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Georgia Sotiropoulou¹, Anna Lewinschal², Paraskevi Georgakaki³, Vaughan Phillips⁴, Sachine Patade⁵, Annica M. L. Ekman⁶, and Athanasios Nenes³

¹ICE-HT, Foundation for Research and Technology Hellas (FORTH), Patras, Greece

²Department of Meteorology, Stockholm University & Bolin Center for Climate Research, 9 Sweden

³Laboratory of Atmospheric Processes and their Impacts (LAPI), Ecole Polytechnique Fédérale de Lausanne (EPFL), Lausanne, Switzerland

⁴Department of Physical Geography

⁵Department of Physical Geography, University of Lund, Lund, Sweden

⁶Department of Meteorology, Stockholm University

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Georgia Sotiropoulou^{1,3*}, Anna Lewinschal², Paraskevi Georgakaki³, Vaughan Phillips⁴, Sachin Patade⁴, Annica M. L. Ekman², Athanasios Nenes^{1,3}

¹ICE-HT, Foundation for Research and Technology Hellas (FORTH), Patras, Greece

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

³Laboratory of Atmospheric Processes and their Impacts (LAPI), Ecole Polytechnique Fédérale de Lausanne (EPFL), Lausanne, Switzerland

⁴Department of Physical Geography, University of Lund, Lund, Sweden

**Currently at the Department of Physics, Sector of Environmental Physics & Meteorology, National & Kapodistrian University of Athens, Greece*

Corresponding author: georgia.sotiropoulou@epfl.ch , athanasios.nenes@epfl.ch

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Abstract

Ice formation remains one of the most poorly represented microphysical processes in climate models. While primary ice production (PIP) parameterizations are known to have a large influence on the modeled cloud properties, the representation of secondary ice production (SIP) is incomplete and its corresponding impact is therefore largely unquantified. Furthermore, ice aggregation is another important process for the total cloud ice budget, which also remains largely unconstrained. In this study we examine the impact of PIP, SIP and ice aggregation on Arctic clouds, using the Norwegian Earth System model version 2 (NorESM2). Simulations with both prognostic and diagnostic PIP show that heterogeneous freezing alone cannot reproduce the observed cloud ice content. The implementation of missing SIP mechanisms (collisional break-up, drop-shattering and sublimation break-up) in

NorESM2 improves the modeled ice properties, while improvements in liquid content occur only in simulations with prognostic PIP. However, results are sensitive to the description of collisional break-up. This mechanism, which dominates SIP in the examined conditions, is very sensitive to the treatment of the sublimation correction factor, a poorly-constrained parameter that is included in the utilized parameterization. Finally, variations in ice aggregation treatment can also significantly impact cloud properties, mainly through its impact on collisional break-up efficiency. Overall, enhancement in ice production through the addition of SIP mechanisms and the reduction of ice aggregation (in line with radar observations of shallow Arctic clouds) result in enhanced cloud cover and decreased TOA radiation biases, compared to satellite measurements, especially during the cold months.

Significance

Arctic clouds remain a large source of uncertainty in projections of the future climate due to the poor representation of the microphysical processes that govern their life cycle. Ice formation is among the least understood processes. While it is widely recognized that better constraints on primary ice production (PIP) are needed to improve existing parameterizations, we show that secondary ice production (SIP) and ice aggregation can have also a significant impact on the ice number concentrations. Constraining ice formation through the addition of missing SIP mechanisms and reducing ice aggregation can improve the representation of the cloud macrophysical properties and enhance total cloud cover in the Arctic region, which in turn contributes to decreased TOA radiation biases in the cold months.

1. Introduction

Clouds and cloud feedbacks remain the largest source of uncertainty in predictions of the future climate (Boucher et al. 2013). In the most recent Climate Model Intercomparison Project (phase 6 – CMIP6) many general circulation models (GCMs) exhibited larger sensitivity to changes in carbon dioxide concentrations, a metric known as Equilibrium Climate Sensitivity (ECS), compared to CMIP5 models (Zelinka et al. 2020). Murray et al. (2021) showed that ECS values in CMIP6 correlate with mid-to-high latitude low-level cloud feedbacks. Moreover, CMIP6 models suffer from biases in high-latitude cloud cover (Vignesh et al. 2020), cloud radiative impacts (Sledd and L'ecuyer 2020) and snowfall patterns (Thomas et al. 2019).

Mixed-phase clouds, consisting of both supercooled liquid and ice, are the most

abundant Arctic cloud type at temperatures between -25°C and 0°C (Shupe et al. 2006; 2011). While these clouds are theoretically thermodynamically unstable and can easily glaciate through the Wegener-Bergeron-Findeisen (WBF) mechanism, they have been observed to persist for days to weeks (Morrison et al. 2012). Moreover, as ice crystals grow through vapor deposition, they can start forming aggregates through collisions with other ice particles or they can gain mass through the collection of liquid droplets (i.e. riming) until they eventually fall out in the form of snow or graupel. Mixed-phase cloud observations often indicate that the supercooled liquid layer is concentrated near cloud top with ice particles falling below, which allows the liquid phase to be maintained (Morrison et al. 2012). Modeling the life-cycle of these clouds is challenging since errors in the representation of the complex processes that maintain them can lead to rapid glaciation. At the same time a correct representation of the vertical structure and cloud phase is crucial for an accurate estimation of the cloud radiative impact (Curry et al. 1996). Predictions of Arctic warming are particularly sensitive to cloud ice formation (Tan et al. 2019). While ice formation processes are likely an important contributor to the CMIP6 spread in predicted mid- and high-latitude cloud feedbacks (Murray et al. 2021), they remain among the most poorly understood microphysical processes in mixed-phase clouds (Seinfeld et al. 2016; Storelvmo 2017).

Primary ice production (PIP) at temperatures above -38°C can only happen heterogeneously in the atmosphere, which means that the assistance of insoluble aerosols that act as Ice Nucleating Particles (INPs) is required (Hoose and Möhler 2012). However, primary ice crystal concentrations can further be enhanced through multiplication processes (Field et al. 2017; Korolev and Leisner 2020), known as secondary ice production (SIP). SIP has received substantially less attention than PIP in the past decades, which is the reason behind its poor (or absent) representation in atmospheric models. Several observational (Gayet et al. 2009; Lloyd et al. 2015; Luke et al. 2021; Pasquier et al. 2022) and modeling (Sotiropoulou et al. 2020; 2021b; Zhao et al. 2021; Zhao and Liu 2021; 2022) studies have indicated that SIP might be particularly important for Arctic clouds, as INP concentrations in the Arctic region are generally low (Wex et al. 2020) to account for the high ice crystal number concentrations (ICNCs) observed (Hobbs and Rangno 1998).

Several mechanisms that can trigger ice multiplication have been identified in laboratory experiments (Korolev and Leisner 2020), however only one SIP mechanism has until now been considered in GCMs: the Hallett-Mossop (HM) process (Hallett and Mossop, 1974). This is also the case for the Norwegian Earth System model version 2 (NorESM2),

which allows HM to occur after cloud drop-snow collisions. However, observational (Rangno and Hobbs 2001; Schwarzenboeck et al. 2009; Luke et al. 2021) and modeling studies (Sotiropoulou et al. 2020; 2021b; Zhao et al. 2021; Zhao and Liu 2021; 2022) suggest that other SIP processes, like collisional break-up (Vardiman 1978; Takahashi et al. 1995) and drop-shattering (Lauber et al. 2018; Keinert et al. 2020), also have a significant influence on Arctic cloud microphysical structure.

In this study we implement descriptions for drop-shattering (DSH) and collisional break-up (BR) in NorESM2, using parameterizations from the recent literature (Phillips et al. 2017a,b; 2018). We further test the efficiency of sublimation break-up (SUBBR) (Oraltay and Hallett 1989; Bacon et al. 1998), a process whose efficiency remains unknown in Arctic atmospheric conditions, using the parameterization developed by Deshmukh et al. (2022). In addition, we modify the existing HM description to further account for rain-snow collisions. Sensitivity simulations with varying PIP, SIP and ice aggregation treatment are conducted to quantify the ice-related processes that are most impactful on ice particle number. Results are initially evaluated against two-year surface-based observations from Ny-Ålesund for the period June 2016 - May 2018 to assess the most realistic simulation set-up. Satellite radiation and cloud measurements are further used to quantify the impact of the examined processes on the current climate state over the whole Arctic region.

2. Methods

a. Observations

Field observations of clouds were collected at Ny-Ålesund in 2016–2018 in the context of the Arctic Amplification: Climate Relevant Atmospheric and Surface Processes, and Feedback Mechanisms (AC)³ project. With the addition of a W-band cloud radar, this observation site became one of the few Arctic sites capable of state-of-the-art long-term cloud profiling with high temporal and spatial resolution. A detailed analysis of the observed cloud properties is offered by Nomokonova et al. (2019; 2020). The total occurrence of clouds was found to be ~81%. The most predominant type of clouds was multi-layer clouds with a frequency of occurrence of 44.8%. Single-layer clouds occurred 36%, with the vast majority of them being mixed-phase; liquid hydrometeors were generally observed within the lowest two kilometers in the atmosphere.

Below the measurements utilized to evaluate the model are described. Macro- and micro- physical cloud properties are derived from a combination of instruments that includes a 94 GHz cloud radar, a ceilometer and a HATPRO radiometer (Nomokonova et al. 2019d). The cloud Liquid water path (LWP) is derived from a HATPRO microwave radiometer (Nomokonova et al. 2019a,b,c) with typical uncertainty around $\pm 20\text{--}25 \text{ g m}^{-2}$, using a multivariate linear regression algorithm developed at the University of Cologne (Löhnert and Crewell 2003). HATPRO cannot provide reliable estimates under rainy conditions, when the instrument radome becomes wet; data flagged for precipitation are excluded from this analysis. Thermodynamic variables such as temperature (Nomokonova et al. 2019d,e,f) and integrated water vapor (IWV; Nomokonova et al. 2019g,h,i) are also derived from HATPRO.

Once the Cloudnet retrieval algorithm (Illingworth et al. 2007) has been applied to categorize the measured particles as liquid droplets, ice, melting ice, and drizzle/rain, ice water content (IWC) is derived from radar reflectivity and temperature measurements following the methodology of Hogan et al. (2006). The uncertainties in this IWC retrieval range from -33% to +50% for temperatures above -20°C and from -50% to +100% for temperatures below -40°C . The effective radius of ice particles (r_{ieff}) is calculated following Delanoë and Hogan (2010), using IWC and visible extinction coefficient estimates (Ebell et al. 2020); the latter is also derived following Hogan et al. (2006). The uncertainty in r_{ieff} retrieval described by Delanoë and Hogan (2010) is about 30%, while the uncertainty for the radar-derived visible extinction coefficient that is used in the ice effective radii retrieval is 62% to 160% (Hogan et al. 2006). de Boer et al. (2009) reported that assumptions in the shape of ice particles might result in a $200 \text{ }\mu\text{m}$ uncertainty in r_{ieff} estimations that are based on cloud radar and lidar techniques.

Surface in-situ cloud measurements were collected at the Zeppelin station, on mount Zeppelin near Ny-Alesund town, with the Zeppelin Observatory counterflow virtual impactor (CVI) inlet and a fog monitor (Droplet Measurement Technologies Inc., USA, Model FM-120) (Karlsson et al. 2021a,b) for a similar period (until February 2018) as the remote sensing observations. CVI and FM-120 number concentrations show very good agreement, except for a small portion of the data (7-8%) that are characterized by overestimated CVI values (Karlsson et al. 2021a). These deviations have been attributed to either varying sampling efficiencies, physical processes or spurious measurements (Karlsson et al. 2021a). FM-120 is used in our study to evaluate the modeled cloud particle spectrum over a limited size range, since these instruments measure only small cloud particles with diameters below $50 \text{ }\mu\text{m}$.

Finally, since Ny-Alesund conditions differ from those observed at other pan-Arctic sites and in the central Arctic, in terms of both thermodynamic (e.g. Naaka et al. 2018) and aerosol (e.g. Schmeisser et al. 2018) properties, this is expected to lead in a variable impact of the examined processes across the Arctic. For this reason, local measurements are complemented with satellite datasets to evaluate the modeled radiation and cloud characteristics over the whole Arctic region. These include the Clouds and Earth's Radiant Energy Systems (CERES; Wielicki et al. 1996) Energy Balanced and Filled (EBAF) product, edition 4.1 (Kato et al. 2018) and the GCM-Oriented CALIPSO Cloud Product (GOCCP) Version 3 (Chepfer et al. 2010).

b. Model description

For our investigations we use the NorESM2-MM version (Selund et al. 2021) with approximately 1° horizontal resolution (development branch). Wind and pressure fields are nudged towards ERA-Interim profiles to limit the influence of meteorological errors on microphysical fields. The relaxation time for nudging is set to 6 hours, same as the time resolution of the reanalysis data (Dee et al. 2011). Simulations are run for 29 months, from 1 January 2016 to 31 May 2018, with fixed sea-surface temperatures (SSTs). The first five months are considered as spin-up, while the rest of the data are used for comparison with surface-based observations from Ny-Ålesund. A description of the modeled ice microphysics, which is the main focus of this study, and the implemented modifications follow below.

The atmospheric component of NorESM2 is CAM6-Oslo, which consists of the Community Atmosphere Model version 6 (CAM6) and the OsloAero5.3 (Kirkevåg et al. 2018) aerosol scheme. CAM6-Oslo employs the Morrison and Gettelman (2015) microphysics scheme (MG2), which accounts for four hydrometeor types: cloud droplet, raindrop, cloud ice and snow. Heterogeneous PIP parameterizations follow the Classical Nucleation Theory (CNT; Hoose et al. 2010; Wang et al. 2014) which accounts for immersion, contact and deposition freezing of two INP species, dust and soot. Immersion freezing is only allowed to occur below -10°C in this scheme for both INP species, while only 10% of the soot concentrations are considered efficient INPs. While CNT is the default nucleation scheme used in CMIP6, the model employs an alternative option for PIP: CNT can be replaced by diagnostic parameterizations that are a function of basic thermodynamic variables and do not account for explicit cloud-aerosol interactions. These include the Bigg (1953), Young (1974) and Meyers et al. (1992) parameterizations for immersion, contact and

deposition freezing, respectively. The Bigg (1953) and Young (1974) parameterizations are activated at temperatures below -4°C , while Meyers et al. (1992) is active within the -37°C – 0°C temperature range.

Secondary ice production is accounted in MG2 scheme only through the HM mechanism, which is parameterized following Cotton et al. (1986). This formulation considers a maximum splinter production of 350 splinters per milligram of rime at -5°C , while the process efficiency decreases to zero at temperatures below (above) -8°C (-3°C). However, HM is only activated after cloud droplets collide with snow; in our modified code, we further account for the contribution from raindrop-snow collisions, following Morrison et al. (2005) scheme, using the same parameterization (Cotton et al. 1986) for the prediction of the generated fragments. Estimations of mass and number collision tendencies for raindrop-snow collisions are available in the standard MG2 scheme.

To represent the BR mechanism, we implement the parameterization of Phillips et al. (2017a). The process is initiated after snow particles collide with each other or with cloud ice. We assume that the collisions that do not result in sticking (aggregation) at an instant timestep, can bounce to initiate the break-up. Phillips et al. (2017a) is a physically-based parameterization that predicts the number of generated fragments as a function of collisional kinetic energy, while the effect of the colliding particles' size, rimed fraction and ice habit is further accounted. MG2 however does not predict rimed fraction and ice habit. For this reason, in our simulations planar ice particles with a 0.4 rimed fraction are assumed; planar shape accounts for a larger range of shapes and is valid for a wider temperature range, while a high fraction has been shown to give the most optimal results in simulations of polar clouds (Sotiropoulou et al. 2020; 2021a). All generated fragments from this mechanism are added to the cloud ice category.

The DSH description follows Phillips et al. (2018) and is initiated after raindrop-INP (immersion freezing), raindrop-snow and raindrop-ice collisions. For ice multiplication due to raindrop-INP and raindrop-cloud ice collisions we utilize the formulation referred as 'mode 1' in Phillips et al. (2018), which concerns the accretion of small particles by more massive raindrops, while for snow-raindrop the 'mode 2' formulation is applied. Mode 1 can generate both tiny and big fragments; the former are added to the cloud ice category, while the latter is considered snow. The new tiny fragments are assumed to have a fixed diameter of 10^{-5} m (Phillips et al. 2018) and a constant ice density of 500 kg m^{-3} (which is the default cloud ice density in the MG2 scheme), while the rest of the colliding rain mass is transferred to snow.

Freezing probability in this mode is set to unity and zero, at temperatures below -6°C and above -3°C , respectively, while it takes intermediate values at temperatures between -6°C and -3° . Similarly, the shattering probability is a function of raindrop size, set to 0 and 1 at sizes smaller than $50\text{ }\mu\text{m}$ and larger than $60\text{ }\mu\text{m}$, respectively. Mode 2 can only generate tiny fragments. Tiny fragments are added to the cloud ice category, while big fragments are treated as snow.

Deshmukh et al. (2022) recently developed an empirical formulation for sublimation break-up of graupel and dendritic snow, in which the total number of the ejected fragments (N) is proportional to the square root of the sublimated mass (M), $N = KM^{0.57}$, where K is a function of size (diameter) and relative humidity with respect to ice. Since graupel is not accounted in the MG2 scheme, we apply this parameterization to sublimating snow and cloud ice, as long as the diameter for the latter exceeds $200\text{ }\mu\text{m}$ (note that the cloud-ice to snow autoconversion diameter is set to $500\text{ }\mu\text{m}$ in NorESM2). Sublimating cloud ice and snow mass is calculated by the default MG2 scheme. Moreover, since Deshmukh et al. (2022) parameterization is developed based on the observation of dendritic particles, we only allow for sublimation break-up to activate between -10°C and -20°C , where such ice habits are more likely to occur in reality (Bailey and Hallet 2009). All new fragments are added to the cloud-ice category. Sublimation break-up of graupel, which is expected to occur at all temperatures (Deshmukh et al. 2022), is not accounted in the model, since graupel is not treated in MG2.

Finally, while PIP and SIP are significant ice-crystal sources, aggregation is a critical sink that can substantially decrease the cloud-ice number, while its parameterization is also a source of uncertainty in atmospheric models (Karrer et al. 2021). MG2 scheme accounts for aggregation through cloud ice-snow and snow-snow collisions. The accretion of cloud ice by snow follows the “continuous collection” approach as described in Rutledge and Hobbs (1983), while snow-snow aggregation follows Passarelli (1978). Aggregation efficiency (E_{ii}) between ice particles is considered the product of collision efficiency and sticking efficiency, with the latter depending on collisional kinetic energy and size (Phillips et al. 2015). However, a very simplified approach for E_{ii} is usually found in climate models; in CAM6-Oslo this parameter is set constant and to 0.5 (while it was 0.1 in the previous model version).

c. Sensitivity simulations

In this study, we examine the sensitivity of Arctic clouds to three main processes that determine cloud ice number: PIP, SIP and ice aggregation. At this point, it is worth noting

that a bug has been recently identified in MG2 (Shaw et al. 2021), which limits ice formation in mixed-phase clouds. This is due to an upper limit (n_{imax}) imposed for the ICNCs, which is equal to the INP number. Neither heterogeneous freezing processes nor SIP contribute to this INP limit, preventing them from producing new ice crystals (Shaw et al. 2021). In all our simulations we remove this n_{imax} limit, allowing PIP and SIP to evolve prognostically in the stratocumulus clouds. Our investigations on PIP effects include the use of either the prognostic or the diagnostic treatment for the freezing processes (see section 2b). Simulations that employ the Hoose and Möhler (2012) parameterization include the abbreviation 'CNT' in their name, while the ones that are run with diagnostic descriptions (Meyers et al. 1992; Bigg 1953; Young et al. 1974) include the prefix 'MBY' (Table 1). The CNT simulation is also referred as 'control' simulation in the text, as this is the model set-up utilized in CMIP6.

Sensitivity to SIP descriptions is examined by (a) either accounting for the standard SIP treatment in CAM6-Oslo which includes only the HM process after cloud droplet - snow collisions or (b) activating all the additional mechanisms, described in section 2b, simultaneously. Moreover, the performance of SIP processes like BR, which are a function of collisional kinetic energy, can be sensitive to different implementation methods. In this study we examine the performance of bulk vs hybrid-bin descriptions of SIP. Our bulk implementations follow the methodology of Sotiropoulou et al. (2020; 2021a,b) and Georgakaki et al. (2022) for BR and DSH, respectively. In their studies, the characteristic diameters and number-weighted velocities for each hydrometeor are used as input parameters for Phillips et al. (2017a) and (2018) schemes, while the standard MG2 formulations for accretion/aggregation rates are used to estimate the collisions that lead to SIP.

However, the MG2 scheme does not account for the accretion of cloud ice on raindrops. To estimate the number and mass collision tendencies for these interactions, we further implement the formulation proposed by Reisner et al. (1998), which is also utilized in the Morrison et al. (2005) scheme. Furthermore, to account for underestimations in collisional kinetic energy when the terminal velocity of the two colliding particles is similar ($u_1 \approx u_2$), we adapt the corrections in the mass- or number-weighted difference in terminal velocity (Δu_{12}) proposed by Mizuno (1990) and Reisner et al. (1998) in the bulk SIP implementations. When snowflakes collide with each other, it is assumed that 0.1% of the colliding mass is transferred to the generated fragments (Phillips et al. 2017a). The same assumption is applied for mode 2 of the drop-shattering process, thus only 0.1% of the colliding mass is transferred to the tiny fragments (Phillips et al. 2018). A detailed description of the implementation method can be

found in Sotiropoulou et al. (2021a) and Georgakaki et al. (2022).

On the contrary, Zhao et al. (2021) used an emulated bin approach for these two mechanisms, that better accounts for the impact of the size spectra variability. In their framework, the collision rates are calculated for each bin as $E_c \delta N_1 \delta N_2 \pi (r_1 + r_2)^2 |u_1 - u_2|$, where E_c is the collision efficiency, and δN_1 and δN_2 are the number concentrations in the two bins with particle radii r_1 and r_2 , respectively. Similarly, to the bulk approach described above, the number of generated fragments per collision is estimated following Phillips et al. (2017, 2018). Each new fragment produced by these two processes is assumed to have a 10- μm size (Phillips et al. 2018). Sensitivity simulations that account for all SIP mechanisms include the abbreviation 'SIP' in their name (Table I), while if an emulated bin framework is used instead of a bulk description, this suffix is modified to 'SIPBN'. Note that the emulated bin framework is only tested for BR and DSH; the adapted bin diameter ranges follow Zhao et al. (2020), being from 0.1 to 6.5 mm for raindrops (24 bins) and 0.1 to 50 mm for snow and cloud ice particles (35 bins). Each bin diameter (D) is estimated following $D_{k+1} = C D_k$ with $C=1.2$, discretizing the raindrop and ice particle size range in 24 and 35 bins respectively. A bulk approach is used for HM and sublimation break-up in all simulations.

Previous applications of these parameterizations in Arctic conditions (Sotiropoulou et al. 2020; Zhao et al. 2022) has shown that BR is the dominant SIP mechanism. However, Sotiropoulou et al. (2021b) showed that the Phillips et al. (2017a) parameterization is largely sensitive to the sublimation factor (ψ) – a correction factor for ice enhancement due to sublimation included in the BR formulation (see Appendix A). This factor was induced to account for the fact that the field data (Vardiman, 1978) used to constrain the number of fragments generated by this the prescribed ψ in Phillips et al. (2017a) study is overestimated, leading to underestimation of the BR efficiency. For this reason we perform two more sensitivity simulations, with both prognostic and diagnostic PIP, with this factor removed from the BR formulation. These experiments include the suffix 'SIPBN ψ ' in their name, as they are combined with the more advanced emulated bin framework.

Finally, ice aggregation is another process that has a significant impact on ICNCs but is highly-tuned in climate models. Generally observations from mid-latitudes indicate the presence of two temperature zones that promote aggregation: one around -15°C (Barret et al. 2019) associated with enhanced dendritic growth that facilitates the interlocking of the ice crystal branches (Connolly et al. 2012), and a second one close to the melting layer (Lamb and Verlinde 2011), caused by the increased sticking efficiency of melting snowflakes. However,

an analysis of recent dual-wavelength radar observations of shallow clouds from Ny-Ålesund suggest that enhanced aggregation occurs between -10°C and -15°C (Chellini et al. 2022), while no evidence of this process is found at higher temperatures. To adjust the aggregation efficiency to these new findings we perform simulations with modified E_{ii} . While in the standard scheme the aggregation efficiency remains constant to 0.5 throughout the whole temperature range, in our sensitivity simulations with the suffix 'AGG' this high value is only sustained between -10°C and -15°C . At colder temperatures E_{ii} is set to 0.1, while at warmer temperatures aggregation is deactivated ($E_{ii}=0$). A description of all the performed sensitivity tests and the different combinations of PIP, SIP and aggregation treatments is given in Table 1.

TABLE 1: Description of the sensitivity simulations

	Primary Ice Production	Secondary Ice Production	Aggregation
CNT (CONTROL)	prognostic (CNT)	HM (cloud droplet-snow)	constant E_{ii}
MBY	diagnostic (Meyers et al., Bigg, Young)	HM (cloud droplet-snow)	constant E_{ii}
CNT_AGG	prognostic (CNT)	HM (cloud droplet-snow)	variable E_{ii}
MBY_AGG	diagnostic (Meyers et al., Bigg, Young)	HM (cloud droplet-snow)	variable E_{ii}
CNT_SIP	prognostic (CNT)	HM (cloud droplet/rain-snow), bulk BR, bulk DS, SUBBR	constant E_{ii}
MBY_SIP	diagnostic (Meyers et al., Bigg, Young)	HM (cloud droplet/rain-snow), bulk BR, bulk DS, SUBBR	constant E_{ii}
CNT_SIPBN	prognostic (CNT)	HM (cloud droplet/rain-snow), bin BR, bin DS, SUBBR	constant E_{ii}
MBY_SIPBN	diagnostic (Meyers et al., Bigg, Young)	HM (cloud droplet/rain-snow), bin BR, bin DS, SUBBR	constant E_{ii}
CNT_SIPBN ψ	prognostic (CNT)	HM (cloud droplet/rain-snow), bin BR ($\psi=1$), bin DS, SUBBR	constant E_{ii}
MBY_SIPBN ψ	diagnostic (Meyers et al., Bigg, Young)	HM (cloud droplet/rain-snow), bin BR ($\psi=1$), bin DS, SUBBR	constant E_{ii}
CNT_SIPBN_AGG	prognostic (CNT)	HM (cloud droplet/rain-snow), bin BR, bin DS, SUBBR	variable E_{ii}
MBY_SIPBN_AGG	diagnostic (Meyers et al., Bigg, Young)	HM (cloud droplet/rain-snow), bin BR, bin DS, SUBBR	variable E_{ii}
CNT_SIPBN ψ _AGG	prognostic (CNT)	HM (cloud droplet/rain-snow), bin BR ($\psi=1$), bin DS, SUBBR	variable E_{ii}

MBY_SIPBN ψ _AGG	diagnostic (Meyers et al., Bigg, Young)	HM (cloud droplet/rain-snow), bin BR ($\psi=1$), bin DS, SUBBR	variable E_{ii}
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3. Results

a. Ny-Ålesund site

1) Cloud properties

In this section we focus on the evaluation of the simulated cloud macrophysical properties against remote-sensing surface observations collected at Ny-Ålesund (see section 2a). An evaluation of the modeled thermodynamic conditions is presented in Figs. S1 and S2 in the Supporting Information. NorESM2 offers a realistic representation of the temperature (Fig. S1a,b) and IWV (Fig. S2a) fields. Somewhat colder conditions are often found in the model within the lowest first two kilometers of the atmosphere (Fig. S1c), but most of the instantaneous modeled values show a discrepancy lower than 5° from the observed (Fig. S1d). In Figure S2, while instantaneous modeled and observed IWV values occasionally show large discrepancies (Fig. S2b), the overall mean observed (modeled) IWV is 7.4 (8.6) kg m⁻² suggesting a reasonable agreement.

Instantaneous modeled ICNC and IWC values derived at 3-hour time resolution are used in Fig. 1, which presents the median estimates as a function of temperature. ICNCs are constructed from the in-cloud cloud ice number and the in-precipitation snow number, predicted by the model. Similarly, modeled IWC is constructed from the respective in-cloud cloud ice and in-precipitation snow mass mixing ratios. IWC retrievals are averaged over a ± 10 -minute window around the model output timesteps and within ± 20 meters around the model vertical levels, while ICNC measurements are not available at this site. Measurement uncertainty is also plotted in Fig. 1b.

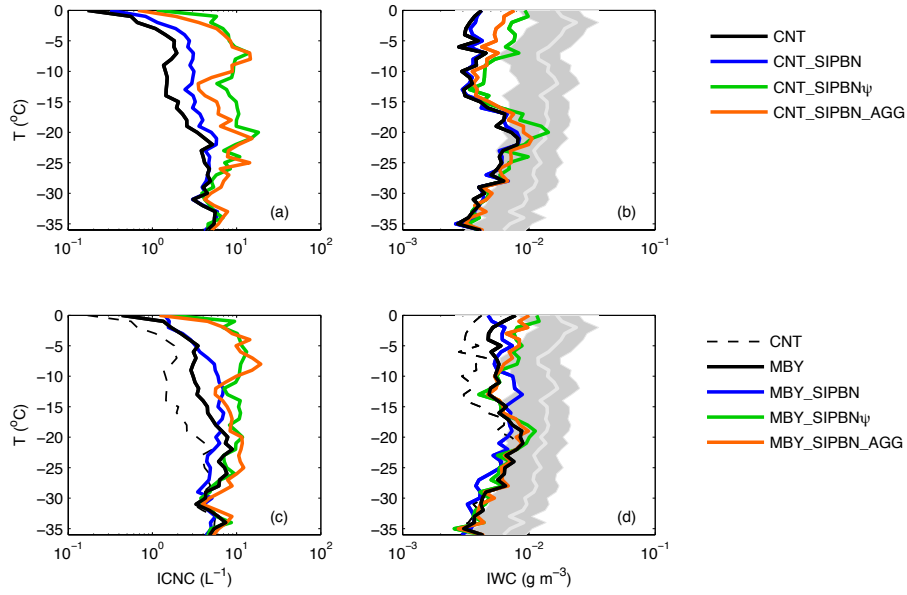


FIG 1. (a, c) Ice crystal number concentration (ICNC) and (b, d) ice water content (IWC) as a function of temperature. Grey shading (line) indicate the uncertainty range (50%) in the measured values. Results are derived from the Ny-Ålesund site (grid-point) for the period June 2016- May 2018. The observed IWC values are averaged over a ± 10 -minute window around the model output timesteps and within ± 20 meters round the model vertical levels.

The aerosol-aware CNT (control) simulation produces median ICNC concentrations round 1.5 L^{-1} within the -5°C to -15°C temperature range (Fig. 1a), which results in a median IWC that is on average five times lower than the observed (Fig. 1b). The IWC discrepancies between CNT and observations are reduced below -15°C : the median IWC is only two times lower than the observed median at these cold temperatures and lays very close to the uncertainty range. CNT_SIP does not result in any ice enhancement; for this reason it is shown in the Supplementary Information (Figure S3). CNT_SIPBN results in a weak ICNC enhancement within the temperature range that is favorable for SIP (Fig. 1a), compared to CNT, with hardly any impact on median IWC (Fig. 1b). CNT_AGG produces similar results to CNT_SIPBN (Fig. S3), thus activating SIP or decreasing ice aggregation has a similar effect on ICNCs. CNT_SIPBN ψ and CNT_SIPBN_AGG produce similar ICNC enhancements, resulting in 5-15 times larger median values (Fig. 1a) at the relatively high ($> -15^{\circ}\text{C}$) temperatures compared to CNT. Median ICNC values are close to 10 L^{-1} in these two simulations (Fig. 1a), which are in agreement with recent SIP observations from Arctic clouds at Ny-Alesund (Pasquier et al. 2022). Median IWC is 2-3 times larger in CNT_SIPBN ψ and CNT_SIPBN_AGG at temperatures above -15°C , compared to CNT, in closer agreement with observations (Fig. 1b). CNT_SIPBN ψ _AGG, which includes both the modified ψ factor and decreased aggregation results in reasonable agreement with the observed IWC (Fig. S3b),

however median ICNCs exceed 100 L^{-1} (Fig. S3a). Such median values are extreme and have not been observed in the Arctic. Since this set-up results in unrealistic microphysical properties, it is excluded from the rest of the analysis.

The MBY simulation (Fig. 1c) produces about 2-2.5 times higher median ICNCs than CNT at temperatures above -15°C , which increases median IWC values by 50-80% (Fig. 1d), in slightly better agreement with observations. No improvement in IWC is found at colder temperatures with the diagnostic PIP treatment. MBY_SIPBN results in negligible differentiations compared to the MBY simulation that do not affect the ice macrophysical state of the modeled clouds (Fig. 1d). The same applies for MBY_SIP and MBY_AGG, shown in the supplementary information (Fig. S3). Similarly to CNT_SIPBN ψ and CNT_SIPBN_AGG, MBY_SIPBN ψ and MBY_SIPBN_AGG produce median ICNCs close to 10 L^{-1} , which are realistic for Arctic SIP conditions observed at Ny-Alesund (Pasquier et al. 2022); these set-ups in somewhat improved IWC at temperatures above -15°C . Despite the improved median IWC in MBY_SIPBN ψ _AGG (Fig. S3d), this simulation produces unrealistically high median ICNCs ($> 100 \text{ L}^{-1}$), similar to CNT_SIPBN ψ _AGG (Fig. S3a,c). and thus this simulation is also excluded from the following analysis.

It is worth noting that CNT_SIPBN_AGG (MBY_SIPBN_AGG) is substantially more efficient in ICNC enhancement than CNT_SIPBN (MBY_SIPBN) and CNT_AGG (MBY_AGG), which are more similar (Fig. S3). This indicates an important interplay between SIP and decreased ice aggregation, when combined together. An overestimated aggregation rate can substantially limit ice multiplication, as the new fragments will rapidly aggregate and form precipitation-sized particles that will lead to IWC depletion through sedimentation (not shown). It is worth noting that the worst CNT_SIPBN_AGG and MBY_SIPBN_AGG performance is found at temperatures between -10°C and -15°C , where the default aggregation efficiency remains unaffected (see section 2c). This suggests that constraining ice aggregation is critical for the representation of Arctic cloud properties, particularly in conditions that favor SIP.

ICNC measurements were not conducted at Ny-Ålesund in 2016-2018, thus the ICNC profiles presented in Fig. 1 cannot be evaluated against observations. Only measured cloud particle concentrations over a limited size range ($<50 \mu\text{m}$) collected with the CVI and the FM-120 monitor are available (see section 2a). The FM-120 measurements are shown in Fig. 2 along with the modeled in-cloud droplet and cloud ice size spectra that include the sampled size range. Size spectra of larger particles, rain and snow are also shown in the same figure to

give a complete overview of the microphysical differences between the different simulations. The spectral differences between the standard NorESM2 set-up (CNT or MBY) and the sensitivity simulations are further highlighted in Fig. S4.

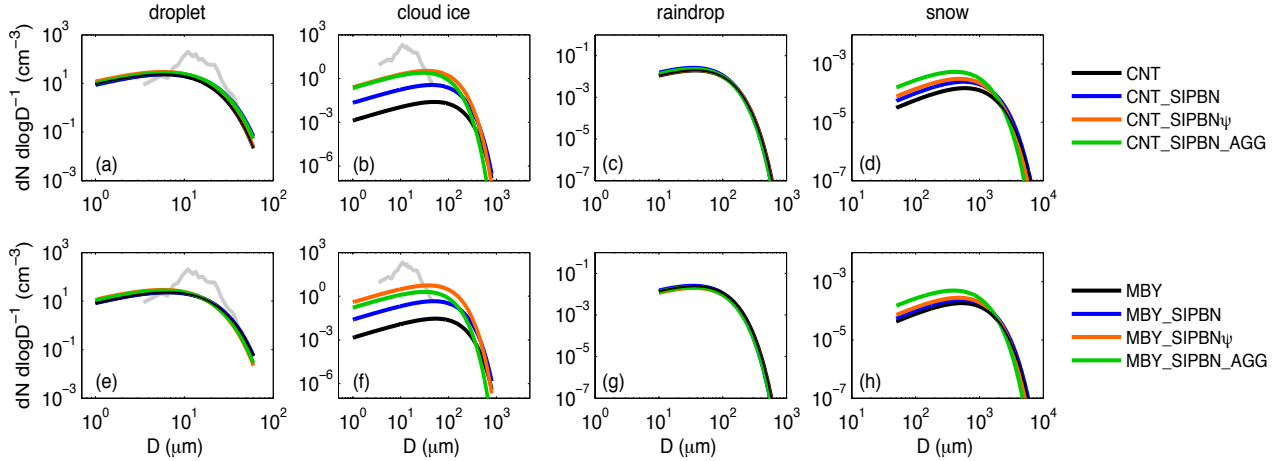


FIG 2. (a, e) droplet, (b, f) cloud ice, (c, g) raindrop and (d, h) snow size distributions for the different model sensitivity simulations. The first (second) row of panels presents simulations conducted with prognostic (diagnostic) PIP. Grey lines in panels (a, e) and (b, f) represent the observed spectrum derived from the fog monitor FM-120 for the size range 3.5-46 μm . All data span the period June 2015 - February 2018, as cloud particle measurements were not collected beyond this date.

All model simulations underestimate the measured hydrometeor concentrations at sizes between 10-30 μm . Differentiations in liquid hydrometeors among the simulations are generally small (Fig. 2a,c,e,g), while the set-ups that significantly enhance ICNCs in Fig. 1 produce more droplets within the 1-10 μm size range. More pronounced differences among the simulations are found in the cloud ice particle spectra: increasing ice production (Fig. 1) substantially enhances the smaller particles (Fig. 2b, f) in CNT_SIPBN ψ , CNT_SIPBN_AGG, MBY_SIPBN ψ and MBY_SIPBN_AGG simulations. This improves the agreement with observations particularly within the 10-30 μm size range, where CNT and MBY produce the largest cloud-ice underestimations. CNT_AGG (MBY_AGG) and CNT_SIP (MBY_AGG) are not included in Fig. 2, as they produce very similar spectra to CNT (MBY) and CNT_SIPBN (MBY_SIPBN), respectively. Distinct differences are also found in the snow size spectra (Fig. 2d, h), with the most pronounced shifts towards smaller snowflakes produced by simulations with reduced aggregation. This is however expected as aggregation directly augments the mass of snow particles either through self-collection or collection of cloud ice. Differences in the rain spectra are not pronounced (Fig. 2d, h): simulations with modified SIP or/and aggregation generally produce somewhat larger

raindrop concentrations compared to the default set-up (Fig. S4c, g), albeit these deviations are small in magnitude.

Overall, the large concentrations of the small particles measured at Zeppelin Station are not produced by any simulation. Atlas et al. (2021) have previously shown similar deviations between the default CAM6 and cloud particle measurements within the 5-30 μm size range, conducted with cloud probes, in high-latitude mixed-phase stratocumulus conditions (Southern Ocean clouds in their case). However, in our study the addition of missing SIP mechanisms did not eliminate this discrepancy. Another fact that may affect our model's performance is that it does not account for blowing snow, a mechanism that is commonly observed in mountainous regions and is known to provide the clouds with small ice particles raised from the surface during windy conditions (Gossart et al. 2017).

Apart from the cloud measurements collected on mount Zeppelin, insights into the particle sizes can be obtained from the radar-retrieved r_{ieff} . However, this dataset is associated with large uncertainties (see section 2a). The retrievals result in a median r_{ieff} of 44 μm for measurements collected above -20°C . This value is 79.8 μm for CNT and 79.3 μm for MBY and somewhat decreases in simulation with increased ice production. Among the simulations that utilize CNT PIP scheme, shown in Figs 1 and 2, CNT_SIPBN_AGG produces the median r_{ieff} closest to the observed (67.8 μm), while among the simulations with the diagnostic PIP, MBY_SIPBN ψ produces the smaller radii (64.5 μm). However, the differences in the modeled r_{ieff} do not exceed ~ 15 μm between the different model set-ups, which is substantially smaller than the uncertainty in the r_{ieff} retrieval, indicating these measurements cannot be used for a robust microphysical evaluation.

LWP measurements exhibit considerable variability throughout the year; for this seasonal LWP statistics are presented in Table 2. Moreover, as LWP distribution appears highly skewed, especially during winter and spring, both mean and median values are included in the Table. Observational statistics are also included in Table 2, derived from LWP measurements interpolated at the model timesteps. Modeled in-cloud LWP is constructed from the cloud droplet mixing ratios. Outliers with in-cloud LWP values > 700 g m^{-2} are excluded from the analysis. Moreover, for a consistent comparison with the processed observations, modeled cases with liquid surface precipitation > 0.05 mm day^{-1} (Kiszler et al. 2023) are also discarded from the LWP statistics.

Simulations with CNT produce generally more LWP than those that utilize the diagnostic PIP scheme. All simulations substantially overestimate LWP in summer, while

activation has minor impact on the mean/median values. In contrast, activation of SIP in simulations with diagnostic PIP has more pronounced effects: MBY_SIPBN ψ and MBY_SIPBN_AGG produce the lowest LWP values in summer, in better agreement with observations, although still overestimated. MBY simulation produces a somewhat better LWP in autumn compared to CNT, as it only deviates only $\sim 10 \text{ g m}^{-2}$ ($\sim 22 \text{ g m}^{-2}$) from the mean (median) observed value. Activation of SIP in the simulations with prognostic PIP has weak impact on LWP statistics, as the differences in mean values are smaller than 5 g m^{-2} compared to CNT. On contrary, SIP impacts are more pronounced in the simulations with diagnostic PIP, as increasing ice production results in decreasing mean/median LWP in autumn. MBY overestimates the observed mean/median LWP by $\sim 11/22 \text{ g m}^{-2}$, while MBY_SIPBN and MBY_SIPBN ψ exhibit a very good agreement with observations.

CNT overestimates mean LWP in winter by $\sim 20 \text{ g m}^{-2}$, while MBY produces in-cloud values very similar to the observed. Activation of SIP in simulations that treat PIP through CNT results in decreased LWP and improves agreement with observational statistics. In particular, CNT_SIPBN ψ (CNT_SIPBN_AGG) gives the best representation of the mean (median) winter LWP. In contrast, enhanced ice production through SIP in MBY_SIPBN ψ produces underestimated LWP compared to MBY, leading to larger deviations from the observations. MBY_SIPBN and MBY_SIP_AGG only slightly differentiate from MBY. Finally, model performance in spring for LWP is similar to winter: CNT overestimates LWP (albeit the deviations are less pronounced than for summer and autumn seasons), while increasing ice production in CNT_SIPBN and CNT_SIPBN_AGG improves agreement with measurements, with CNT_SIPBN being more realistic in this season. On contrary, mean and median LWP in MBY and MBY_SIP set-ups is very close to the observed spring values, while increasing ice production in MBY_SIPBN and MBY_SIPBN_AGG results in underestimated cloud liquid

TABLE 2: in-cloud Liquid Water Path (LWP, g m^{-2}) for observations and sensitivity simulations, segregated into mean/median seasonal values.

	summer	autumn	winter	spring
Observations	83.6/ 39.8	81.1/ 23.6	34.7/ 2.7	38.3/ 2.9
CNT (CONTROL)	135.2/ 116.4	99.1/ 57.3	54.7/ 5.9	58.1/ 10.6
CNT_SIPBN	141.6/ 120.6	101.4/ 61.9	52.8/ 4.2	58.6/ 7.9

CNT_SIPBN ψ	134.3/ 115.0	94.3/ 54.6	26.1/ 5×10^{-5}	45.2/ 5.9
CNT_SIPBN_AGG	136.6/ 116.0	103.1/ 63.5	46.7/ 3.3	53.7/ 7.4
MBY	126.4/ 108.5	91.6/ 55.5	33.6/ 3.6	37.3/ 5.5
MBY_SIPBN	129.8/ 107.2	81.8/ 36.8	32.9/ 3.3	39.4/ 4.7
MBY_SIPBN ψ	119.1/ 97.4	78.7/ 32.9	17.5/ 4×10^{-5}	27.2/ 2.0
MBY_SIPBN_AGG	118.9/ 95.5	62.5/ 12.5	30.9/ 1.15	29.9/ 4.1

2) Microphysical processes

To better understand the interactions between the underlying microphysical processes that drive the macrophysical differences between the different sensitivity simulations, vertical profiles of mean PIP, SIP, WBF and riming tendencies are plotted in Fig. 3. The ice multiplication tendencies of the individual SIP mechanisms are shown in Fig. 4. Interestingly, when a diagnostic PIP treatment is applied (Fig. 3e), PIP rates generally decrease with increasing ice production through modifications in SIP or aggregation, a behavior that is not found in simulations with CNT (Fig. 3a). An analysis of the changes in thermodynamic profiles between the simulations (Fig. S5a, c) indicate warmer temperatures with increasing ice production, especially at heights above 1 km, while the specific humidity response is more variable (Fig. S5b, d); since the diagnostic PIP parameterizations are solely dependent on the thermodynamic conditions, these temperature variations can explain to a large extent the variable PIP rates in Fig. 3e. In Fig. 3a substantial differences in PIP are mainly found in CNT_SIPBN ψ ; these differences seem to follow changes in specific humidity profiles (Fig. S5b, d) suggesting that the prognostic PIP treatment is mostly affected by variations in supersaturation.

SIP rates in CNT_SIP and MBY_SIP are very similar to CNT and MBY (Fig. 4b, f). This is in agreement with the findings of Fig. S1, which reveal that the bulk implementations of BR and DSH hardly result in any ice multiplication. This result is further confirmed by Fig. 4 which shows that BR and DSH tendencies are orders of magnitude smaller than those of HM. Another interesting finding is that including rain-snow collisions in the HM description in the CNT_SIP and MBY_SIP simulations does not enhance the efficiency of this process compared to CNT and MBY that account only for cloud drop-snow collisions (Fig. 4a, e), as the precipitation particle concentrations are generally limited (Fig. 2c,d,g,h). Furthermore, sublimation breakup activates in the lowest five atmospheric kilometers but remains extremely weak through the whole layer (Fig. 4d, h).

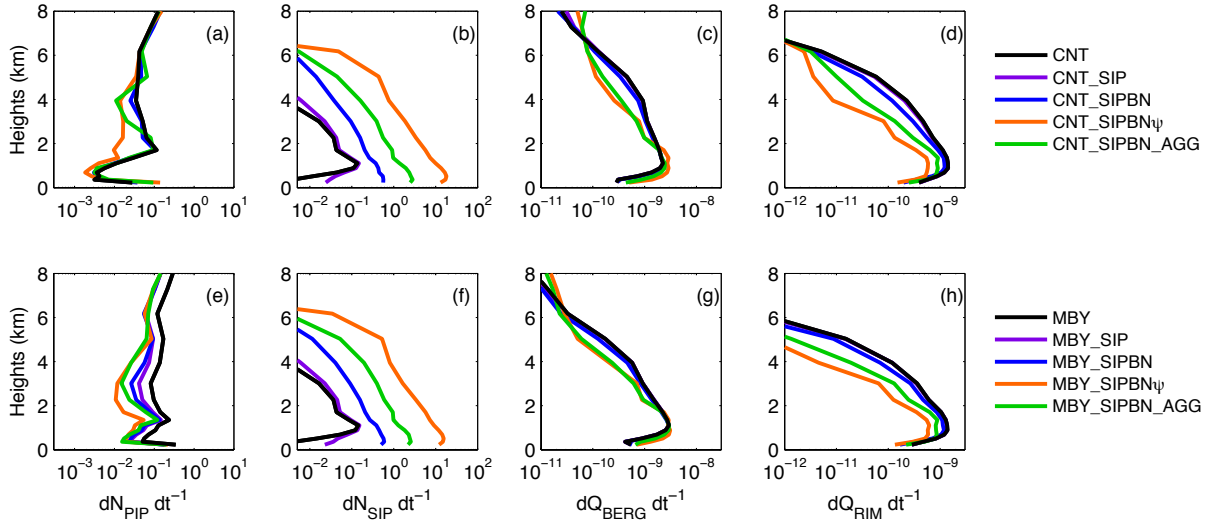


FIG 3. Mean vertical profiles of number concentration tendencies ($\text{kg}^{-1} \text{s}^{-1}$) due to (a, e) PIP ($dN_{\text{PIP}} dt^{-1}$) and (b, f) SIP ($dN_{\text{SIP}} dt^{-1}$), (c, g), and mass concentration tendencies ($\text{kg kg}^{-1} \text{s}^{-1}$) due to WBF ($dQ_{\text{BERG}} dt^{-1}$) and (d, h) riming ($dQ_{\text{RIM}} dt^{-1}$) for the different model sensitivity simulations. The WBF rate is the sum of the individual rates for cloud ice and snow particles, while riming is the sum of cloud droplet and rain accretion on snow. The first (second) row of panels presents simulations conducted with prognostic (diagnostic) PIP.

Utilizing an emulated bin framework for BR and DSH enhances SIP rates by on average a factor of ~ 5 in the lowest 4 atmospheric kilometers, compared to the simulations that adapt bulk frameworks (Fig. 3b, f). SIP also becomes prominent at higher altitudes (> 4 km), where bulk parameterizations do not produce any ice multiplication. Figure 4 indicates that the SIP is mainly due to the BR process. Although the emulated bin framework enhances DSH efficiency, the DSH rates remain substantially lower than those that correspond to the BR mechanism. Decreasing aggregation in CNT_SIPBN_AGG and MBY_SIPBN_AGG increases SIP efficiency by on average a factor of 5 (Fig. 3b, f), compared to CNT_SIPBN and MBY_SIPBN simulations, mainly through the enhancement of the BR process (Fig. 4b, f). Interestingly, the largest sensitivity of SIP is found in the treatment of the sublimation correction factor ψ in BR description. The simulation with $\psi=1$ (Table 1), that does not account for this correction results in BR rates enhanced by 1-1.5 orders of magnitude (Fig. 4b,f), which highlights the importance of constraining this parameter for an accurate BR representation. It is worth noting that increasing BR efficiency is associated with decreasing HM rates (Fig. 4). This is due to the fact that increasing SIP results in smaller ice particle sizes that are less likely to rime and initiate HM. The impact of SIP on riming and the WBF

efficiency will be discussed below.

The simulations with a modified ψ factor or aggregation efficiency are characterized by an enhanced (reduced) WBF efficiency in the low-level (mid-level) clouds (Fig. 3c, g) compared to the rest of the simulations that produce significantly less ice content (Fig. 1). These simulations are also characterized by decreased riming efficiency throughout the whole troposphere (Fig. 3d, h). This is likely due to the shift of the frozen hydrometeor spectra to smaller particle sizes (Fig. 2) that are less efficient in depositional growth and liquid accretion. The more active WBF mechanism in the low-level clouds is likely responsible for the reduced in-cloud LWPs (Table 2).

Our findings indicate that the inclusion of missing SIP mechanisms in NorESM2 can improve the macrophysical representation of Arctic mixed-phase clouds, but this requires the use of an emulated bin framework for BR and DSH, which is computationally about two times more demanding than the bulk descriptions of SIP. Modifications in the HM description, with the inclusion of rain-snow interactions, did not enhance the efficiency of this process in the examined conditions, suggesting that these modifications are redundant. BR appears to be the dominant SIP mechanism, however its efficiency is very sensitive to the treatment of the poorly constrained parameter ψ . DSH and SUBR processes are substantially weaker in the examined conditions. DSH is likely not favored due to lack of relatively large drops to initiate the process (Fig. 2c, g), while SUBBR is likely limited by the high relative humidity conditions that generally dominate in the Arctic.

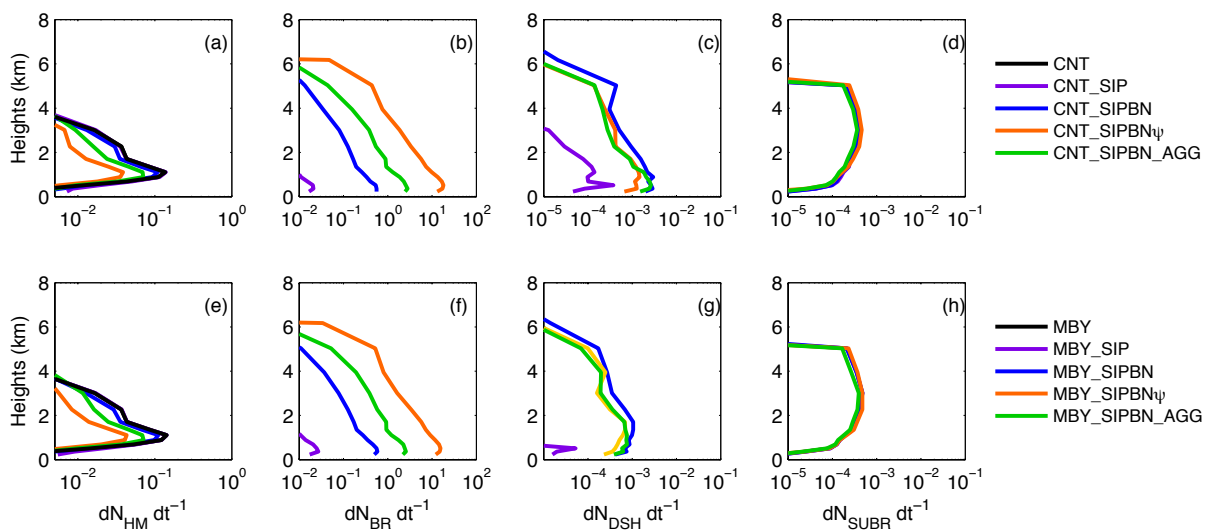


FIG 4. Mean vertical profiles of number concentration tendencies ($\text{kg}^{-1} \text{s}^{-1}$) due to SIP from the (a, d) HM, (b, f) BR and (c, f) DSH and (d, h) SUBBR for the different model sensitivity simulations. The

first (second) row of panels presents simulations conducted with prognostic (diagnostic) PIP.

b. Arctic region

In this section, the performed simulations are evaluated against satellite observations averaged over the whole Arctic region ($>66^{\circ}\text{N}$). In Table 3 the simulated net total cloud radiative effects (CRE) at the Top Of the Atmosphere (TOA) are compared to EBAF v4.1 products; differences in the net surface cloud radiative effect are found less significant and thus are not shown. Furthermore, in Table 4 the modeled and observed total cloud cover is presented; the latter is represented by the GOCCP product. Two values are shown for the different simulations: (a) the COSP (CFMIP Observation Simulator Package) output which is suitable for comparison with the satellite observations (Bodas-Salcedo et al. 2011) and (b) the direct model outputs, which control radiation (Table 4).

Net CRE at TOA is negative in summer and spring, as shortwave effects dominate, while during autumn-winter, when incoming solar radiation is weaker, the dominance of the longwave components result in positive values. The simulations that utilize the CNT PIP scheme produce enhanced warming (cooling) at TOA autumn-spring (summer) than the simulations with diagnostic PIP parameterizations, resulting in slightly better (worse) agreement with EBAF observations. CNT overestimates cloud radiative cooling at TOA in summer by 5.3 W m^{-2} and overestimates cloud induced warming during the rest of the seasons, with the largest deviations from EBAF observations found in winter (4.6 W m^{-2}). CNT_SIPBN produces very similar results to CNT, while the two simulations with the enhanced ice production produce larger net longwave effects (Table S1), shifting the net CRE towards warmer values. This improves the representation of the net cloud radiative effect during most of the year, with the largest improvements found in winter for CNT_SIPBN_AGG ($\sim 3 \text{ W m}^{-2}$). Differences in shortwave CRE at TOA among the simulations are generally smaller, never exceeding 1.5 W m^{-2} (Table S2). The response of the simulations with diagnostic PIP to increasing ice production is similar to those that employ CNT, but weaker in magnitude. As a result, the differences between MBY and MBY_SIPBN_AGG or MBY_SIPBN ψ are generally small ($<1.5 \text{ W m}^{-2}$). The most pronounced improvement in net CRE is found in simulation MBY_SIPBN ψ for the summer season, however this is due to compensating errors between the shortwave and longwave components (Tables S1, S2).

TABLE 3: Net Total Cloud Radiative Forcing at TOA

	summer	autumn	winter	spring
EBAF observations	-44.6	6.6	12.4	-7.1
CNT (CONTROL)	-49.9	5.3	7.8	-1.9
CNT_SIPBN	-49.1	5.3	7.9	-1.8
CNT_SIPBN_AGG	-48.5	7.5	10.8	-1.6
CNT_SIPBN ψ	-47.6	6.2	8.6	-1.2
MBY	-46.7	4.6	7.7	-0.8
MBY_SIPBN	-46.1	4.7	7.8	-0.7
MBY_SIPBN_AGG	-46.5	4.8	8.4	-0.8
MBY_SIPBN ψ	-45.3	4.8	8.4	-0.5

621
622 COSP total cloud cover for CNT and MBY simulations is in good agreement with EBAF
623 observations in summer (Table 4), but underestimates cloud cover during the rest of the
624 seasons, especially in winter and spring. Increasing ice production result in somewhat
625 increased total cloud cover: the difference between CNT (MBY) and the simulation that
626 produces the largest ice content, CNT_SIPBN ψ (MBY_SIPBN ψ), is about 1-1.5% (1-4%).
627 Increasing COSP cloud cover is mainly caused by increased high cloud cover (Tables S4);
628 COSP mid-level cloud cover exhibits little sensitivity to variations in ice treatment (not
629 shown), while COSP low-level cloud cover decreases with increasing ice production (Table
630 S3). However, this behaviour is not found in the direct model output, in which both total and
631 low-level cloud cover increase in the simulations with enhanced ice content (Table S3). A
632 possible explanation for this discrepancy is that as the enhanced ice production results in
633 optically-thinner layers, thus the fraction of the very thin clouds that do not pass the detection
634 thresholds applied in the COSP simulator increases (e.g. only subcolumns generated by COSP
635 per model grid with an optical depth > 0.3 are included in the calculations). The direct model
636 outputs however are generally compatible with changes in CRE_{LW} at TOA (Table S1), as
637 increasing cloud cover reduces outgoing thermal radiation, resulting in a warming effect.

638
639 **TABLE 4:** COSP /model total cloud cover. First value is derived from COSP simulator,
640 while the second one corresponds to the direct model output.

	summer	autumn	winter	spring
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EBAF observations	80.7	79.6	63.3	70.3
CNT (CONTROL)	81.5/ 85.0	75.6/ 85.6	57.8/ 78.0	55.4/ 74.7
CNT_SIPBN	81.5/ 85.0	75.2/ 85.5	57.6/ 78.0	54.8/ 74.6
CNT_SIPBN_AGG	82.3/ 85.7	75.7/ 86.3	57.6/ 79.0	54.7/ 75.5
CNT_SIPBN ψ	82.6/ 86.1	75.7/ 86.8	57.0/ 79.2	53.9/ 75.5
MBY	79.7/ 83.8	69.8/ 83.7	50.7/ 76.7	46.4/ 71.5
MBY_SIPBN	79.7/ 83.8	69.7/ 83.9	60.0/ 76.9	46.2/ 71.5
MBY_SIP_AGG	80.4/ 84.5	70.3/ 84.6	52.2/ 78.1	47.0/ 72.4

4. Discussion

In this study, we examine the sensitivity of Arctic cloud properties to the representation of ice microphysical processes in NorESM2. The primary target is to quantify the impact of PIP and SIP parameterizations on the cloud macrophysical structure and radiative effects. Sensitivity simulations with PIP are performed with two different primary ice treatments: (a) a prognostic CNT scheme that explicitly predicts ice formation from cloud-aerosol interactions and (b) diagnostic temperature-dependent parameterizations for all the heterogeneous freezing processes. The standard version of NorESM2 accounts only for the HM process through droplet-snow collisions. The sensitivity to SIP is examined by implementing additional SIP mechanisms, namely the BR, DSH and SUBBR mechanisms. Furthermore, the HM description is modified to account for rain-snow collisions, following Morrison et al. (2005).

The interactions of PIP and SIP with ice aggregation are also a subject of the present study. The standard parameterization of this process in NorESM2 includes a constant aggregation efficiency (E_{ii}) set to 0.5. To investigate the sensitivity of our results to this parameter, we adapt a variable E_{ii} which is qualitatively constrained by recent dual-wavelength radar measurements of shallow Arctic clouds (Chellini et al. 2022): E_{ii} is set to 0.5 at temperatures between -10°C and -15°C and to 0 (0.1) at temperatures below (above) this range. The model results are evaluated against surface observations from Ny-Ålesund and satellite retrievals over the whole Arctic.

Using CNT instead of diagnostic PIP descriptions results in a worse agreement with IWC observations from Ny-Ålesund at temperatures between -5°C and -15°C , when no other modification in SIP or aggregation is implemented. We speculate that the reason for this behavior is that the NorESM2 CNT parameterization does not account for aerosol types that

are efficient INPs at relatively warm temperatures (e.g. biological aerosols). This larger underestimation in ice content is accompanied by substantially overestimated LWP, compared to the observed.

Activating the missing SIP mechanisms enhances ice content, mainly through the BR process. BR efficiency however highly depends on the treatment of the correction factor ψ , which is included in the Phillips et al. (2017a) parameterization to account for the ice enhancement due to sublimation. This is a poorly constrained parameter, while the value assigned by Phillips et al. (2017a) likely results in underestimations of the BR effect. DSH and SUBBR are the two mechanisms with the weakest efficiency in the examined conditions. Moreover, modifications in the HM description to account for rain-snow collisions do not enhance the efficiency of the process. HM and DSH are likely limited by the fact that relatively large raindrops are generally few in the examined conditions. SUBBR is likely not favored due to the high relative humidity conditions (Wyszyński and Przybylak 2014; Tjernström et al. 2012) that often persist in polar environments.

It is worth noting that the current BR and SUBBR implementations can be affected by the number of frozen hydrometeors that are treated in the cloud scheme and MG2 does not account for graupel particles. While Gettelman et al. (2019) showed that the global climate impact of rimed ice in stratiform clouds is negligible in 100-km scale simulations, their study concerns the standard MG2 scheme that does not account for additional SIP mechanisms. Zhao et al. (2020, 2021) on the other hand showed that including graupel can enhance the efficiency of the BR process in Arctic clouds. Similarly, the SUBBR implementation concerns only the snow particles in our model, which can undergo sublimation break-up only within a limited temperature range (see Section 2c). In contrast, sublimation break-up of graupel can occur at any temperature (Deshmukh et al. 2022). In summary, the fact that graupel category is not treated in NorESM2 suggests that the overall efficiency of both BR and SUBBR mechanisms might be underestimated in our simulations.

Interestingly, SIP efficiency increases substantially with decreasing ice aggregation in our simulations. This is because enhanced SIP results in enhanced ice aggregation when a constant aggregation efficiency is assumed. However, in reality, this might not be necessarily true as enhanced SIP may lead to the prevalence of small ice particles that are not efficient in aggregation or to the reduction of dendritic ice crystal concentrations through break-up; dendrites are the ice habits that are known to be most favorable for aggregation (Karrer et al., 2021; Chellini et al., 2022).

Increasing ice production through changes in SIP and /or aggregation decreases has a direct impact on other microphysical processes, such as riming and WBF efficiency. Specifically simulations with higher ice number are characterized by decreased riming throughout the whole vertical profile. In contrast, WBF exhibits a more variable behaviour: it is less efficient in mid-level clouds, while in low-level clouds below 1-km WBF can become more effective in these simulations. The net effect of all these microphysical processes on the macrophysical structure of the Arctic clouds at Ny-Alesund site is an enhancement in IWC and reduction in cloud liquid, albeit the latter is more pronounced in simulations with diagnostic PIP. This improves the agreement of the simulations that utilize the CNT PIP scheme with the field observations especially in winter/spring, as CNT is generally characterized by substantially overestimated LWP. In contrast, SIP enhancement or decreased aggregation often results in degraded cloud liquid representation in the simulations with the diagnostic PIP scheme.

Finally, as far as SIP/aggregation impacts on cloudiness over the whole Arctic region are concerned, increasing ice production is found to lead to increased total cloud cover. This is mainly due to the fact that these ice microphysical processes shift the overall cloud ice particle spectra towards smaller sizes, extending the cloud particle lifetime in the atmosphere. The largest increases are observed in the modelled low-level cloud cover; weaker increases are found in the high-cloud cover, while mid-level clouds are hardly impacted. The increased cloudiness, results in improved CRE predictions at TOA especially during the cold months, through improvements mainly in the longwave component. The latter is due to enhanced downward longwave emission, which decreases the negative CRE bias that is produced by the standard NorESM2 model in winter.

5. Conclusions

The main objective of this study is to quantify the sensitivity of the Arctic cloud microphysical and radiative properties to the description of three ice microphysical processes (PIP, SIP and ice aggregation) and infer the process hierarchy based on their importance. Changes in PIP treatment have a very pronounced impact on cloud characteristics and radiative impacts, however without the inclusion of missing SIP mechanisms, no PIP scheme alone can reproduce a realistic microphysical structure. The fact that the most realistic simulations include modifications in SIP highlights the importance of this process for modeling Arctic clouds. Yet, SIP representation remains highly sensitive to uncertainties in

the description of the underlying mechanisms and particularly collisional break-up. Finally changes in ice aggregation efficiency have very pronounced impacts in our simulations, however this is mainly due to the impact of this process on collisional break-up efficiency, rather than due to its direct impact on the ice/snow budget. Our results suggest that improving SIP representation in climate models can have pronounced impacts on the Arctic cloud representation and cloud radiative effect, especially during winter, however uncertain parameters (such as the correction factor ψ or E_{ii} that affect collisional break-up efficiency) should be constrained or tuned carefully.

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Data availability statement:

Both surface-based and satellite observations are available online. LWP datasets from Ny-Ålesund for the years 2016, 2017 and 2018 can be found at <https://doi.org/10.1594/PANGAEA.902096> (Nomokonova et al. 2019a), <https://doi.org/10.1594/PANGAEA.902098> (Nomokonova et al. 2019b) and <https://doi.org/10.1594/PANGAEA.902099> (Nomokonova et al. 2019c). IWC and $R_{i_{eff}}$ data can be found at <https://doi.pangaea.de/10.1594/PANGAEA.898556> (Nomokonova et al. 2019d). HATPRO temperature profiles can be downloaded from <https://doi.org/10.1594/PANGAEA.902145> (Nomokova et al. 2019e), <https://doi.org/10.1594/PANGAEA.902146> (Nomokova et al. 2019f) and

<https://doi.org/10.1594/PANGAEA.902147> (Nomokova et al. 2019g). Ny-Ålesund IWV measurements for the same years are available at <https://doi.org/10.1594/PANGAEA.902140> (Nomokonova et al. 2019h), <https://doi.org/10.1594/PANGAEA.902142> (Nomokova et al. 2019i) and <https://doi.org/10.1594/PANGAEA.902143> (Nomokova et al. 2019j). Cloud measurements collected on mount Zeppelin are available at <https://doi.org/10.17043/zeppelin-cloud-aerosol-1> (Karlsson et al. 2021b). The CERES-EBAF data are retrieved from <https://ceres.larc.nasa.gov/data/>, while GOCCP dataset can be downloaded from <https://climserv.ipsl.polytechnique.fr/cfmip-obs/>. ERA-Interim reanalysis products can be accessed through <https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era-interim>. Model datasets will be deposited to zenodo.org upon acceptance of the manuscript.

Appendix A: Sublimation corrector factor in BR formulation

The Phillips et al. (2017a) parameterization predicts the number of fragments (F_{BR}) generated from mechanical break-up upon collisions of two ice particles using the equation:

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

where K_o is the collisional kinetic energy, α is the surface area of the smaller ice particle that undergoes fracturing, A represents the number density of the breakable asperities in the region of contact, γ is a function of the particle's rimed fraction and C is the asperity-fragility coefficient, which is a function of a correction term (ψ) for the effects of sublimation based on the field observations by Vardiman (1978). Specifically, for planar ice the assigned values are: $C = 7.08 \times 10^6 \psi$ and $\psi = 3.5 \times 10^{-3}$. Thus, a ψ value smaller than unity has a decreasing impact on F_{BR} estimation. Setting $\psi=1$ in the sensitivity simulations with ' ψ ' suffix assumes no impact of sublimation break-up on the Vardiman (1978) data used to constrain the above formulation.

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