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

Georgia Sotiropoulou[1,2], Etienne Vignon[3], Gillian Young[4], Hugh Morrison[5,6], Sebastian J. O'Shea[7], Thomas Lachlan-Cope[8], Alexis Berne[3], Athanasios Nenes[1,9]

[1]Laboratory of Atmospheric Processes and their Impacts (LAPI), Ecole Polytechnique Fédérale de Lausanne (EPFL), Lausanne, Switzerland

[2]Department of Meteorology, Stockholm University & Bolin Center for Climate Research, Sweden

[3]Environmental Remote Sensing Laboratory (LTE), EPFL, Lausanne, Switzerland

[4]School of Earth and Environment, University of Leeds, UK

[5]National Center for Atmospheric Research, Boulder, CO, USA

[6]ARC Centre for Excellence in Climate System Science, University of New South Wales, Sydney, Australia

[7]Centre for Atmospheric Science, University of Manchester, UK

[8]British Antarctic Survey, Cambridge, UK

[9]ICE-HT, Foundation for Research and Technology Hellas (FORTH), Patras, Greece

Correspondence to: georgia.sotiropoulou@epfl.ch, athanasios.nenes@epfl.ch

Abstract

The correct representation of Antarctic clouds in atmospheric models is crucial for accurate projections of the future Antarctic climate. This is particularly true for summer clouds which play a critical role in the surface melting of the ice-shelf in the vicinity of Weddell Sea. However these clouds are often poorly represented, as ice crystal number concentrations (ICNCs) are undepredicted by atmospheric models, even when primary ice formation is constrained with aerosol measurements. Rime-splintering, thought to be the dominant secondary ice production (SIP) mechanism at temperatures between -8 and -3°C, is also very weak in summer Antarctic conditions. Including a parameterization for SIP due to break-up (BR) from collisions between ice particles in the Weather and Research Forecasting model bridges the gap between observations and simulations, suggesting that BR could account for the enhanced ICNCs in the pristine Antarctic atmosphere. These results are insensitive to uncertainties in primary ice production. The BR mechanism is currently not represented in

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most weather prediction and climate models; including this process can have a significant impact on the Antarctic radiation budget and thus in projections of the future regional climate.

1. Introduction

Predictions of Antarctic climate are hampered by the poor representation of mixed-phase clouds over the Southern Ocean and the Antarctic Seas (Haynes et al., 2011; Flato et al., 2013; Bodas-Salcedo et al., 2014; Hyder et al., 2018). Model simulations reveal significant discrepancies in the Antarctic surface radiation budget, associated with cloud biases that are driven by errors in the representation of the cloud microphysical structure (Lawson and Gettelman, 2014; King et al., 2015; Listowski and Lachlan-Cope, 2017). A correct representation of the cloud radiative impacts largely depends on the parameterization of cloud microphysical processes (Listowski and Lachlan-Cope, 2017; Hines et al., 2019; Young et al., 2019) and precipitation (Vignon et al., 2019), which determine the concentration and characteristics of liquid drops and ice crystals.

Droplet formation processes are relatively well understood and prediction biases are known to arise from errors in cloud dynamics and aerosol predictions (Sullivan et al., 2016). Errors in ice formation processes, however, are far from resolved. While progress has been made during the past decade in the study of primary ice production (DeMott et al., 2011), secondary ice production (SIP) remains poorly understood (Field et al., 2017). SIP refers to the generation of ice crystals in a cloud that does not require ice-nucleating particles (INPs) (Field et al., 2017) and has been studied primarily in convective clouds (Lawson et al., 2015; Field et al., 2017), where the observed ice crystal concentrations (ICNCs) can be up to 3-4 orders of magnitudes higher than the available INPs. However, observations indicate that SIP can also magnify ICNCs to a significant extent in polar clouds (Lloyd et al., 2015; Lachlan-Cope et al., 2016) where low aerosol concentrations are frequently found (Lachlan-Cope et al., 2016; O'Shea et al. 2017), with important implications for the surface radiative balance (Young et al., 2019). Yet the mechanisms underlying SIP remain uncertain (Field et al., 2017).

The only well-established SIP mechanism that has been extensively implemented in weather forecast and climate models is rime-splintering (Hallett and Mossop, 1974), also known as the Hallett-Mossop process (H-M), which refers to the production of ice splinters after collisions of supercooled droplets with ice particles (Hallett and Mossop, 1974; Heymsfield and Mossop, 1984). This process is effective only in a limited temperature range, between -8 and -3°C, and requires the presence of supercooled liquid droplets both smaller

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and larger than 13 μm and 24 μm, respectively. (Mossop and Hallett, 1974; Choularton et al., 1980). However, recent studies have shown that H-M cannot sufficiently explain the enhanced ICNCs observed in both Arctic (Sotiropoulou et al., 2020) and Antarctic (Young et al., 2019) clouds. While some Antarctic studies (Vergara-Temprado et al., 2018; Young et al., 2019) suggest that the underestimation of ice multiplication in models might be related to uncertainties in the description of the H-M process, we argue that this is likely driven by the fact that almost no models include other SIP mechanisms.

Another SIP mechanism, identified in recent laboratory studies (Leisner et al*.*, 2014; Lauber et al., 2018), is the generation of ice fragments from shattering of relatively large frozen drops. This process however, while it is very efficient in convective clouds (Korolev et al., 2019), it has been found ineffective in polar regions (Fu et al., 2019; Sotiropoulou et al., 2020). This is in agreement with Lawson et al. (2017) and Sullivan et al. (2018a) who have shown that drop-shattering occurs in clouds with a relatively warm cloud base.

Mechanical break-up (BR) of ice particles that collide with each other is another process that results in ice multiplication (Vardiman, 1978; Takahashi et al., 1995) and it operates over a wide temperature range with maximum efficiency around -15◦C. Limited knowledge of the BR mechanism comes from few laboratory experiments (Vardiman, 1978; Takahashi et al., 1995) and small-scale modeling (Fridlind et al., 2007; Yano and Phillips, 2011, 2016; Phillips et al, 2017a,b; Sullivan et al. 2018a; Sotiropoulou et al., 2020). To the authors knowledge only two attempts have been made to incorporate this process in mesoscale models (Sullivan et al., 2018b; Hoarau et al., 2018). Specifically, Hoarau et al. (2018) assumed a constant number of fragments (F_{BR}) generated per snow-graupel collision in Meso-NH model, while Sullivan et al. (2018b) implemented a temperature-dependent relationship for F_{BR} in COSMO-ART for several types of collisions (e.g. crystal-graupel, graupel-hail, etc), based on the results of Takahashi et al. (1995). Phillips et al. (2017a) recently developed a physically-based description of F_{BR}, which is a function of collisional kinetic energy and accounts for the effect of the colliding particles' size and rimed fraction (Ψ). While being more advanced than any other parameterization proposed for BR, this scheme has never been implemented in mesoscale models before; it has only been tested in small-scale models for convective (Phillips et al., 2017b; Qu et al., 2020) and Arctic (Sotiropoulou et al., 2020) cloud conditions.

Sotiropoulou et al. (2020) recently showed that the observed ICNCs in Arctic clouds within the H-M temperature zone can be explained only by the combination of BR with the H-M process, which results in a 10 to 20-fold enhancement of the primary ice crystals. Based

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on their results, we postulate that BR may also play a critical role in Antarctic clouds and can potentially explain the discrepancy between the observed INPs and ICNCs in the region (Young et al., 2019). To test this hypothesis, we implement parameterizations of the BR process in the Morrison microphysics scheme (Morrison et al., 2005) (hereafter M05) in the Weather and Research Forecasting (WRF) model V4.0.1 and examine their influence on the Antarctic clouds observed during the Microphysics of Antarctic Clouds (MAC) flight campaign (O'Shea et al., 2017; Young et al., 2019).

2. Observations

2.1. MAC Instrumentation

The MAC flight campaign was conducted in November–December 2015 over coastal Antarctica and the Weddell Sea, with the aim to offer detailed measurements of the microphysical and aerosol properties of the Antarctic atmosphere. MAC included an extensive suite of airborne and ground-based instruments, a detailed description of which can be found in O'Shea et al. (2017); here we only offer a brief recap of the instrumentation used in this study.

Cloud particle size distributions were derived using the images from two optical array probes (OAPs): a 2D Stereo (2DS, SPEC Inc., USA; Lawson et al., 2006) with a nominal size range of 10 to 1280 μm (10 μm pixel resolution) and a cloud imaging probe (CIP-25, DMT Inc., USA; Baumgardner et al., 2001) with a size range of 25 to 1600 μm. Shattering effect in the probes' inlet were corrected by applying "antishatter" tips (Korolev et al., 2011) and inter-arrival time (IAT) post analysis (Crosier et al., 2011).

Aerosol particle measurements of sizes 0.25 to 32 μm were made using the Grimm optical particle counter (GRIMM model 1.109), while a Cloud Aerosol Spectrometer (CAS, DMT; Baumgardner et al., 2001; Glen and Brooks, 2013) measured particles between 0.6 to 50 μm. Following the methodology of Young et al. (2019) and O'Shea et al. (2017), we only consider Grimm measurements of particles with diameter smaller than 1.6 μm in our analysis to avoid including data subject to inlet losses at larger particle sizes. Finally, the aircraft also included instrumentation to measure temperature, turbulence, humidity, radiation and surface temperature (King et al., 2008).

2.2 Case study

For our investigations we focus on the MAC case examined in Young et al. (2019), for which

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they showed that the H-M process, as currently parameterized in WRF, cannot explain the observed ICNCs. Young et al. (2019) utilized measurements from two MAC flights, M218 and M219, combined in one case study; both flights were conducted on 27 November 2015 over the Weddell Sea (Fig. 1): M218 between 15.3-16.7 UTC and M219 between 20.45-22.5 UTC. On that day, a low pressure system persisted over the eastern Weddell Sea, resulting in a southeasterly flow reaching the aircraft with air mass back trajectories from around the low pressure system, towards the Antarctic Peninsula and southern Patagonia (O'Shea et al., 2017).

The temperature and microphysical conditions encountered during these flights are representative of the MAC campaign (see Table 1 in O'Shea et al., 2017, and Fig. S6 in Young et al., 2019) and generally representative of West Antarctica (Lachlan-Cope et al., 2016). Cloud measurements were collected mainly within the lowest 1.1 km above sea-level (a.s.l.) during both flights and within a temperature range of ~ -9 to -3^{o}C. Aerosol measurements were consistently low (Young et al., 2019), while ICNCs were substantially higher: the mean concentration of aerosols with sizes between 0.5-1.6 μm was 0.56 scm^{-3} and 0.41 scm^{-3} (cm^{-3} at standard temperature and pressure) for M218 and M219, respectively, while the corresponding mean (max) ICNCs were 1.16 (9.03) L^{-1} and 3.33 (87.31) L^{-1} for the two flights. Such low aerosol conditions and concurrent high ICNC concentrations within this temperature range are frequently found in West Antarctica (Lachlan-Cope et al., 2016). Moreover, similar cloud droplet concentrations (N_{drop}) were measured during both flights (Young et al., 2019): the mean N_{drop} was 82.7 cm^{-3} for M218 and 100.4 cm^{-3} for M219, which are comparable with previous observations from the Antarctic Peninsula (Lachlan-Cope et al., 2016).

3. Modeling Methods

3.1. Model set-up

This study is conducted with the WRF model (Skamarock et al., 2008), version 4.0.1, by applying the same model set-up as in Young et al. (2019). Two domains with a respective horizontal resolution of 5 km and 1 km are used, where the inner one is two-way nested to the parent domain (Fig. 1). The polar stereographic projection is applied. The outer domain is centered at 74.2^{o}N, 30^{o}E and includes 201 × 201 grid points, while the second domain consists of 326 × 406 grids. Both domains have a high vertical resolution with 70 eta levels, 25 of which correspond to lowest 2 km of the atmosphere. The model top is set to 50 hPa. The

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simulation period spans from 26 to 28 November 2014, 00:00 UTC, allowing for a 24-hour spin up period before the day of interest (27 November). The model timestep is set to 30 (6) sec for the outer (inner) domain, while output data are produced every 30 minutes.

Input data for the initial, lateral and boundary conditions for the simulations are obtained from the European Centre for Medium-Range Weather Forecasting reanalysis (Dee et al., 2011), as recommended by Bromwich et al. (2013). For both shortwave and longwave radiation components, the RRTMG radiation scheme (Rapid Radiative Transfer Model for GCMs) is applied. The Mellor-Yamada-Nakanishi-Niino (MYNN; Nakanishi and Niino, 2006) 2.5-level closure planetary boundary layer (PBL) and surface options are also implemented, in combination with the Noah Land Surface Model (Noah LSM; Chen and Dudhia, 2001), which includes a simplified thermodynamic sea-ice model. Given the short run length, time-varying sea ice concentrations are not utilized. Young et al. (2019) used the PWRF V3.6.1 to represent fractional sea-ice, a capability not available in standard WRF V3.6. However, this option has been made available in the more recent V4.0.1 that we use in this study. Following Young et al. (2019), the sea-ice albedo is set to 0.82, with a default thickness of 3 m, and snow accumulation depth on sea ice is allowed to vary between 0.001m and 1.0 m.

Cumulus parameterization is neglected in both domains to ensure all cloud processes are represented by the grid-scale microphysics scheme. Note that 5 km is a general upper limit for a convection-resolving resolution (Klemp, 2006; Prein et al., 2015). Cloud microphysics are parameterized following Morrison et al. (2005), hereafter M05. M05 performs well in reproducing Antarctic clouds, resulting in improved representation of the liquid phase and thus the cloud radiative effects (Listowski and Lachlan-Cope, 2017; Hines et al., 2019). This bulk microphysics scheme predicts mixing ratios and number concentrations for cloud water, cloud ice, rain, snow and graupel. N_{drop} is, however, specified. The default value of the scheme is 200 cm^{-3}; here N_{drop} is set to 92 cm^{-3}, which is the mean value of M218 and M219 flight measurements (see Section 2.2).

3.2 Sensitivity Simulations

A detailed description of the ice formation processes in M05 and the implemented BR parameterizations is offered in Appendix A and B, respectively. We assume that collisions that include at least one large particle (thus ice-snow, ice-graupel and graupel-snow, snow-snow and graupel-graupel) result in ice multiplication; contribution from collisions between small ice particles (cloud ice) are neglected. Additionally to the control (CNTRL) simulation,

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which corresponds to the default set-up of M05, we perform seven sensitivity simulations with varying description of F_{BR}.

In two sensitivity simulations we assume, as in Hoarau et al. (2018), a constant number of fragments generated per collision. This number is constrained by in-situ measurements from the Arctic (Schwarzenboeck et al., 2009) which indicated that one-branch ice-crystals are more common in polar clouds, resulting in ejection of a single fragment after collision with another ice particle; however this analysis (Schwarzenboeck et al., 2009) included only dendritic crystals with size larger than 300 μm. Based on these results we perform two simulations: FRAG1 assumes all collision types generate one fragment without any size restrictions, while FRAG1siz allows for ice multiplication only if the particle that undergoes fragmentation is larger than 300 μm. Note that since the separation size between ice and snow in the M05 scheme is 125 μm, collisions that include cloud ice do not result in any multiplication in FRAG1siz.

The standard temperature-dependent formula of Takahashi et al. (1995) for F_{BR}, applied in Sullivan et al. (2018b), is tested here in the TAKAH simulation. However, Takahashi et al. (1995) used 2-cm hailballs in their experiments, which is an unrealistic set-up. For this reason we perform an additional simulation, TAKAHsiz, in which this relationship is further scaled with size (see Appendix B). Finally, the Phillips parameterization is implemented in three simulations with varying Ψ for the cloud ice/snow particles that undergo fragmentation; Ψ is not predicted in most bulk micrphysics scheme, including M05, and thus it is prescribed as a constant. These simulations are referred as PHIL0.2, PHIL0.3 and PHIL0.4 in the text, where the number indicates the assumed values of Ψ.

4. Results

4.1 BR effect on microphysical properties

In Fig. 2a the modeled total ice number concentrations (cloud ice + snow + graupel, N_{isg}) derived for the region encompassing the 2 MAC flights (Fig. 1) are compared with measurements derived from the 2D Stereo (2DS) probe (see Section 2.1 for details). Since 2DS cannot detect ice particles smaller than 80 μm, only modeled ice particles with sizes larger than this threshold are considered in this figure, like in Young et al. (2019). While mean and maximum statistics are discussed below, additional statistical metrics (e.g. median and interquartile range) are shown in Fig. S1 (Text S1). The mean observed N_{isg} for the whole

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MAC campaign generally fluctuates between 0.5–4 L^{-1}. The variation in N_{isg} with temperature is somewhat larger for our case study (November 27), as maximum mean concentration goes up to ~7 L^{-1} at T= -6.5°C. The CNTRL simulation consistently underestimates the mean observations, as it produces mean N_{isg} ~0.1 L^{-1} over the examined temperature range (Fig. 2a).

PHIL0.2 and PHIL0.3 produce similar results to CNTRL (Fig. 2a, b), suggesting that lightly to moderately rimed ice particles do not contribute to ice multiplication through collisional break-up. The higher rimed fraction in PHIL0.4 results in very good agreement with mean observations (Fig. 2a), especially over the whole MAC case. FRAG1siz is also close to mean observations, but when the size restrictions are ignored (FRAG1) the model gives substantial ICNC overestimation. TAKAH simulation also produces unrealistically high mean N_{isg}, while TAKAHsiz is in closer agreement with observations. The largest deviations from observations for TAKAHsiz are observed for temperatures below -7°C; however the case study lacks good measurement statistics at this temperature range, since very few observations are available (Fig. 2a).

Overall, CNTRL, PHIL0.2 and PHIL0.3 cannot reproduce the observed spectrum (Fig. 2b). PHIL0.4, FRAG1siz and TAKAHsiz, however, can successfully reproduce the observed range of values (Fig. 2b), but their relative frequency remains underestimated for ICNCs larger than 0.5 L^{-1}. FRAG1 is in closest agreement with the observed spectrum, while TAKAH often overestimates the relative frequency (Fig. 2b). Maximum ICNCs in FRAG1 and TAKAH are 6403 and 2600 L^{-1}, respectively, which are about 70 and 30 times larger that the observed maximum value: 88 L^{-1}. This suggests that BR parameterizations that do not account for the impact of size are rather unrealistic. The maximum ICNCs in PHIL0.4, FRAG1siz and TAKAHsiz are 174 L^{-1}, 150 L^{-1} and 173 L^{-1}, which agree to within a factor of two with observations, while they are substantially underestimated in CNTRL (7.8 L^{-1}), PHIL0.2 (4.7 L^{-1}) and PHIL0.3 (5.2 L^{-1}).

Vertical distributions of cloud ice (N_i), graupel (N_g) and snow (N_s) number concentration are examined in Fig. 3(a-c) for all simulations except those that produce unrealistically large concentrations (FRAG1 and TAKAH). The observed ICNCs are also shown in Fig. 3a and 3c; for consistency with M05, the threshold size separating measured cloud ice from snow is set to 125 μm. Graupel concentrations cannot be distinguished in the measurements (hence no 'Nov 27' profile in Fig. 3b), however the more realistic model simulations suggest that these are negligible compared to cloud ice/snow concentrations.

PHIL0.2 and PHIL0.3 produce slightly larger N_i (Fig. 3a) than CNTRL, but reduced N_g (Fig. 3b) values and similar or reduced N_s (Fig. 3c); all these mean N_i and N_s profiles are orders of magnitude lower than the observed values. PHIL0.4, FRAG1siz and TAKAHsiz produce somewhat larger N_i than the observations (Fig. 3a), while N_s is somewhat underestimated (Fig. 3c). FRAG1siz is in slightly better agreement with N_i observations than the other two simulations, especially at heights above 750 m a.s.l (Fig. 3a). Activating BR generally results in reduction of N_g (Fig. 3b). This decrease is larger than one order of magnitude in the three realistic simulations, compared to CNTRL, however we cannot assess which of these graupel profiles better represents reality. Nevertheless, we can overall conclude that PHIL0.4, FRAG1siz and TAKAHsiz result in improved agreement of the vertical distribution of total ICNCs with observations compared to the rest of the simulations, including the default set-up of M05.

The simulated liquid water content (LWC) is compared with CAS observations in Fig. 4. All simulations, except TAKAH, produce similar or slightly overestimated mean LWC at temperatures ≤-3.5^{o}C; at -3^{o}C the mean observed values are higher (Fig. 4a). An overestimation of LWC in these runs is more evident in Fig. 4b; the observed spectrum does not include values larger than 0.5 g m^{-2}, while the simulated spectra are wider. An exception to this is TAKAH simulation, which underestimates mean LWCs and glaciates clouds at temperatures below -7^{o}C (Fig. 4a), while it produces a narrower LWC spectrum compared to the observed (Fig. 4b). Apart from TAKAH, the rest of the simulations produce similar liquid water properties with minor improvements in the runs with reduced LWC values, e.g. in FRAG1 (Fig. 4a). Nevertheless, while the produced range of LWC values in FRAG1 is somewhat closer to the observed, it still underestimates the relative frequency for most of the observed spectrum (Fig. 4b).

4.2 BR effect on surface radiation

To examine how deviations in ICNCs affect climate, mean radiative fluxes at the surface and at the top of the atmosphere (TOA) for all model simulations are presented in Table 1. Increasing BR multiplication has a pronounced impact on shortwave radiation, as it results in decreasing sunlight reflection and thus increasing downward surface radiation (SWD_{SFC}). Upward surface radiation (SWU_{SFC}) is a function of SWD and thus exhibits similar behaviour. This is due to the fact that increased BR efficiency (Fig. 2) results in decreased liquid water path (LWP). However, no substantial differences in mean cloud optical thickness are indicated for CNTRL (40.1 g m^{-2}), PHIL0.2 (33.2 g m^{-2}), PHIL0.3 (40.2 g m^{-2}), PHIL0.4 (29.1 g m^{-2}) and FRAG1siz (30.1 g m^{-2}), which have a mean LWP almost within the black body emission range (Stephens, 1978). Optically thinner clouds are produced in TAKAHsiz

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(23.1 g m^{-2}), and especially in FRAG1 (8.2 g m^{-2}) and TAKAH (3.2 g m^{-2}) runs. Note that most simulations, including CNTRL, produce wider LWC spectra than the observed, by overestimating cloud liquid (Fig. 4b). Generally, decreasing liquid content is in better agreement with observations (see section 4.1), suggesting that including the BR process in M05 likely shifts the simulated LWPs towards more realistic values. However, excessive ice multiplication, as in TAKAH, results in unrealistic liquid properties (Fig. 4a) and thus errors in surface radiation.

The difference between CNTRL and the simulations that improve ICNC representation (PHIL0.4, FRAG1siz and TAKAHsiz) fluctuates between 11.9-25.7 Wm^{-2} for SWD_{SFC} and 6.7-12.4 Wm^{-2} for SWU_{SFC} (Table 1). Pronounced reduction in downward longwave surface radiation (LWD_{SFC}) is only found for the simulations FRAG1siz, FRAG1 and TAKAHsiz, which have a mean LWP well below 30 g m^{-2}, the lowest limit of the black body emission range (Stephens, 1978). In all other simulations, the reduction in cloud liquid due to BR is not large enough to alter the cloud emissivity significantly. The upward longwave component (LWU_{SFC}) is only slightly affected in all simulations (<~ 1.3 Wm^{-2}). Young et al. (2019) showed that underestimation of ICNCs results in significant both positive and negative biases in the surface Cloud Radiative Forcing (CRF) over the coastal areas; our results agree with their findings, as CRF biases vary between -78 Wm^{-2} and +86 Wm^{-2} for the most realistic simulations (Fig. S2, Text S2). Furthermore, the difference between CNTRL and the realistic simulations in upward radiation flux at TOA (Table 1) is also more pronounced for the shortwave component (SWU_{TOA}), fluctuating between 4.7-9.2 Wm^{-2}, and less significant for LWU_{TOA} (1.4-3.6 Wm^{-2}). All in all, both surface and TOA radiation results indicate that a correct representation of SIP in the atmospheric models is critical for the projection of the energy budget and thus for the future Antarctic climate.

4.3 Sensitivity to uncertainties in H-M and primary ice production

The accuracy of SIP representation strongly depends on the uncertainties in the H-M description. To investigate this aspect we allow for a more efficient H-M process that is active over the whole droplet size spectrum (Fig. S3, Text S3). While this modification is not in agreement with the current knowledge on the H-M mechanism derived from laboratory studies (Hallet and Mossop, 1974; Choularton et al., 1980), it has been applied in previous modeling studies (Sinclair et al., 2016; Young et al., 2019) resulting in improved representation of the cloud ice properties. However, in these additional sensitivity tests the H-M process remains very weak and cannot explain the observed concentrations, if BR is not

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accounted for in the simulations. The BR mechanism, on the other hand, can explain the observed ICNCs even when H-M is deactivated (Fig. S3).

BR efficiency can be largely affected by the uncertainty in primary ice production, which e.g. is a factor of 10 in the applied deposition/condensation-freezing nucleation scheme (DeMott et al., 2010). Additional sensitivity tests with the Phillips parameterization for collisional break-up indicate that decreasing the efficiency of all primary ice production mechanisms by a factor of 10 inhibits BR multiplication (Fig. S4, Text S4). Increasing primary ice production by the same factor in CNTRL still results in substantially underestimated ice concentrations than the observed (Fig. S4) if BR is not accounted for, providing additional evidence for the significant role of SIP in these conditions. Activating BR in these high INP conditions produces similar ICNCs as when the standard primary ice production mechanisms are employed. These findings suggest that BR is not very sensitive to uncertainties in primary ice production, as long as there are enough primary ice crystals to initiate this process.

5. Conclusions

Our results indicate that collisional break-up can explain observations of enhanced ICNCs in coastal Antarctic clouds, in which primary ice nucleation and H-M efficiency are limited. The studied case is representative for summer low-level clouds in West Antarctica, which is the region where large discrepancies between INPs and ICNCs are known to occur within the examined temperature range (Lachlan-Cope et al., 2016; O'Shea et al., 2017). The conclusions may not hold for winter clouds in the region, which contain less supercooled liquid water (Listowski et al., 2019) and are less prone to riming, and thus may not favour BR. Nevertheless, summer clouds play a critical role in the surface melting of the ice-shelf in the vicinity of Weddell Sea (Gilbert et al., 2020) in West Antarctica and thus their accurate microphysical representation in models is of great importance.

Although BR has been observed in polar conditions before (Rangno and Hobbs, 2001; Schwarzenboeck et al., 2009), this mechanism is currently not implemented in most weather prediction and climate models. The more advanced Phillips parameterization produces realistic ICNCs in Antarctic clouds, as long as a high rimed fraction is prescribed for the particles that undergo fracture, in agreement with Sotiropoulou et al. (2020). A comparison of vapor deposition rates with riming rates (which include mass changes due to collisions with droplets/raindrops and due to contact/immersion freezing) for CNTRL simulation indicate that these two are on average comparable for cloud ice, while riming rates are substantially

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larger than vapor deposition rates for snow (not shown). These results indicate that prescribing a high rimed fraction for cloud ice and snow in M05 is not unreasonable; nevertheless Ψ in reality is highly variable for different temperature and microphysical conditions. More simplified parameterizations also produce improved results as long as the impact of the dependence of F_{BR} on the ice particle size is accounted for.

The very few existing BR descriptions in mesoscale models either do not account for size limitations (Sullivan et al., 2018b) or do not account for all collision types (Hoarau et al., 2018), which limits their realism. Increasing ICNCs from BR alters significantly the radiative effects of summer Antarctic clouds, suggesting that an accurate representation of this process in models can impact the future projection of regional climate.

Appendix A: Ice formation processes in M05 scheme

The standard M05 scheme includes three primary ice production mechanisms (immersion freezing, contact freezing and deposition/condensation-freezing nucleation), and one SIP process (H-M). Immersion freezing of cloud droplets and rain is parameterized after Bigg (1953). This mechanism is active below -4°C and produces a raindrop freezing rate that depends on the degree of supercooling and the number concentration and volume of supercooled drops. Meyers et al. (1992) description is used for contact freezing, also active below -4°C, but the rates are further weighted by the effective diffusivity of the contact nuclei.

The default parameterization for deposition/condensation-freezing ice nucleation in M05 is Cooper (1986), which depends only on temperature and is active below -8°C in liquid saturated conditions or when ice supersaturation exceeds 8%. However, the DeMott et al. (2010) parameterization for heterogeneous nucleation has been shown to compare better with in-cloud ice measurements over the Antarctic Peninsula than Cooper (Listowski and Lachlan-Cope, 2017). For this reason, we apply the DeMott description in our simulations, where the mean aerosol concentration of particles with sizes between 0.5-1.6 μm for the two flights (0.49 scm^{-3}) is used as input (Young et al., 2019).

The H-M description, adapted from Reisner et al. (1998), is based on the laboratory experiments conducted by Hallett and Mossop (1974), who found a maximum of ~350 splinters per milligram of rime generated around -5°C:

$$\frac{dNi_{HM}}{dt} = \rho\, 3.5 \cdot 10^{8}\, f(T) \frac{dm_{rime}}{dt} \quad (1)$$

where dNi_{HM}/dt is the number of new fragments produced at a given timestep, $f(T)$ is the

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temperature-dependent efficiency of the process, ρ is the air density, and dm_{rime}/dt is the mass production rate of rime on snow or graupel due to accretion of cloud and rain drops. $f(T)$ is 0 for $T < -8^oC$ and $T > -3^oC$, 1 for $T = -5^oC$, and increases linearly between these two extremes for $T \geq -8^oC$ and $T \leq -3^oC$.

Furthermore, for H-M to become activated in M05, two conditions must be met: (a) snow (or graupel) mass mixing ratios must be greater than 0.1 g kg^{-1} and (b) cloud liquid (or rain) water mass mixing ratios shoud be greater than 0.5 (or 0.1) g kg^{-1}. To achieve a good agreement between modeled and observed ICNCs for the simulated case, Young et al. (2019) had to remove condition (b) and multiply the H-M efficiency by a factor of 10.

Appendix B: Parameterizing collisional break-up in M05

There are three types of ice particles considered in the M05 scheme: small (cloud) ice, snow, and graupel. Ice multiplication is allowed after cloud ice-snow, cloud ice-graupel, graupel-snow, snow-snow and graupel-graupel collisions. The standard M05 scheme includes a description for collisions between cloud ice and snow to represent the accretion process, following the "continuous collection" approach:

$$\frac{dN_{i_{AC}}}{dt} = \frac{\pi}{4}\rho E_{col}\,\Gamma(b_s+3)\,a_s\,\frac{N_i N_{0s}}{\lambda_s^{(b_s+3)}} \quad (2)$$

dN_{iAC}/dt is the rate of ice crystal number concentration collected by snow. N_{0S} and λ_s are the intercept and slope parameters of the snow size distribution, represented by an inverse exponential function, and Γ is the Euler gamma function. a_s and b_s are the characteristic parameters for snow in the fallspeed-diameter relationship (Morrison et al., 2005); a_s includes a density correction factor (Heymsfield et al., 2007). Note that the diameter (d_i) and terminal velocity (u_i) of cloud ice particles are considered much smaller than those of snow: $d_i << d_s$ and $u_i << u_s$, so that they are neglected in Eq. (2). E_{col} is the collection (sticking) efficiency between ice particles, set to 0.1; hence, it is assumed that only 10% of cloud ice particles that collide with snow are actually collected. We assume the remaining 90% of collisions result in ice particle break-up, hence the following relationship gives the rate of cloud ice-snow collisions that contribute to ice multiplication:

$$\frac{dN_{i_{is}}}{dt} = \frac{\pi}{4}\rho(1-E_{col})\,\Gamma(b_s+3)\,a_s\,\frac{N_i N_{0s}}{\lambda_s^{(b_s+3)}} \quad (3)$$

In the default M05, collisions between cloud ice and graupel particles are neglected as it is assumed that the collection efficiency of such collisions is negligible. To represent cloud ice-graupel collisions for ice multiplication, we use Eq. (3), but the size distribution and

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fallspeed parameters of snow are replaced by those for graupel. Moreover, since cloud ice is not collected by graupel particles, we assume that 100% of these collisions result in cloud ice break-up:

$$\frac{dN_{i_{ig}}}{dt} = \frac{\pi}{4}\rho\Gamma\left(b_g+3\right) a_g \frac{N_i N_{0g}}{{\lambda_g}^{(bg+3)}} \quad (4)$$

In the default M05 scheme, collisions between snow and graupel are also neglected because it is assumed that the collection efficiency for such collisions is negligible. For this study, graupel-snow collisions are treated using expressions similar to those for raindrop-snow collisions in M05. These are adapted from Ikawa and Saito (1991) and represent collisions between relatively large precipitation-sized particles:

$$\frac{dQ_{i_{sg}}}{dt} = \pi^2\rho_s\rho\left|\Delta u_{m_{sg}}\right|\frac{N_{0s}N_{0g}}{\lambda_s^3}\left(\frac{5}{\lambda_s^3\lambda_g}+\frac{2}{\lambda_s^2\lambda_g^2}+\frac{0.5}{\lambda_s\lambda_g^3}\right) \quad (5)$$

$$\frac{dN_{i_{sg}}}{dt} = \frac{\pi}{2}\rho\left|\Delta u_{n_{sg}}\right| N_{0s}N_{0g}\left(\frac{1}{\lambda_s^3\lambda_g}+\frac{1}{\lambda_s^2\lambda_g^2}+\frac{1}{\lambda_s\lambda_g^3}\right) \quad (6)$$

where $$\left|\Delta u_{m_{sg}}\right| = \left(\left(1.2u_{ms}-0.95u_{mg}\right)^2+0.08u_{mg}u_{ms}\right)^{1/2} \quad (7)$$

and $$\left|\Delta u_{n_{sg}}\right| = \left(\left(1.7u_{ns}-u_{ng}\right)^2+0.3u_{ng}u_{ns}\right)^{1/2} \quad (8)$$

dQ_{isg}/dt and dN_{isg}/dt represent the bulk rates that snow mass and number concentration collide with graupel and contribute to ice multiplication through fragmentation. Corrections in the mass (or number) -weighted difference in terminal velocity $\Delta u_{m_{sg}}$(or $\Delta u_{n_{sg}}$) of the colliding particles (Eq. 7,8) are adapted from Mizuno (1990) and Reisner et al. (1998), to account for underestimates when $u_{ns} \approx u_{ng}$.

M05 also includes a description for collisions between snowflakes to represent snow aggregation, following Passarelli (1978):

$$\frac{dN_{s_{AG}}}{dt} = \frac{-1108a_sE_{col}}{4\times720}\pi^{\frac{1-b_s}{3}}\rho^{\frac{2+b_s}{3}}\rho_s^{\frac{-2-b_s}{3}}Q_s^{\frac{2+b_s}{3}}N_s^{\frac{4-b_s}{3}} \quad (9)$$

Based on this expression we parameterize the number of snow-snow collisions that contribute to ice multiplication as:

$$\frac{dN_{i_{ss}}}{dt} = \frac{1108a_s(1-E_{col})}{4\times720}\pi^{\frac{1-b_s}{3}}\rho^{\frac{2+b_s}{3}}\rho_s^{\frac{-2-b_s}{3}}Q_s^{\frac{2+b_s}{3}}N_s^{\frac{4-b_s}{3}} \quad (10)$$

Because snow aggregation does not result in any mass transfer, the snow mass involved in these collisions is not calculated by the default M05 scheme. We obtain a description of dQ_{iss}/dt by applying the size distribution and fallspeed parameters of snow in the analytical solution for self-collection derived by Verlinde et al. (1990):

$$\frac{dQ_{i_{ss}}}{dt} = \frac{914\pi^2}{48\rho\rho_s}(1-E_{col})a_s d_s^{b_s+5}N_s^2 \quad (11)$$

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To test the consistency of Eq. (10) and (11), which were derived using different methods, we repeated the CNTRL and PHIL0.4 simulations but with the Eq. (9) and (10) replaced by the analytical solution for the change in number concentration from self-collection derived by Verlinde and Cotton (1993). The sensitivity of the results to this modification was found to be insignificant.

Graupel-graupel collisions are also parameterized in a similar manner. Since there is no graupel aggregation (collection efficiency of such collisions is assumed to be negligible), 100% of the collisions are assumed to contribute to break-up:

$$\frac{dN_{igg}}{dt} = \frac{1108 a_g}{4\times720}\pi^{\frac{1-b_g}{3}}\rho^{\frac{2+b_g}{3}}\rho_g^{\frac{-2-b_g}{3}}Q_g^{\frac{2+b_g}{3}}N_g^{\frac{4-b_g}{3}} \quad (12)$$

$$\frac{dQ_{igg}}{dt} = \frac{836\pi^2}{48\rho\rho_g}a_g d_g^{b_g+5}N_g^2 \quad (13)$$

The value 1108 in Eq. (10) is valid for b_s=0.4 (Passarelli, 1978); in M05 b_s=0.41 and b_g=0.37, thus adapting this value for both snow-snow (10) and graupel-graupel (12) collisions is a reasonable approximation.

Following the methodology of Sullivan et al. (2018b) in TAKAH simulation, the number of fragments generated due to ice-ice particle collisions (F_{BR}) is:

$$F_{BR} = 280\,(T-252)^{1.2}e^{-(T-252)/5} \quad (14)$$

However, Takahashi et al. (1995) used 2-cm hailballs in their experiments, thus to further include the influence of size in this formulation, we implement a size-scaled expression in TAKAHsiz simulation, assuming that F_{BR} depends linearly on D, decreasing to 0 at D = 0:

$$F_{BR} = 280\,(T-252)^{1.2}e^{-(T-252)/5}\frac{D}{D_o} \quad (15)$$

where D (in meters) is the size of the ice particle that undergoes fracturing and D_o=0.02 m, the size of haiballs used by Takahashi et al. (1995) .

The Phillips et al. (2017a) parameterization allows for varying treatment of F_{BR} depending on the ice crystal type and habit.

$$F_{BR} = \alpha A\left(1 - exp\left\{-\left[\frac{CK_o}{\alpha A}\right]^{\gamma}\right\}\right) \quad (16)$$

$$\text{where :}\ K_o = \frac{m_1 m_2}{m_1+m_2}\left(\Delta u_{n_{12}}\right)^2,$$

$$\psi = 3.5\times10^{-3}$$

$$a = \pi D^2$$

m_1, m_2 are the masses of the colliding particles and Δu_{n12} is the difference in their terminal velocities. The correction applied in Eq. (8) is also adapted here to account for underestimates when $u_{n1} \approx u_{n2}$. D (in meters) is the size of the smaller ice particle which undergoes

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fracturing and α is its surface area. The parameterization was developed based on particles with diameters 500 μm < D < 5 mm, however Phillips et al. (2017a) suggest that it can be used for particle sizes outside the recommended range as long as the input variables to the scheme are set to the nearest limit of the range. C is the asperity-fragility coefficient and ψ is a correction term for the effects of sublimation based on the field observations by Vardiman (1978). For cloud ice-snow, cloud ice-graupel, snow-graupel and snow-snow collisions:

$$A = 1.58 \cdot 10^7 (1 + 100\Psi^2)\left(1 + \frac{1.33 \cdot 10^{-4}}{D^{1.5}}\right),$$

$$\gamma = 0.5 - 0.25\Psi,$$

$$C = 7.08 \times 10^6 \psi$$

The above parameters adapted from Phillips et al. (2017a) concern planar crystals or snow with rimed fraction $\Psi < 0.5$ that undergo fracturing: $\Psi \leq 0.2$ corresponds to lightly rimed particles, while $\Psi \approx 0.4$ represents highly rimed crystals/snow. The choice of the ice habit is based on particle images collected during the MAC flights, which indicate the presence of needles and planar particles (O'Shea et al., 2017); needles are often considered secondary ice (Field et al., 2017). However, the M05 scheme does not explicitly consider habit and assumes spherical particles for all processes except sedimentation, for which the fallspeed-diameter relationships are for non-spherical ice.

For graupel-graupel collisions the parameters implemented in Eq. (16) are somewhat different (Phillips et al., 2017a):

$$A = \frac{a_o}{3} + \max\left(\frac{2a_o}{3} - \frac{a_o}{9}|T - 258|, 0\right)$$

$$\gamma = 0.3,$$

$$C = 6.3 \times 10^6 \psi$$

Finally, an upper limit for the number of fragments produced per collision is imposed, set to $F_{BR_{max}} = 100$; this is the same for all collision types (Phillips et al., 2017a).

We estimate the production rate of fragments for cloud ice-snow collisions and cloud ice -graupel collisions using Eq. (3) or (4) and one of the proposed formulations for F_{BR} above: $\frac{dN_{i_{is}}}{dt} F_{BR}$ and $\frac{dN_{i_{ig}}}{dt} F_{BR}$. For both of these collision types we assume that the cloud ice particles undergo break-up and the new smaller ice fragments remain within the same ice particle category. For snow-graupel collisions, where the snow particle is assumed to undergo fracturing, the production term $\frac{dN_{i_{sg}}}{dt} F_{BR}$ is added to the cloud ice category. In this case mass transfer from the snow to the cloud ice category also occurs, but according to Phillips et al. (2017a) this is only 0.1% of the snow mass that collides with graupel (5). Snow-snow and

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graupel-graupel collisions are handled in the same way: $\frac{dN_{i_{ss}}}{dt} F_{BR}$ and $\frac{dN_{i_{gg}}}{dt} F_{BR}$ are added to the cloud ice number equation, while 0.1% of $\frac{dQ_{i_{ss}}}{dt}$ (11) and $\frac{dQ_{i_{gg}}}{dt}$ (13) is added to the corresponding mass equation.

Code and data availability: MAC data are available at https://catalogue.ceda.ac.uk/uuid/da17dab196f74d64af3ccbc35624027b. The modified Morrison scheme is available upon request

Competing interests: The authors declare that they have no conflict of interest.

Author contribution: GS and AN conceived and led this study. EV helped with the model configuration and set-up, and provided Fig. 1. GY provided the observations and the model set-up for the MAC case. SJO post-processed MAC data. GS implemented the BR parameterizations, performed the WRF simulations, analyzed the results and, together with AN, led the manuscript writing. All authors contributed to the scientific interpretation, discussion and writing of the manuscript.

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Tables:

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Preprint. Discussion started: 25 May 2020
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Table 1: Mean modeled downward and upward shortwave (SWD_{SFC}, SWU_{SFC}) and longwave (LWD_{SFC}, LWU_{SFC}) surface radiation, along with upward shortwave and longwave (SWU_{TOA}, LWU_{TOA}) radiation at the top of the atmosphere, during flights M218 and M219. Model
865 results are averaged over the dashed rectangular area in Fig. 1.

Simulation	**SWD_{SFC} (Wm^{-2})**	**SWU_{SFC} (Wm^{-2})**	**LWD_{SFC} (Wm^{-2})**	**LWU_{SFC} (Wm^{-2})**	**SWU_{TOA} (Wm^{-2})**	**LWU_{TOA} (Wm^{-2})**
CNTRL	323.9	182.1	244.3	304.6	255.8	218.4
PHIL0.2	328.6	184.5	244.1	304.6	254.8	218.5
PHIL0.3	322.3	181.0	247.4	305.3	256.6	217.9
PHIL0.4	339.7	190.8	243.3	304.9	251.1	219.8
FRAG1	354.1	198.6	236.7	303.8	246.9	221.5
FRAG1siz	335.7	188.8	244.0	304.6	250.5	220.7
TAKAH	365.9	206.5	229.8	303.3	242.5	221.2
TAKAHsiz	349.5	194.5	237.0	304.2	246.6	222.0

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Figures:

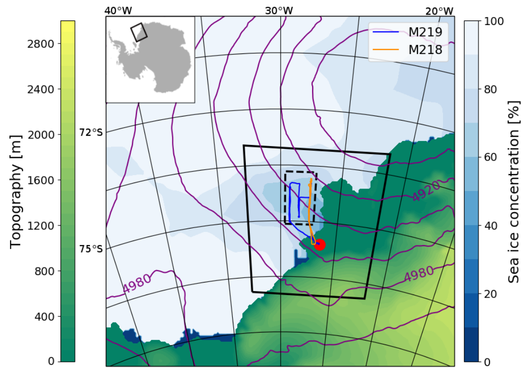

Figure 1: Map of Antarctic domains. Colors indicate terrain heights (green to yellow) and sea-ice concentrations (blue to white), whereas the purple contours correspond to 500 hPa geopotential heights from the CNTRL simulation at 18:00 UTC, 27 November 2015. The black solid line delimits the 1-km horizontal grid spacing domain, while the dashed one outlines the subset of the nest used for direct comparison with the aircraft data. Orange and blue lines indicate the flight tracks, while the red circle represents Halley station. The small figure in the top right corner indicates the location of the 1-km horizontal grid spacing domain relative to the Antarctic continent.

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Atmospheric Chemistry and Physics Discussions

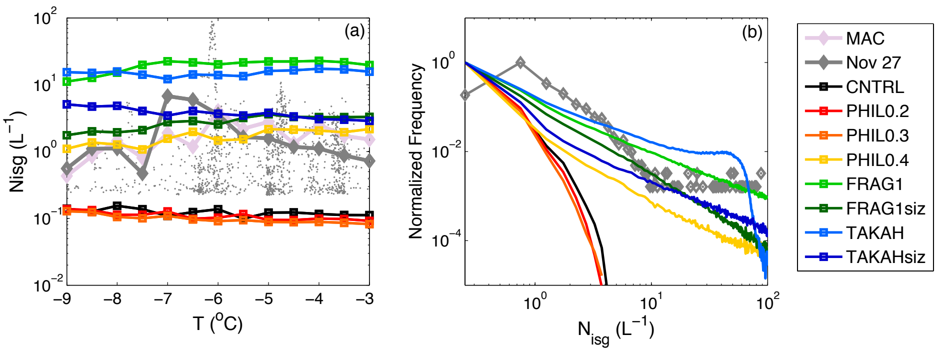

Figure 2: (a) Mean ice number concentrations (cloud ice+snow+graupel, N_{isg}) as a function of temperature for the whole MAC campaign (pink), our case study (grey) and the eight model simulations. Grey dots indicate point observations. (b) Relative frequency distribution of N_{isg}, binned in 0.5 L^{-1} intervals, scaled with maximum frequency. Ice properties are calculated for particles > 80 µm and for N_{isg} > 0.005 L^{-1} within the lowest 1.5 km a.s.l.

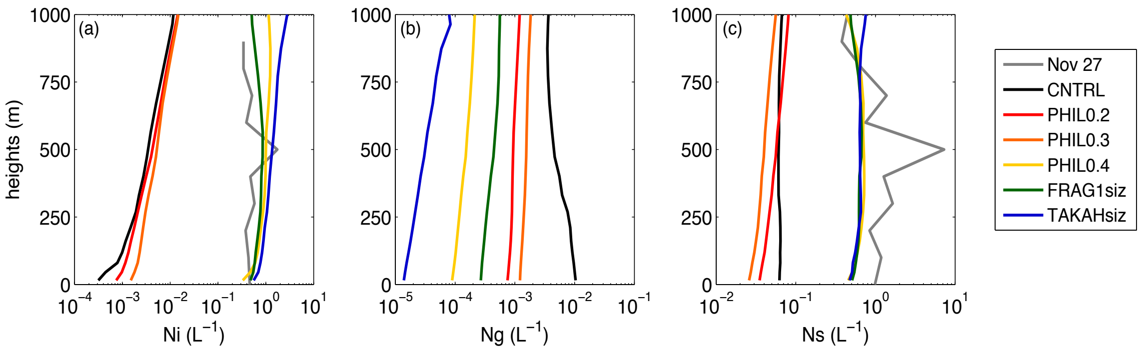

Figure 3: Mean vertical profiles of number concentrations of modeled (a) cloud ice, (b) graupel and (c) snow for six simulations. Grey lines represent measured concentrations with diameters (a) smaller and (c) larger than 125 µm. Ice properties from the model are calculated for N_{isg}>0.005 L^{-1}. For consistency with observations, only particles with sizes > 80 µm are included in the modeled profiles in panels (a) and (c).

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Atmospheric Chemistry and Physics
Discussions

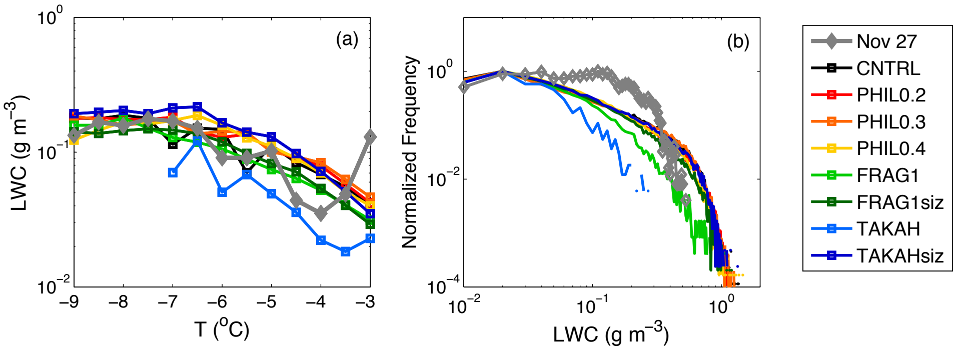

Figure 4: (a) Mean liquid water content (LWC) as a function of temperature for our case study (grey) and the eight model simulations. (b) Relative frequency distribution of LWC, binned in 0.01 g m^{-3} intervals, scaled with maximum frequency. Only values greater than 0.01 g m^{-3} within the lowest 1.5 km a.s.l. are included in the analysis.