2018	0	+
DS_SULL	off	Sullivan et al., 2018a	0	+
TAKAH	Takahashi et al., 1995	off	0	+

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<u>TAKAHrim</u>	<u>Takahashi et al., 1995</u> <u>activated only after</u> <u>collisions between rimed</u> <u>ice particles</u>	<u>off</u>	<u>0</u>
PHILL	Phillips et al., 2017a	off	0
BIPS10	off	off	10
BIPS100	off	off	100
BIPS100_PHILL	Phillips et al., 2017a	off	100

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563 Finally, the performance of the more advanced Phillips et al. (2017a) parameterization is 564 tested in the PHILL simulation. Parameters involved in the Phillips parameterization that are 565 not explicitly resolved in the M05 microphysics scheme are the rimed fraction and the ice habit 566 of colliding ice particles. The choice of ice habit is based on particle images collected during 567 the CLACE 2014 campaign, showing the presence of non-dendritic sectored plates and oblate 568 particles at temperatures ~ -15 °C (Lloyd et al., 2015). Grazioli et al. (2015) also presented 569 some examples of particle imagery produced by a 2D-S imaging probe, revealing the presence 570 of heavily rimed hydrometeors, as well as highly oblate particles (probably columns or 571 needles). The rimed fraction, is prescribed to a value of 0.4 (0.3) to account, respectively, for 572 heavily and moderately rimed ice particles present under NW (SE) wind conditions. A high 573 degree of riming is expected in the simulated cases, as they both occur under ice-seeding 574 situations (Section 3.1.1), where large precipitating ice particles from the seeder clouds 575 effectively gain mass in the mixed-phase zone through riming. Direct observations with 576 balloon-borne measurements carried out within ice-seeding events in the region around Davos 577 in the Swiss Alps support the presence of a large fraction of rimed particles and graupel 578 (Ramelli et al., 2021). The higher riming degree is prescribed under NW-winds because the 579 orographic forcing (i.e., vertical velocity) is stronger and helps maintaining mixed-phase 580 conditions in the feeder clouds - which in turn promotes ice crystal growth through riming. 581 However, our results were not very sensitive to the choice of the rimed fraction. 582 The remaining sensitivity simulations focus on the potential impact of BIPS. A recently-583 developed blowing snow scheme, used to simulate alpine snowpacks, reported significant mass 584 and number mixing ratios of BIPS that can be found up to ~1 km above the surface under high

wind speed conditions (see Fig. 17 in Sharma et al., 2021) with the potential to trigger cloud
 microphysical processes. Given that in the default M05 scheme there is no parameterization of
 a flux of ice particles from the surface, we parameterize the effect of BIPS lofting into the

simulated orographic clouds by applying a constant ice crystal source to the first atmospheric

level of WRF over the whole model domain. Although the source of BIPS at the first model level remained constant, yet their number will be affected by processes such as advection, sublimation and sedimentation, that are described in the M05 scheme. Note that the relatively coarse horizontal resolution in the innermost domain of our simulations (i.e., 1 km) does not allow the accurate representation of the small-scale turbulent flow over the orographic terrain. This is considered a limitation of our methodology, since turbulent diffusion is a key process affecting the amount of BIPS that will be resuspended from the surface.

The applied concentrations of BIPS varied between 10⁻² and 100 L⁻¹, which is the upper 596 597 limit proposed by Lloyd et al. (2015) and observed within in-cloud conditions by Beck et al. 598 (2018). Number concentrations of BIPS (i.e., NBIPS) lower than 10 L⁻¹ were found incapable 599 of affecting the simulated cloud properties and are, therefore, not included in the following 600 discussion. Two sensitivity simulations are finally performed, BIPS10 and BIPS100 (Table 2), in which the number indicates the NBIPS in L⁻¹. In our approach we assume BIPS are spherical 601 602 with diameters of 100 µm, based on typical sizes that are frequently reported in the literature 603 (e.g., Schlenczek et al., 2014; Schmidt, 1984; Geerts et al., 2015; Sharma et al., 2021). The 604 relatively small fall speed of these particles (e.g., Pruppacher and Klett, 1997) will allow them 605 to remain suspended in the atmosphere. As a sensitivity we also considered smaller particles 606 with sizes of 10 µm, but our results did not change significantly (supplement Fig. S3). 607 BesidesNevertheless, such small ice particles are not expected to substantially contribute to the 608 simulated IWC, as shown by Farrington et al. (2016).

As SIP through BR and blowing snow are both important when trying to explain the high ICNCs observed in alpine environments, their combined effect is addressed in our last simulation, BIPS100_PHILL (Table 2). In this sensitivity simulation the effect of BR is parameterized after Phillips et al. (2017a), while a constant ice crystal concentration of 100 L⁻ ¹ is applied to the first atmospheric level of WRF to represent the effect of BIPS.

614

615 3. Results and discussion

- 616
- 617 3.1 Impact of SIP through BR on simulated microphysical properties
- Fig. 3 displays the The temporal evolution of the N_{isg} , IWC and LWC, at the first model level
- 619 (~ 10 m a.g.l.) from the nearest to JFJ model grid point of the CNTRL, TAKAH and PHILL
- 620 simulations. Note that instead is presented in Fig. 3. Instead of focusing on a single grid point,
- 621 we <u>also</u> averaged the results from the 9-km² area surrounding the point of interest. However,

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622 the produced time series showed only little difference when compared to the nearest grid point 623 time series, further validating the robustness of our results (not shown)., ensuring our analysis 624 is robust. Besides, the region in the vicinity of JFJ is very heterogeneous supporting the single 625 point comparison presented in the following discussion. The grey dots shown in Fig. 3 represent the measurements taken by the 2D-S and CDP instruments at JFJ throughout the two 626 627 periods of interest. The displayed time frequency of the observations is 30 min to match the 628 output frequency of the model. Note that the simulated LWC includes liquid water from cloud 629 droplets and rain, while the simulated IWC includes cloud ice, snow and graupel. The 630 contribution of rain in our simulations is, however, negligible (supplement Fig. S2). Several 631 statistical metrics for Nisg, IWC and LWC are summarized in Table 3, 4 and 5, respectively. 632 Note that periods Periods with missing data in the measurement time series are excluded from 633 the statistical analysis.

634 During the NW flow, between 26 and 28 January, the measured ICNCs exceed 100 L⁻¹ 635 for >50 % of the time, whereas during the SE flow the ICNCs usually fluctuate between 10 and 636 100 L⁻¹ (Fig. 3a). The highest ICNCs are generally observed at temperatures higher than ~ -15 637 °C, where SIP processes are thought to be dominant and primary ice nucleation in the absence 638 of bioaerosols is limited (e.g., Hoose and Möhler, 2012; Kanji et al., 2017). The CNTRL simulation fails to reproduce N_{isg} higher than 10 L⁻¹, with the mean simulated values being ~2-639 640 2.5 L⁻¹ during both periods. At the same time the mean observed ICNC values are ~200 (70) 641 L⁻¹ during the NW (SE) case. Thus, CNTRL systematically underestimates the amount of ice 642 by up to 2 orders of magnitude, which is also consistent with the interquartile statistics 643 presented in Table 3. With the HM process being totally ineffective in the prevailing 644 temperatures, this discrepancy suggests that ice crystals produced by heterogeneous ice 645 nucleation in CNTRL are not high enough to match the observations. A similar discrepancy between predicted INPs and measured ICNCs was also documented in Lloyd et al. (2015). 646

Table 3. The 25^{th} , 50^{th} (median) and 75^{th} percentiles of ICNC time series (in L⁻¹).

Simulation	25 th perc.		Mee	Median		75 th perc.	
	NW winds	SE winds	NW winds	SE winds	NW winds	SE winds	
OBSERVED	8.69	6.64	80.47	34.53	261.25	88.69	
CNTRL	0.76	0.84	<u>2.02</u> 1.68	<u>1.682.02</u>	2.80	3.60	
TAKAH	2.27	1.08	9.85	122.56	362.51	358.38	
TAKAHrim	2.56	0.75	12.30	4.17	<u>101.46</u>	<u>82.16</u>	

PHILL	2.49	0.76	6.27	2.09	118.21	59.23
BIPS10	1.60	1.90	2.42	2.72	3.30	4.78
BIPS100	6.17	10.74	10.36	13.88	12.32	17.39
BIPS100_PHILL	8.95	11.51	15.87	16.30	138.92	98.43

650 Activating the BR process in TAKAH, TAKAHrim and PHILL simulations is found to produce N_{isg} as high as 400 L⁻¹ during both case studies (Fig. 3a), resulting in a substantially 651 652 better agreement with observations. At times when the simulated temperatures drop below ~ -18 °C, the N_{isg} modeled by bothall three simulations coincide with the CNTRL simulation. At 653 654 relatively warmer subzero temperatures though, the significant contribution of the BR process 655 is evident, elevating the predicted Nisg by up to 3 orders of magnitude during the NW case and 656 by more than 2 orders of magnitude during the SE case. Although the median N_{isg} in TAKAH 657 and PHILLall three sensitivity simulations with active break-up remains underestimated 658 compared to observations during the NW flow, the first TAKAH seems to produce unrealistically high median and 75th percentile values during the SE flow (Table 3). Indeed, 659 660 focusing on the N_{isg} time series (Fig. 3a) TAKAH is ~25% of the time shown to overestimate 661 the observed ICNCs by a factor of ~3, reaching up a factor of 10 on 30 January at 00:00. 662 TAKAHrim and PHILL, on the other hand, produces produce more reasonable Ning 663 concentrations of ice particles throughout both case studies, increasing with the Nisg values in 664 the 75th percentile by more thanexceeding 100 (50) L⁻¹ during the NW (SE) case study (Table 665 3), reducing which is found to reduce the gap between observations and model predictions.

It is worth noting that, despite the fact that the Takahashi parameterization (Eq. 2) is applied to both TAKAH and TAKAHrim simulations, the former seems to systematically overestimate the number of secondary ice fragments, while the latter produces ICNCs that are more consistent with the observations. Hence the Takahashi parameterization predicts reasonable results if it is allowed to generate fragments from collisions between rimed ice particles only (Section 2.4).

672

Table 4. The 25th, 50th (median) and 75th percentiles of IWC (in gm⁻³) time series.

Simulation	25 th perc.		Mee	lian	75 th	perc.
	NW winds	SE winds	NW winds	SE winds	NW winds	SE winds
OBSERVED	31.5×10 - ³ 0.03	<u>22.0×10</u> - ³ .02	0. 63<u>19</u>	0. 24<u>11</u>	0.66	0.26
CNTRL	4.3×10 ⁻³	5.0×10 ⁻³	0.03	0.04	0.15	0.12

TAKAH	1.3×10 ⁻³	2.0×10 ⁻³	0.10	0.09	0.52	0.34
TAKAHrim	4.7×10 ⁻³	<u>3.1×10⁻³</u>	<u>0.08</u>	<u>0.04</u>	<u>0.33</u>	0.27
PHILL	3.8×10 ⁻³	3.7×10 ⁻³	0.10	0.02	0.38	0.30
BIPS100_PHILL	3.9×10 ⁻³	9. <u>01</u> ×10 ⁻³	0.09	0.03	0.40	0. 31<u>30</u>

The observed IWC time series (Fig. 3b) are frequently reaching ~1 gm⁻³ during the NW case, with the median values being a factor of 2.5 higher than those observed during the SE 676 677 case (Table 4). This highlights indicates the presence of more massive ice particles when higher 678 updraft velocities prevail. The CNTRL simulation cannot produce IWC values > 0.8 gm⁻³ and 679 is most of the time below the observed range. Adding a description of the BR process (i.e., in 680 TAKAH, TAKAHrim and PHILL-simulations) sufficiently increases the modeled IWC by up 681 to ~1 order of magnitude between 26 January 12:00 UTC and 27 January 06:00 UTC, when 682 the modeled N_{isg} exceeds 100 L⁻¹ and the temperatures remain temperature remains higher than -16 °C. The same conditions are observed in the SE case, between 12:00 and 18:00 UTC on 683 684 30 January, when IWC shows a ~3 fold enhancement reaching the observed levels. The IWC 685 values in the third quartile predicted by TAKAH, TAKAHrim and PHILL are more than a 686 factor of 2 higher than the ones predicted by CNTRL (Table 4). This increase improves the 687 model performance although the modeled IWC remains slightly underestimated 688 (overestimated) during the NW (SE) case. The size distribution of the three ice hydrometeors 689 assumed by all three sensitivity simulations species considered in the M05 scheme (supplement 690 Fig. S4) reveals that, the implementation of the BR mechanism producesleads to elevated 691 concentrations of relatively smaller cloud ice crystals, but at the same time increases the 692 concentrations of snow particles. This is the reason why the modeled total ice mass is also 693 increased compared with the CNTRL simulation.

694 Fig. 3c compares the simulated cloud LWC to the concurrent CDP observations at JFJ 695 station. The LWC values recorded during the NW case are highly variant, reaching up to 0.75 696 gm⁻³, which is substantially higher than the respective maximum LWC observed during the SE 697 case (0.30 gm⁻³). On 26 January before 12:00 UTC, all three-sensitivity simulations predict 698 LWC > 1 gm⁻³, which, however, cannot be validated against measurements due to missing 699 valuesdata in the CDP time series. Note that this period is excluded from the statistics derived 700 in Table 5. The CNTRL simulation is found to overestimate the cloud LWC, predicting 0.42 701 (0.25) gm⁻³ in the third quartile, which is a factor of \sim 2 higher than the mean LWC observed 702 values during the NW (SE) case (Table 5).

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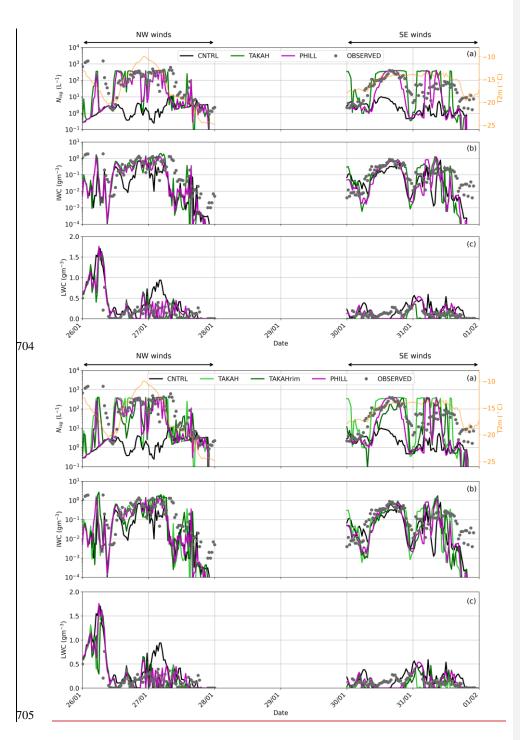


Figure 3. Time series of (a) total N_{isg} and temperature at 2 m height (orange line), (b) IWC and
(c) LWC, predicted by the CNTRL (black line), TAKAH (light green line), TAKAHrim (dark
green) and PHILL (magenta line) simulations between 26 January and 1 February 2014. The
grey dots in all three panels represent the 2D-S ICNCs, the inferred IWC and the CDP LWC
measured at the JFJ station, respectively. Note the logarithmic y-axes in panels a and b.

711

712 The modeled LWC in the 75th percentile is decreased by a factor of >2.5 (-1.5)-5 in 713 TAKAH (PHILL)-the simulations that account for the BR process (Table 5), improving the 714 agreement with observations (Fig. 3c). This The reduction in LWC is expected, considering that 715 the higher Nisg produced when BR is activated can readily deplete the surrounding droplets 716 under liquid water subsaturated conditions through the WBF process. This introduces a 717 challenging environment to simulate, as the model is sometimes seen to convert water to ice 718 too rapidly, leading to cloud glaciation (e.g., on 30 January after 12:00 UTC). Despite all sinks 719 of cloud water (i.e., condensation freezing, WBF or riming), observations at JFJ suggest that mixed-phase regions are generally sustained (Lloyd et al., 2015). This is particularly true for 720 721 the NW case, when the sufficiently large updrafts caused by the steep ascent of the air masses 722 help maintain the supersaturation with respect to liquid water (Lohmann et al., 2016). PHILL 723 and TAKAHrim can more efficiently sustain the observed mixed-phase conditions compared 724 to TAKAH, which frequently results in explosive ice multiplication – especially during the SE 725 case - leading to an underestimation of the LWC (see Fig. 3c and Table 5).3c and Table 5). 726 TAKAH is, therefore, excluded from the following discussion as it fails to reproduce an 727 accurate liquid and ice partitioning.

728

729 **Table 5.** The 25th, 50th (median) and 75th percentiles of LWC (in gm⁻³) time series.

Simulation	25 th perc.		Me	dian	75 th perc.	
	NW winds	SE winds	NW winds	SE winds	NW winds	SE winds
OBSERVED	8.5×10 ⁻³	70.0×10 ⁻³	0.12	0.11	0.21	0.14
CNTRL	87.7×10 ⁻³	26.0×10 ⁻³	0.19	0.17	0.42	0.25
TAKAH	1.3×10 ⁻¹⁰	0.0	0.01	6.7×10 ⁻¹⁰	0.16	0.05
TAKAHrim	1.2×10 ⁻⁷	0.0	0.06	2.8×10 ⁻⁶	0.23	0.09
PHILL	6.3×10 ⁻⁸	0.0	0.09	0.03	0.26	0.18
BIPS10	82.0×10 ⁻³	4.6×10 ⁻³	0.18	0.12	0.33	0.24
BIPS100	67.1×10 ⁻³	13.1×10 ⁻³	0.18	0.13	0.36	0.23
BIPS100_PHILL	6.3×10 ⁻¹⁰	0.0	0.09	0.06	0.27	0.10

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731	The time-averaged vertical profiles of number concentrations of cloud ice (N_i) , graupel
732	(N_g) , snow (N_s) and total N_{isg} <u>number concentrations</u> are illustrated in Fig. 4 for the CNTRL,
733	TAKAHTAKAHrim and PHILL simulations. The mean N_i (Fig. 4a) and N_{isg} profiles (Fig. 4d)
734	are enhanced by more than up to 2 orders of magnitude enhanced in TAKAHTAKAHrim and
735	PHILL compared to CNTRL. It is again obvious that TAKAH produces higher concentrations
736	than PHILL, at leastDuring the NW flow both simulations including the BR process produce
737	similar vertical distribution of the ice hydrometeors in the lower 2 (lowest 1)-1.5 km ofin the
738	atmosphere. This is not the case for the SE case, where TAKAHrim seems to predict a rapid
739	decrease in N_i and N_s and thus in total N_{isg} with altitude. The main difference between these
740	two simulations lies in the fact that the total LWC and hence, the probability of riming,
741	decreases with height limiting the efficiency of BR in TAKAHrim. This become more evident
742	during the NW (SE) case. As discussed above, this regularly leads to overestimated Nise
743	compared with the observed amount of ice close to the surface. All three simulations, however,
744	produce similar resultsSE case where mixed-phase conditions are exclusively confined below
745	1.5 km in the atmosphere (Section 3.1.1). However, we cannot estimate which vertical
746	distribution better represents reality, due to the lack of corresponding measured profiles.
747	TAKAHrim coincides with PHILL only when there is sufficient liquid water in the atmosphere,
748	allowing for the riming of the ice hydrometeors. Moreover, at heights above ~2.5 km, where
749	the simulated temperatures drop well below -20 °C (supplement Fig. S5), <u>S5</u>), all three
750	simulations are seen to produce similar results. This implies the greater importance of SIP
751	through BR at heights below 2-3 km in the atmosphere (i.e., in the temperature range between
752	$\sim -18 \text{ °C and } \sim -10 \text{ °C}$).
753	

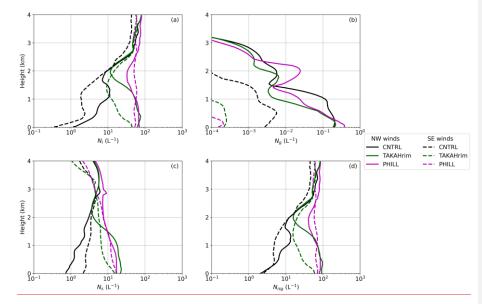


Figure 4. Mean vertical profiles of (a) N_{i} , (b) N_{g} , (c) N_{s} and (d) total N_{isg} , predicted by the CNTRL (black), TAKAHrim (dark green) and PHILL (magenta) simulations for the NW (solid lines) and SE (dashed lines) cases. Note the different scale on the x axis of the N_g vertical distribution. The height is given in km a.g.l.

Graupel number concentrations (Fig. 4b) do not contribute much to the modeled ice phase, especially during the SE case when the simulated N_g is negligible compared with the N_i and N_s (Fig. 4c). In the M05 scheme, portion of the rimed cloud or rain water onto snow is allowed to convert into graupel (Reisner et al. 1998), provided that snow, cloud liquid and rain water mixing ratios exceed a threshold of 0.1, 0.5 and 0.1 g kg⁻¹, respectively. These mixing ratio thresholds for graupel formation are arbitrary and might not be suitable for the examined conditions, preventing the formation of graupel from rimed snowflakes (Morrison and Grabowski, 2008). During the NW case, however, we can identify substantially higher N_g than the SE case, owing to the presence of sufficient supercooled liquid water especially during the first half of 26 January. Activating the BR mechanism in TAKAHTAKAHrim and PHILL 770 generally decreases the simulated N_g in both cases (Fig. 4c), suggesting that break-up of graupel 771 contributes to ice multiplication.

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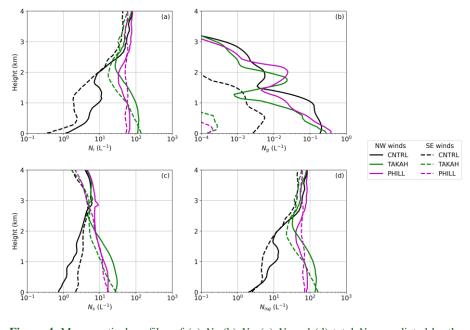


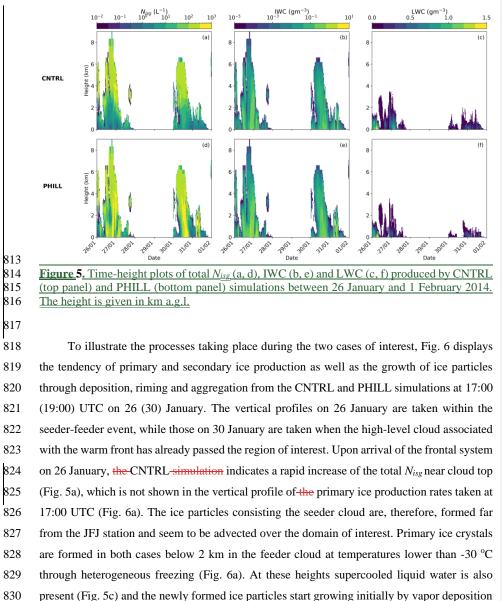
Figure 4. Mean vertical profiles of (a) N_s, (b) N_g, (c) N_s and (d) total N_{isg}, predicted by the CNTRL (black),-TAKAH (green) and PHILL (magenta) simulations for the NW (solid lines) and SE (dashed lines) cases. Note the different scale on the x axis of the N_e vertical distribution. The height is given in km a.g.l.

The mean vertical profile of N_s (Fig. 4c) seems to follow the respective profile of N_i (Fig. 4a). Unlike the graupel concentrations, including the BR mechanism is found to enhance N_s up to one order of magnitude compared to the CNTRL simulation. Focusing on a single model time step when the BR mechanism is activated, the size distribution of snow particles shown in the supplement Fig. S4 reveals that the increase in snow number concentrations can reach up to 2 orders of magnitude during the NW case. This is a logical consequence of the increase in number concentration of ice crystals, which are converting to snow particles after ice crystal growth (i.e., cloud-ice-to-snow autoconversion), when surpassing a characteristic mean diameter of 250 µm. This will be discussed in detail in the following section, which is focused. 788 We focus subsequent discussion on the PHILL simulation as it because i) TAKAHrim provides 789 a slightly more accurate representation comparable results in terms of the in-cloud phase 790 partitioning-compared with TAKAH, and, ii) we explore the sensitivity of simulation results to 791 parameters not considered by the Takahashi formulation. 792

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793 3.1.1 Conditions favoring BR in the two considered events

The temporal evolution of the vertical profiles of Nisg, IWC and LWC can provide valuable 794 795 insight on the drivers of enhanced ice formation in the wintertime alpine MPCs. Fig. 5 reveals 796 the presence of a seeder-feeder cloud system with sustained mixed-phase conditions confined 797 to levels below ~3 km (~1.5 km) in the NW (SE) case and a pure ice cloud aloft. Such 798 configurations are a well-known type of orographic multi-layer clouds that enhances 799 precipitation over mountains (e.g., Browning et al., 1974, 1975; Roe, 2005). Cloud 800 condensation is promoted by the synergy between a midlatitude frontal system and its 801 orographically induced ascent over the mountain range (Fig. 1). The separation between the 802 seeder and feeder clouds is often nonexistent, meaning that ice seeding can occur either in 803 layered clouds or internally within one cloud (Roe, 2005; Proske et al., 2021). In the first case, 804 which seems to occur here as well, there can be vertical continuum of cloud condensates 805 between the seeder and the feeder cloud due to precipitation of ice crystals from the higher-806 level cloud (Fig. 5a). This means that the seeding ice crystals fall through subsaturated cloud-807 free air before reaching the feeder region of the cloud and might sublimate. AThe remote-808 sensing analysis to 11-year of data over Switzerland presented by Proske et al. (2021), showed 809 that in-cloud seeding occurs in 18% of the observations, while the external seeder-feeder 810 mechanism is present 15% of the time (Proske et al., 2021) when the seeder is a cirrus 811 cloud.when the seeder is a cirrus cloud.



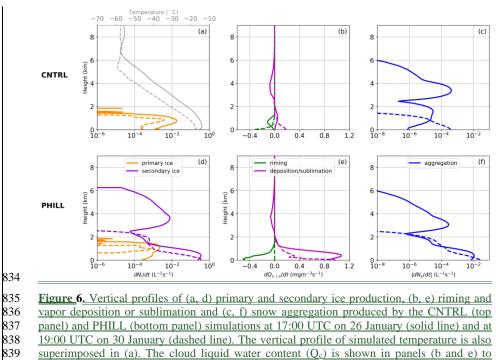
831 due to supersaturation with respect to ice, followed by riming (Fig. 6b). This is also indicated

by the increased IWC values closer to the ground (Fig. 5b).

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840 represent the tendency due to riming, while the mass mixing ratio of the ice and snow species

 $(Q_i + Q_s)$, are representing the relative tendencies due to vapor deposition or sublimation. Note that the tendencies due to snow aggregation in (c, f) are presented in absolute values. The height

841 842 843 is given in km a.g.l.

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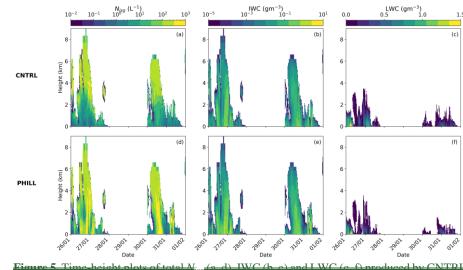
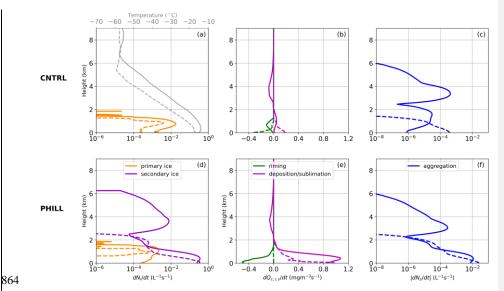


Figure (top panel) and PHILL (t panel) simulations January and The height is given in km a.g.l.

Focusing on the ice-seeding event of 26 January, the enhanced aggregation rate observed at heights above ~2.5 km in the atmosphere indicates the enhanced collision efficiencies of the precipitating ice particles while falling from the seeder cloud (Fig. 6c). Note that a portion of the sedimented ice particles sublimates before reaching the feeder cloud at heights ~3-5 km, indicating the prevailing unsaturated conditions in this layer (Fig. 6b). Within this layer-the aggregation of snowflakes weakens, while it is enhanced again when the falling hydrometeors 856 enter the feeder cloud. The bottom line is that, even under the simulated seeder-feeder events the concentrations of ice particles reaching the ground in CNTRL simulation remain severely 858 underestimated (Section 3.1). Despite the low concentrations of ice crystals simulated by the 859 CNTRL simulation, the low-level cloud is glaciated more frequently during the SE_ than during 860 the NW-winds case (Fig. 5c). This is probably because of the higher updraft velocities 861 prevailing until 28 January (Fig. 2d), preventing ice crystals from falling through the lower 862 parts of the cloud (Lohmann et al., 2016).



865 Figure 6. Vertical profiles of (a. d) primary and secondary icc production, (b, c) riming and 866 the CNTPL vapor deposition or sublimation and (c, f) snow aggregation produced d PHILI 867 868 The vertical profile 10.00 LITC 30 January (dashad lina) 869 870 871 (a). The cloud liquid water content (Q_e) in and superim represent the tendency due to riming, while the mass mixing ratio of the ice and snow species $(Q_{i} + Q_{j})$, are representing the relative tendencies due to vapor deposition or sublimation. Note 872 that the tendencies due to snow aggregation in (c, f) are presented in absolute values. The height 873 is given in km a.g.l.

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874 Activating the BR mechanism along with the seeding of precipitating hydrometeors in 875 PHILL-simulation shifts the simulated N_{isg} towards higher concentrations that are found to 876 exceed 300 L⁻¹ in the lower-level part of the cloud (Fig. 5d). On 26 January the mode of the 877 cloud ice distribution shifts to slightly bigger sizes, while on 30 January the modal sizes 878 become almost an order of magnitude smaller compared with the CNTRL simulation 879 (supplement Fig. S4). The enhanced concentrations of bigger ice particles simulated in the first 880 case experience rapid growth through vapor deposition and riming (Fig. 6e) causing a slight increase in the simulated IWC (Fig. 5e) at the expense of the surrounding cloud droplets in the 881 882 low-level feeder cloud (Fig. 5f). Nevertheless, the smaller ice particles simulated in the second 883 case grow less efficiently through vapor deposition, while the explosive multiplication of ice 884 through BR seems to fully glaciate the low-level cloud below ~1 km resulting in an almost zero 885 riming rate (Fig. 6e). The reduced primary ice production rate observed during both case 886 studies is a consequence of the depletion of liquid water when BR is considered (Fig. 6d). A 887 suppression of heterogeneous ice nucleation following the introduction of SIP into models has

already been reported in previous studies (Phillips et al., 2017b; Dedekind et al., 2021; Zhao and Liu, 2021b).

890 The key difference between CNTRL and PHILL simulations is that the latter takes 891 advantage of the enhanced ice particle growth through aggregation while falling to the feeder 892 cloud below ~2 km, -where large snowflakes coexist with smaller ice crystals (Fig. 4a, 6a, 6d). 893 This allows for differential settling, which enhances collision efficiency facilitating ice 894 multiplication through BR. This is the reason why the vertical profile of secondary ice 895 formation agrees with the corresponding profile of aggregation during both case studies (Fig. 896 6d, 6f). On 26 January the first secondary ice particles start forming already within the seeder 897 cloud with the contribution of SIP increasing considerably when reaching the feeder cloud, 898 where the tendency due to SIP is more than 3 orders of magnitude higher than primary ice 899 production (Fig. 6d). The significant role of SIP stands out also on 30 January at altitudes below 900 2 km. It is, therefore, essential to consider SIP though BR in the feeder cloud, in order to 901 achieve the enhanced levels of ICNCs frequently observed within seeder-feeder events in the 902 alpine region. This is in agreement with the observational study of Ramelli et al. (2021) on an 903 ice-seeding case occurring in the region around Davos in the Swiss Alps. In this study, they 904 proposed that SIP though HM and BR were necessary to explain the elevated ICNCs in feeder 905 clouds.

906 A classification of the dominant type of precipitation was applied to the polarimetric data 907 collected by a weather radar deployed at the Kleine Scheidegg station (2061 m a.s.l) during the 908 SE case between 30 and 31 January (supplement Fig. S6). In the derived time series, we can 909 identify periods when individual ice crystals (not aggregated and not significantly rimed) 910 dominate over the entire precipitation column followed by periods when a clear stratification 911 is present with ice crystals aloft and mostly aggregates and rimed ice particles below. This 912 stratification is observed on 30 January at 19:00 UTC when the model tendencies are extracted 913 (dashed lines in Fig. 6). Allowing for the BR process in PHILL-simulation results in a 2 orders 914 of magnitude enhancement in the aggregation rates close to the ground, which can better 915 reproduce the signatures observed in the hydrometeor classification at that time. An increase 916 in the simulated aggregates and rimed particles is expected to increase orographic precipitation, 917 which is important given that these low-level feeder clouds are incapable of producing 918 significant amounts of precipitation. Indeed, the mean surface precipitation produced by 919 PHILL is 30% (10%) increased during the NW (SE) case compared with CNTRL (supplement 920 Fig. S7), which is in contrast to Dedekind et al. (2021) where the activation of the BR process 921 is found to suppress the regions of strong surface precipitation. This was attributed to the

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922 limited efficiency of the small secondary ice particles to grow sufficiently to precipitation sizes923 when the local updrafts -lift them to the upper parts of the cloud that were glaciated. The radar-

based hydrometeor classification reveals also the predominance of ice crystals at the beginning

925 and the end of the precipitating periods (e.g., on 30 January at 15:00-17:30 or 31 January at

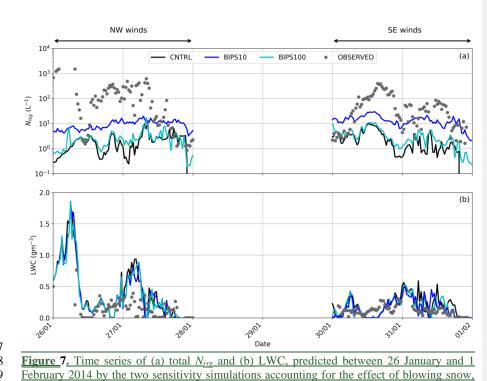
926 04:30-06:00), which is again more consistent with the vertical profile of N_i produced by PHILL

927 rather than the CNTRL simulation (supplement Fig. S6, S8).

928

929 3.2 Sensitivity to the injection of ice crystals from the surface

In this section we examine if the surface-originating small ice particles could have the potential 930 931 to initiate and enhance ice particle growth in the near-surface MPCs present in our case studies. 932 Fig. 7 illustrates two additional WRF simulations - BIPS10 and BIPS100 - where the ice 933 crystal source applied to the first model level is equal to 10 and 100 L⁻¹, respectively (Table 2). 934 Note that these two sensitivity tests do not consider any SIP process to analyze the influence 935 of BIPS only. The total Nisg values produced in BIPS10 are only slightly increased compared 936 to the CNTRL simulation and generally remain outside the observed range at JFJ (Fig. 7a). An 937 order of magnitude increase in the applied NBIPS is seen to enhance the modeled Nisg during 938 both case studies, however our simulations are still lacking ice particles. This is particularly 939 evident during the NW winds case, where the simulated Nisg varies most of the time around 10 940 L^{-1} , remaining an order of magnitude lower than the observations. During the SE case, the 941 model performance is slightly improved with the N_{isg} reaching up to ~25 L⁻¹ in BIPS100, which 942 occasionally falls within the lower limit of the observed ICNC values (e.g., in the evening of 943 31 January). At times when the detected ICNCs remain quite low (i.e., on the order of 10 L⁻¹), 944 the contribution of blowing snow particles probably from the Aletsch Glacier is sufficient to 945 explain the observations at JFJ.



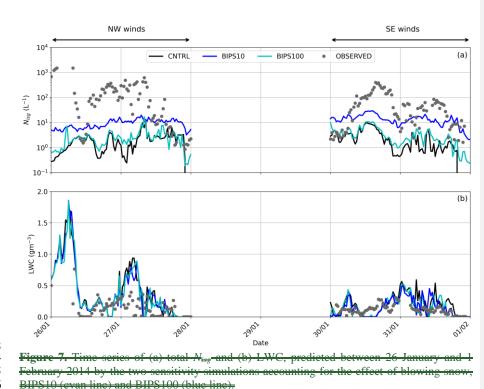
As indicated in Fig. 7b, during the NW flow the simulated LWC at the first model level in BIPS10 and BIPS100 almost coincides with the CNTRL simulation of WRF. The three sensitivity simulations are producing comparable median and quartile LWC values (Table 5), with BIPS10 and BIPS100 producing median LWC values closer to the observed ones during the SE flow. When comparing against the LWC values in the third quartile though, the two simulations lead to an overestimation up to a factor of ~1.5 during both case studies. Given that there is approximately a factor of >20 (5) difference between the modeled and observed ICNCs

BIPS10 (cyan line) and BIPS100 (blue line).

during the NW (SE) winds case (Table 3), Fig. 7 overall reveals that the addition of a source
of ice crystals from the effect of blowing snow cannot account for the observed liquid-ice phase
partitioning in the simulated orographic MPCs.

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Our findings are in contrast with the modeling study of Farrington et al. (2016), where a different approach was proposed to include the surface effect on the ICNCs simulated with WRF. In this study, a single model domain was used with a horizontal resolution of 1 km. To account for the flux of hoar crystals being detached from the surface by mechanical fracturing, Farrington et al. (2016) included a wind-dependent surface flux of frost flowers adapted from 973 Xu et al. (2013). Despite the improved performance of the-WRF-model in terms of predicted 974 ICNCs and LWC, the wind-dependent formulation of the surface flux caused the modeled 975 ICNCs to become strongly correlated with the simulated horizontal wind speed - a behavior 976 that was not confirmed by the observations of Lloyd et al. (2015). Nonetheless, the highest 977 observed ICNCs at the beginning of the NW case correspond to the time when both the 978 observed and modeled wind speed is the strongest (Fig. 2c), implying that a wind-dependent 979 surface flux of BIPS could potentially elevate the simulated N_{isg} to the observed levels at this 980 time.

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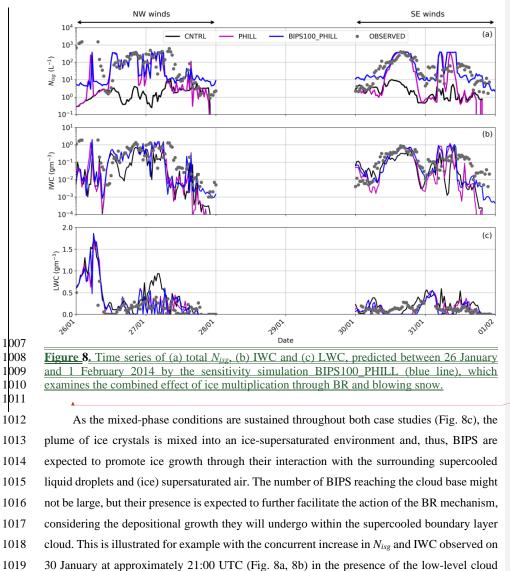
982 3.3 The synergistic impact of BR and surface-induced ice crystals

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It is deducible from the above discussion that the sole inclusion of a constant source of BIPS in our simulations cannot efficiently bridge the gap between modeled and measured ICNCs. Our aim in this section is to explore the combined effect of SIP through BR and blowing snow on the simulated orographic MPCs, since both processes are deemed to be important when trying to explain the high ICNCs observed in alpine environments. This is addressed in the final sensitivity simulation, BIPS100_PHILL, the results of which are compared with the CNTRL and PHILL simulations in Fig. 8.

990 In terms of the modeled ice particle concentrations, the combination of the simplified 991 blowing snow treatment and BR parameterization can account for most of the discrepancy between modeled and observed ICNCs, particularly during the SE case (Fig. 8a), when the 992 993 simulation leads to best agreement with the observed interquartile values (Table 3). 994 BIPS100_PHILL and CNTRL generally differ by an average factor of ~100 (40) during the 995 NW (SE) case, with the former producing N_{isg} values that are sometimes elevated by up to ~3 996 (2) orders of magnitude (Fig. 8a). Compared to the PHILL setup, including a source of BIPS 997 is found to improve the modeled ICNCs close to the surface episodically - for instance in the 998 evening of 30 and 31 January, with the Nisg in BIPS100_PHILL efficiently reaching the 999 observed levels (Fig. 8a). Note that BIPS can contribute to the modeled N_{isg} even without the 1000 presence of a near-surface orographic cloud (e.g., Geerts et al., 2015; Beck et al., 2018). For 1001 instance, BIPS100_PHILL is the only sensitivity simulation producing high Nisg values in the 1002 evening of 27 and 31 January, when the low-level cloud is dissipated (Fig. 5c, f). In the former 1003 case, however, the model results in an overestimate of the ICNCs, which is also observed 1004 during the early hours of 30 January, suggesting that the applied source of ice crystals is 1005 unrealistically high at this time.

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(Fig. 8c). Note that the elevated N_{isg} caused by the addition of BIPS is not always followed by

an efficient increase in the simulated IWC. This can be observed for example on 27 January at

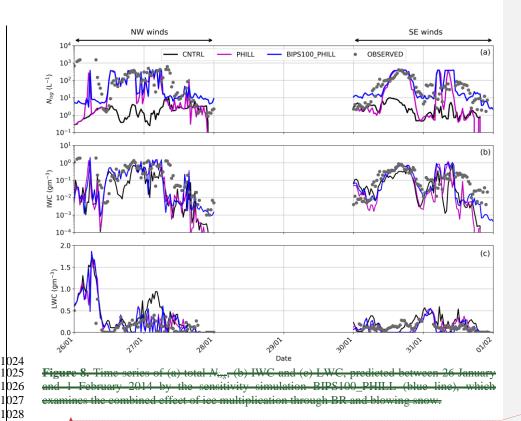
1022 12:00 UTC or in the evening of 31 January (Fig. 8b).

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1029 A discrepancy between modeled and observed IWC was also highlighted in the study of 1030 Farrington et al. (2016), and was attributed to the small sizes of the hoar frost particles assumed 1031 (i.e., 10 µm). Although here BIPS are assumed to have sizes of 100 µm, still the 1032 underestimation in the cloud IWC has not been overcome. This suggests that the applied source of BIPS combined with the effect of SIP through BR shifts the ice particle spectra to smaller 1033 1034 sizes, which are not very efficient in riming and the WBF process and, thus, do not always 1035 contribute to significant increases in IWC values. Overall, the interquartile values presented in 1036 Table 4 reveal that BIPS100_PHILL and PHILL yield almost identical IWC values, suggesting 1037 that the implementation of a constant source of BIPS does not further improve the representation of the total ice mass despite the improvements in the simulated Nisg. Focusing 1038 1039 on the LWC values in the third quartile, though, including a source of BIPS results in better 1040 agreement with the CLACE observations during the SE case, while it is shown to have little 1041 effect on the cloud liquid phase during the NW case (Table 5). Despite the increase in the 1042 modeled Nisg observed in BIPS100_PHILL especially during the SE case, the liquid water in the low-level orographic cloud is not further depleted (Fig. 8c). This is presumably because the
mean surface precipitation produced is also enhanced by almost ~20% compared to PHILL
(supplement Fig. S7), which seems to balance the excessive ice production.

1046 One final point that is worth noting here is that there are still some certain periods when 1047 BIPS100 PHILL fails to reproduce the observed range of ICNCs. This could imply the 1048 potential contribution of additional ice multiplication processes to the observed ice particle 1049 concentrations. Indeed, the seeder-feeder configuration observed in the examined case studies could favor the fragmentation of sublimating hydrometeors while falling through an 1050 1051 subsaturated environment before entering the feeder cloud (e.g., Bacon et al., 1998). The so-1052 called "sublimational break-up" is an overlooked SIP process which is not yet described in the 1053 M05 scheme. Also, note that the periods when the modeled ICNCs remain below the observed 1054 ice number levels are mainly identified when the simulated temperature drops below -15 °C 1055 and the wind speed exceeds 10 ms⁻¹ or even 20 ms⁻¹ (e.g., in the morning of 26 January or 27 1056 January at around 12:00 UTC). This is when the incorporation of surface-based processes 1057 becomes of primary importance. The simplified methodology we followed here although 1058 instructive, yet it faces several limitations. For instance, the constant source of BIPS is 1059 sometimes found to overestimate the modeled N_{isg} and IWC. In order to accurately assess the potential role of the snow-covered surfaces in elevating the simulated ICNCs, an improved 1060 1061 spatio-temporal description of the concentration and distribution of BIPS is required. 1062 Furthermore, the applied ice crystal source is independent of some key parameters controlling 1063 its resuspension, such as the horizontal wind speed, the updrafts or the friction velocity (e.g., 1064 Vionnet et al., 2013, 2014). For example, in the early morning hours of 26 January, the high 1065 simulated horizontal and vertical velocities (Fig. 2c, 2d) are expected to loft significant BIPS concentrations into the cloud layer, owing to enhanced mechanical mixing and momentum flux 1066 1067 close to the surface. Nonetheless, the contribution of the induced plume of BIPS remains 1068 constant throughout the NW case study (Fig. 7a), which seems to lead to an underestimation 1069 of the total ice particle concentration and mass. A more realistic parameterization of the BIPS 1070 flux or the coupling with a detailed snowpack model would, therefore, be essential for a more 1071 accurate representation of the effect of blowing snow.

1072

1073 **4.** Summary and conclusions

1074 This study employs the mesoscale model WRF to explore the potential impact of ice 1075 multiplication processes on the liquid-ice phase partitioning in the orographic MPCs observed 1076 during the CLACE 2014 campaign at the mountain-top site of JFJ in the Swiss Alps. The 1077 orography surrounding JFJ channels the direction of the horizontal wind speed, giving us the 1078 opportunity to analyze two frontal cases occurring under NW and SE conditions.

1079 DS and BR mechanisms were implemented in the default M05 scheme in WRF, in 1080 addition to the HM parameterization, which however remained inactive in the simulated 1081 temperature range (-10 to -24 °C). The DS process is parameterized following either the latest 1082 theoretical formulation developed by Phillips et al. (2018) or the more simplified 1083 parameterization proposed by Sullivan et al. (2018a). Our sensitivity simulations revealed that 1084 the DS mechanism is ineffective in the two considered alpine MPCs, even under the higher 1085 updraft velocity conditions associated with the NW winds case study. This is due, owing to 1086 athe lack of sufficiently big raindrops, necessary to initiate this large drops required for the 1087 process.

1088 To parameterize the number of fragments generated per ice-ice collision we followed 1089 again two different approaches: either the simplified temperature dependent formulation of 1090 Takahashi et al. (1995) scaled for the size of the particle that undergo fragmentation (Sotiropoulou et al., 2021a) or the more advanced physically-based Phillips et al. (2017a) 1091 1092 parameterization. It is important to apply the Takahashi parameterization only to consider 1093 collisions between rimed ice particles, otherwise the number of generated fragments is 1094 significantly overestimated. Including a description of the BR mechanism is essential for 1095 reproducing the ICNCs observed in the simulated orographic clouds, especially at temperatures higher than ~ -15 °C, where INPs are generally sparse. SIP through BR is found to enhance the 1096 1097 modeled ICNCs by up to 3 (2) orders of magnitude during the NW (SE) case, improving the 1098 model agreement with observations. This ice enhancement can cause up to an order of 1099 magnitude increase in the mean simulated IWC values compared with the CNTRL simulation, 1100 which is attributed to the enhanced ice crystal growth and cloud-ice-to-snow autoconversion. 1101 The increase in the simulated ICNCs also depletes the cloud LWC by at least a factor of 2 1102 during both cases, which is more consistent with the measured LWC values.

1103 One of the most interesting outcomes of this study is the association of the enhanced BR 1104 efficiency with the occurrence of in-cloud seeder-feeder events, which are commonly found in 1105 Switzerland (Proske et al., 2021). While ice-seeding situations are associated with enhanced 1106 orographic precipitation in the alpine region, the CNTRL simulation fails to reproduce the 1107 elevated ICNCs reaching the ground. The falling ice hydrometeors experience efficient growth 1108 through aggregation in the seeder part of the cloud, which is enhanced when reaching the feeder

1109 cloud at altitudes below 2 km, where primary ice crystals form and grow through vapor 1110 deposition and riming. Aggregation of snowflakes seems to be the major driver of secondary 1111 ice formation in the examined seeder-feeder events. SIP though BR is initiated already within 1112 the seeder cloud, while it becomes immensely important in the feeder cloud where its 1113 production rate exceeds the one of primary ice formation. The increased generation of 1114 secondary ice fragments does not always lead to ice explosion and cloud glaciation, as it is 1115 followed by an enhancement in the precipitation sink owing to a shift in the ice particle 1116 spectrum. Including a description of the BR mechanism is, therefore, crucial for explaining the 1117 ice particle concentration and mass observed in the low-level feeder clouds.

1118 To assess the potential role of blowing snow in the simulated orographic clouds, a 1119 constant source of ice crystals was introduced in the first atmospheric level of WRF. Our results 1120 indicate that blowing snow alone cannot explain the high ICNCs observed at JFJ, but when this 1121 source is combined with the BR mechanism then the gap between modeled and measured 1122 ICNCs is sufficiently bridged. The biggest influence of blowing snow is mainly detected at 1123 times when the simulated temperatures are low enough (< -15 °C), while the presence of a low-1124 level cloud is required for SIP to manifest. The concentrations of BIPS reaching the cloud base 1125 are not high, but when they are mixed among supercooled liquid droplets they are expected to 1126 grow, facilitating ice multiplication through BR. Nonetheless, including a wind-dependence or 1127 a spatio-temporal variability in the applied ice crystal source would be essential to provide a 1128 more precise description of the effect of blowing snow on the simulated clouds.

1129 Overall, our findings indicate that outside the HM temperature range, a correct 1130 representation of both secondary ice (through BR) and an external ice seeding mechanism, 1131 which is primarily precipitating ice particles formed aloft and to a lesser degree wind-blown 1132 ice from the surface, will improve the accuracy of the liquid-ice partitioning in MPCs predicted 1133 by atmospheric numerical models. More precisely, the implementation of SIP through BR can 1134 effectively shift the number concentrations of ice particles in the right direction dictated by 1135 observations of alpine MPCs, which is in turn critical not only for the determination of their 1136 optical properties but also for the accurate estimate of precipitation patternsis fundamentally 1137 important for accurately predicting the liquid-ice partitioning and properties of MPCs. Given 1138 the high frequency of seeder-feeder events in orographic environments, including the new 1139 physics of BR may address a large source of predictive bias in atmospheric models.

1140

1141	Code and data availability. The WRF outputs presented in this study will be made available at	
1142	https://zenodo.org/, while the updated Morrison scheme is available upon request. @Note by	
1143	authors: Data will be made available upon acceptance of final publication.	
1144		
1145	Competing interests. The authors declare no conflict of interest.	
1146		
1147	Author Contributions. PG and AN conceived and led this study with input from GS. EV helped	
1148	with the WRF configuration and setup. GS provided the updated microphysics scheme with	
1149	the detailed BR parameterizations. PG implemented the DS parameterizations with help from	
1150	GS, conducted the simulations, analyzed the results and, together with AN, wrote the main	
1151	paper. All authors contributed to the scientific interpretation and writing of the paper.	
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