Global Importance of Secondary Ice Production

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Key Points:

- Secondary ice production can enhance ice number concentrations by three orders of magnitude in moderately cold clouds
- Secondary ice production decreases the global annual average liquid water path by 22% and increases ice water path by 23% in a global climate model
- Secondary ice production changes the global annual average shortwave, longwave, and net cloud forcing by 2.1, –1.0, and 1.1 W m⁻², respectively
Abstract

Measured ice crystal number concentrations are often orders of magnitude higher than the number concentrations of ice nucleating particles, indicating the existence of secondary ice production (SIP) in clouds. Here, we present the first study to examine the global importance of SIP through the droplet shattering during freezing of rain, ice-ice collision fragmentation, and rime splintering, using a global climate model. Our results show that SIP happens quite uniformly in the two hemispheres and dominates the ice formation in the moderately cold clouds with temperatures warmer than -15°C. SIP decreases the global annual average liquid water path by −14.6 g m⁻² (−22%), increases the ice water path by 8.7 g m⁻² (23%), improving the model agreement with observations. SIP changes the global annual average shortwave, longwave, and net cloud forcing by 2.1, −1.0, and 1.1 W m⁻², respectively, highlighting the importance of SIP on cloud properties on the global scale.

Plain Language Summary

Global climate models (GCMs) show large cloud biases in terms of phase partitioning due to their poor representations of ice microphysical processes. Ice formation in mixed-phase clouds can remarkably change the cloud albedo, lifetime, and precipitation efficiency. Despite strong evidence of SIP in moderately cold clouds, it is still poorly

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represented in GCMs and its impacts are not quantified. Here, we present the first study to examine the global importance of SIP using a GCM. Our study shows that SIP decreases the global annual average liquid water path by \(-14.6 \text{ g m}^{-2} (-22\%)\) and increases the ice water path by \(8.7 \text{ g m}^{-2} (23\%)\), improving the model agreement with observations. SIP changes the global annual average net cloud radiative forcing by \(1.1 \text{ W m}^{-2}\), highlighting the importance of SIP on cloud properties on the global scale.
1. Introduction

It has been frequently reported that measured ice crystal number concentrations (Ni) are often orders of magnitude higher than number concentrations of ice nucleating particles (INPs). Previous studies interpreted this observed discrepancy as evidence of secondary ice production (SIP) in clouds (Mossop, 1985; Field et al., 2016; Korolev et al., 2020). SIP is referred to as the ice production processes from preexisting ice crystals. Ice nucleation (or primary ice production, hereafter referred to as PIP), is the ice production processes involving aerosol particles via droplet freezing or deposition nucleation (Vali et al., 2015). Currently, there are several proposed SIP mechanisms: rime splintering (also known as the Hallett and Mossop (HM) process), ice-ice collision fragmentation (IIC), droplet shattering during freezing (FR), fragmentation during the sublimation of ice bridge, ice fragmentation during thermal shock, and activation of INPs in a transient supersaturation (Field et al., 2016; Korolev and Leisner, 2020).

Observational studies have provided strong evidence of SIP in different types of clouds and in different geographic regions. By analyzing the aircraft data, Hobbs and Rangno (1985,1990,1998) attributed the occurrence of high Ni in maritime cumulus clouds off the Washington coast to SIP, and suggested the importance of other SIP mechanisms in addition to the HM. Based on in-situ measurements of the Arctic stratus clouds, Rangno and Hobbs (2001) revealed that IIC and FR are the two important SIP mechanisms besides the HM. Based on in-situ measurements by particle imaging probes,
Lawson et al. (2015) and Heymsfield and Willis (2014) found that FR contributed significantly to the high Ni in cumulus clouds in the Caribbean and Africa. Similar results were reported by Taylor et al. (2016) in maritime cumulus over the southwest peninsula of the United Kingdom. Using radar and aircraft measurements of convection in a warm-frontal mixed-phase cloud, Hogan et al. (2002) attributed the formation of column ice crystals in the convection to the SIP. Evidence of SIP was also reported by Crosier et al. (2014) in their study of convection in a strong cold front.

Despite its potential importance, representations of SIP in global climate models (GCMs) are still very crude. Most of the current GCMs only consider the HM. However, this process operates at a relatively narrow temperature range of –3 to –8°C, and is not efficient to reproduce observed Ni (Sullivan et al., 2018; Sotiropoulou et al., 2020). Recent studies indicated that GCMs such as the DOE’s Energy Exascale Earth System Model version 1 (E3SMv1) and the NCAR’s Community Earth System Model version 2 (CESM2) significantly underestimate observed ice water path (IWP) while overestimate observed liquid water path (LWP) in mid- and high-latitude mixed-phase clouds (D’Alessandro et al., 2019; Zhang et al., 2019; Zhang et al., 2020). The cloud phase regulates the cloud radiative forcing (Shupe and Intrieri, 2004; Lawson and Gettelman, 2014; Korolev et al., 2017), and plays an important role in determining the mixed-phase cloud feedbacks (Stephens, 2005; Gettelman and Sherwood, 2016; Ceppi et al., 2017; Tan et al., 2019) and climate sensitivity (Tan et al., 2016; Bodas-Salcedo et al., 2019;
Gettelman et al., 2019; Zelinka et al., 2020). Furthermore, cloud phase can also affect the Arctic amplification (Tan and Storelvmo, 2019).

To bridge the gap between measurements and GCM representations of SIP, we incorporated two new types of SIP, i.e., FR and IIC into CESM2, and improved the agreement of modeled clouds with observations in the Arctic (Zhao et al., 2020). The parameterization methods for the two new types of SIP were based on the latest theoretical developments of Phillips et al. (2017, 2018). In this study, we attempt to further quantify the global importance of SIP on cloud properties and cloud radiative forcing on the global scale.

2. Method

In this study, we use the Community Atmosphere Model version 6 (CAM6), the atmosphere component of CESM2 (Danabasoglu et al., 2020). The treatment of ice nucleation in cirrus clouds includes the homogeneous freezing of sulfate and heterogeneous immersion freezing on dust, and the competition between these two mechanisms at temperatures below −37℃ (Liu and Penner, 2005). In mixed-phase clouds with temperatures between −37℃ and 0℃, the classical nucleation theory (CNT) is used to describe the three mechanisms of heterogeneous ice nucleation, i.e., immersion freezing, contact freezing, and deposition freezing on mineral dust and black carbon (Hoose et al., 2010; Wang et al., 2014). The aerosol processes are represented by the 4- mode version of
the Modal Aerosol Module (Liu et al., 2016, Liu et al., 2012). The cloud microphysics in stratiform clouds is described by a two-moment bulk scheme, called the MG2 scheme (Gettelman et al., 2015). MG2 includes the prognostic precipitation and considers only one type of SIP, the HM. As mentioned before, this process only operates at the temperature range from –8 to –3°C.

We implemented two new types of SIP into CAM6, the fragmentation of freezing rain droplets (FR) and ice-ice collision breakup (IIC), following Phillips et al. (2017, 2018). The FR contains two modes (Phillips et al., 2017). The first mode describes the fragmentation of a freezing rain droplet triggered by contacting with an INP or a smaller ice crystal. In this process, the rain droplet is more massive. In the second mode, a rain droplet collides with a more massive ice particle so that the rain particle freezes and breaks up. The IIC includes snow-snow, snow-cloud ice, snow-graupel, cloud ice-graupel, and graupel-graupel collisions (Phillips et al., 2018). A approach based on Zhao et al. (2017) was applied to the MG2 scheme to diagnose the graupel mass and number mixing ratios for the IIC calculation. Since these processes happen at the particle scale, an emulated bin framework was developed for implementing the two new types of SIP in the MG2 cloud microphysics (Zhao et al., 2020).

We conducted four model experiments in this study, as listed in Table S1. The control (CTL) experiment uses the default CAM6 model in which only the HM is considered. The SIP_CNT experiment is the same as CTL but considers all the three
types of SIP, i.e., HM, FR, and IIC. To examine the impacts of primary ice nucleation on SIP, we conducted two sensitivity experiments: one with the DeMott et al. (2015) ice nucleation scheme to replace CNT in SIP_CNT, hereafter the experiment as SIP_D15, and one with the Niemand et al. (2012) ice nucleation scheme to replace CNT in SIP_CNT, hereafter the experiment as SIP_N12. All simulations were performed for 6 years with a horizontal resolution of 0.9° (latitudes) by 1.25° (longitude) and 32 vertical levels from the surface to 3 hPa. The model is initialized with prescribed greenhouse gas concentrations and the climatological sea ice and sea surface temperature. The present-day (year 2000) precursor gas and aerosol emissions are used to drive the model. The last five-year simulations are used in the analysis with the first-year simulation used as the model spin-up.

3. Results

3.1 Relative importance of SIP and PIP

Figure 1 shows the annual zonal mean ice production rates of SIP and PIP from the SIP_CNT experiment. The SIP rate includes contributions from the FR, IIC, and HM. The SIP production rate has a relatively homogeneous distribution pattern between the two hemispheres: the rate over the Southern Hemisphere (SH) is in the same magnitudes as that over the Northern hemisphere (NH) (Figure 1a). In contrast, the PIP production rate shows an obvious hemispheric asymmetry, with the rate over NH two orders of magnitude larger than that over SH (Figure 1b). PIP dominates the ice production over
the NH, which is mainly contributed from the immersion freezing on dust aerosol (Figure 1c and Figure S1). The PIP rate in the NH is around 10 to 50 kg$^{-1}$ s$^{-1}$ with the maximum rate at ~400 hPa over 45° N. As a comparison, the PIP rate in the SH is around 1 to 5 kg$^{-1}$ s$^{-1}$, and is much smaller than that in the NH where most of the dust INPs are originated from. On a global perspective, the SIP rate is 1 kg$^{-1}$ s$^{-1}$ with two peaks. The first peak locates at ~60° S over the Southern Ocean (SO), extended from surface to 700 hPa with the maximum value of 5 kg$^{-1}$ s$^{-1}$ at ~10°C. The FR is the main contributor to the ice production there (Figure S1). The second peak is at 400 hPa over the tropics, with the most contribution from the FR and IIC. The slightly larger SIP rate in the SH than that in the NH is related to the larger SH cloud and precipitation mass mixing ratios and number concentrations (Figure S2).

Figure 1b also shows that the maximum PIP rate locates at about −25 °C isotherm altitude because of the inverse dependence of immersion freezing rate on temperature as well as its positive dependence on dust concentration. However, SIP has maximum rates at lower levels because the FR and HM rates are dependent on liquid water amount, which is higher at lower levels. The seasonal variation of SIP rate shows that maximum SIP rates at about 7 kg$^{-1}$ s$^{-1}$ occur below 700 hPa altitude over 60° S in the austral winter (Figure S3). As a result, the SIP rate is larger than the PIP rate in the temperature range from −25 to 0°C in the tropics and SH. The largest ice multiplication (i.e., SIP/PIP) by 3 orders of magnitude occurs in these regions at temperatures warmer than −10°C (Figure
1c). In fact, the ice multiplication depends on PIP. The PIP rate is much larger in NH than in SH, while the SIP rate has similar magnitudes in NH as in SH. Therefore, the ice multiplication is lower in NH. The dominant role of SIP in the tropics and SH at warmer temperatures than –10°C is related to the lower PIP rate there.

3.2 SIP influence on clouds and radiation

Figure 2 shows annual zonal-mean liquid water path (LWP), ice water path (IWP), shortwave cloud forcing (SWCF), longwave cloud forcing (LWCF), and cloud fractions from CTL, SIP_CNT, and observations. SWCF and LWCF observation data are from the Cloud and the Earth’s Radiant Energy System Energy Balanced and Filled (CERES-EBAF) (Loeb et al., 2009). Six LWP observation datasets are used in the comparison with modeled LWP. Since some of the LWP datasets are only available over the ocean and there are large uncertainties with observed LWP over the land, we only include LWP over the ocean in the comparison. IWP observation data are from the CloudSat version 5.1 (Waliser et al., 2009). The IWP retrievals from CloudSat may be underestimated since the satellite is unable to penetrate through the liquid-top cloud layer, while a large amount of ice particles exist near cloud base. Since cloud ice is not separated from snow in the observation, we include the snow component in modeled IWP to be consistent with the observation. More details about the observation datasets and their uncertainties can be found in the supplementary.
The CTL experiment substantially overestimates the LWP in mid- to high
latitudes, i.e., 40° N north and 40° S south, which was also shown in previous studies
with CESM (e.g., D’Alessandro et al., 2019). The maximum bias occurs at 60° S, with
the LWP values of 140 and 58 g m\(^{-2}\) in CTL and observation, respectively. This positive
bias is largely reduced in the SIP_CNT experiment, which gives a LWP value of 60 g m\(^{-2}\)
at 60° S, in an excellent agreement with the observation. LWP is still underestimated in
the tropical regions by the model compared with the observation. This calls for the
improvement of convective cloud parameterizations in the model, as our implementation
of SIP is only for the stratiform clouds which are more important in mid- and high
latitudes than in the tropics.

Meanwhile, IWP is systematically underestimated by 10 to 30 g m\(^{-2}\) in all
latitudes in CTL. After considering SIP in the model, this low bias is much reduced in the
SIP_CNT experiment. The maximum IWP in SH is 85, 105, and 115 g m\(^{-2}\) in CTL,
SIP_CNT, and observation, respectively. We also notice the increase of IWP in the
tropics in SIP_CNT compared with CTL, despite no obvious change in LWP there.

Although the model agreement with the observation is improved in SIP_CNT, modeled
IWP is still lower than the observation in the tropics and NH mid- to high latitudes.

Patnaude et al. (2021) showed that CAM6 underestimates ice water content (IWC) for the
cirrus cloud regime (T ≤ −40°C), and the underestimation of IWC in the NH midlatitudes
is stronger than that in the SH midlatitudes. This underestimation of IWC in the cirrus

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clouds could contribute to the overall model low biases of IWP, particularly in the NH midlatitudes.

Reduced LWP leads to weaker SWCF in mid- and high-latitudes, with a maximum change of 5 W m\(^{-2}\) at 50\(^\circ\) S (Fig. 2c). SWCF from SIP_CNT is closer to the observation from 40 to 55\(^\circ\) S and from 50 to 90\(^\circ\) N compared to CTL. Meanwhile, LWCF decreases slightly in SIP_CNT compared to CTL, especially over the SH mid-latitudes. Need to mention that SWCF is more sensitive to the low-cloud amount, cloud droplet size, and LWP, while LWCF is more sensitive to the high-cloud amount, ice crystal size, and IWP. The increase of IWC mainly happens in the mixed-phase clouds below 400 hPa altitude (Figure S2), which has a small influence on LWCF. However, IWC and Ni are slightly reduced in SIP_CNT at around 300 hPa in high clouds (Figure S2), due to less cloud droplets available for the homogeneous freezing to form ice after the introduction of SIP. As a result, LWCF is slightly reduced in SIP_CNT compared to CTL. The model does poorly in simulating LWP and IWP in the tropics, while still captures the SWCF and LWCF there compared to the observation due to the model compensating errors. Our implementation of SIP influences the microphysics in stratiform mixed-phase clouds but does not directly change the properties of convective clouds, which dominate the cloud forcing in the tropics.

Table 1 shows the global annual mean cloud water path and cloud forcing from CTL and SIP_CNT as well as absolute and relative differences between these two.
experiments. Global annual mean LWP decreases by 14.6 g m\(^{-2}\) (-22\%) with the introduction of SIP, while IWP increases by 8.7 g m\(^{-2}\) (23\%). Global annual mean LWCF is reduced by 1.0 W m\(^{-2}\), while SWCF is weakened by 2.1 W m\(^{-2}\). As a result, the net cloud forcing (sum of LWCF and SWCF) is increased (positive) by 1.1 W m\(^{-2}\). Global annual mean low-, mid-, high-, and total cloud fractions are all slightly reduced. Global spatial distributions of LWP, IWP, SWCF, and LWCF changes between CTL and SIP_CTL are shown in Figure S4. In summary, the CESM2 model with SIP improves simulated LWP and IWP compared with observations, leading to weaker cloud forcings, in terms of SWCF, LWCF, and net cloud forcing.

3.3 Ice budget analysis

As shown in Figure 2, SIP has significant impacts on the global LWP and IWP. However, the SIP rate is smaller than the PIP rate, particularly in the NH and at cold temperatures (Figure 1). Furthermore, although the SIP rate is 1-3 orders of magnitude smaller than the PIP rate in the NH mid- and high latitudes, we still see significant changes in LWP and IWP there caused by SIP (Figure 2). Below, we examine the physical mechanisms behind these changes.

Figure 3 shows the global annual mean vertical profiles of rates of several key ice microphysical processes from the CTL and SIP_CNT experiments, with a complete list of ice budgets shown in Figure S5. We notice that both SIP and PIP are very small terms in the ice mass budgets compared with other terms such as water vapor deposition,
Wegener–Bergeron–Findeisen (WBF) process, ice melting, ice sedimentation, and accretion of cloud droplets by snow (Figure S5). However, when introducing the SIP into the model, Ni increases. This leads to a larger surface area of ice for water vapor deposition (Figure 3a). Consequently, the cloud ice mixing ratio increases. This further accelerates the autoconversion of cloud ice to snow (i.e., more negative rate in Figure 3b as this is a loss term for cloud ice), because this process is proportional to the cloud ice mixing ratio. The snow accretion of cloud ice only shows slight changes (Figure 3c). Higher rates of these processes give rise to higher snow mixing ratio, by which the WBF process of cloud ice and snow increases substantially (Figure 3d). This results in a strong reduction in LWP and a noticeable increase in IWP from CTL to SIP_CNT. The ice-related processes largely determine the liquid and ice partitioning in mixed-phase clouds (Solomon et al., 2015; Tan & Storelvmo, 2016; Kalesse et al., 2016). Although SIP is not the direct contributor to the changes of these process rates but it triggers a chain of interactions among these processes.

4. Comparing SIP versus PIP

We further examine the sensitivity of model results to different ice nucleation parameterizations, as shown in Figure 4. The SIP_N12 experiment shows the largest PIP rate compared with the SIP_CNT and SIP_D15 experiments. The SIP rate is quite similar among the three PIP parameterizations. Consequently, the SIP is more important in
SIP_CNT than SIP_N12. However, general patterns for the relative importance between SIP and PIP are similar among these experiments. Figure S6 shows that the LWP, IWP, SWCF, and LWCF from SIP_CNT, SIP_N12, and SIP_D15 experiments are close to each other. This suggests that the global importance of SIP is not significantly dependent on the primary ice nucleation parameterization. We also give the global annual means of LWP, IWP, cloud forcings, and cloud fractions from these experiments in Table S2. Differences among these experiments are much smaller than the differences between these experiments and the CTL experiment.

It is worth noting that the modeled primary ice nucleation currently does not include INPs from marine organic aerosol, which is identified as the dominant INPs over the remote Southern Ocean (Burrows et al., 2013; Wilson et al., 2015; Zhao et al., 2021). The model also does not include the ice nucleation on terrestrial biogenic INPs (O’Sullivan et al., 2018; Kanji et al., 2017). Future work will focus on improving the representations of primary ice nucleation on these INPs as well as representations of other SIP mechanisms (e.g., fragmentation during the sublimation of ice bridge). The modeled ice number concentration will be compared against recent observations over the Southern Ocean (e.g., from the Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study (SOCRATES)).

5. Conclusions

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Physical representations of the two new SIP mechanisms (i.e., FR and IIC) were introduced to CAM6 and evaluated for simulating the Arctic clouds against the ARM observations in our earlier study (Zhao et al., 2020). In this study, we further examine the impacts of SIP on cloud properties and cloud radiative forcing on the global scale. Our results show that the SIP occurs pretty uniformly in the two hemispheres with a maximum process rate of 5 kg m⁻¹ s⁻¹ at −10°C. SIP dominates the ice formation in moderately-cold clouds in SH and in the tropics, where the ice multiplication reaches up to three orders of magnitude at temperatures warmer than −10°C. In contrast, PIP dominates the ice formation in the relatively-cold clouds with temperatures lower than −25°C in all latitudes.

Comparing with the CTL experiment, the model with the two new SIP mechanisms decreases the LWP by 40 to 60 g m⁻² at mid- to high latitudes of both hemispheres, improving the model agreement with the observation. IWP increases at all latitudes with a globally averaged increase of 8.7 g m⁻² (23%). Finally, SIP results in weaker cloud forcings in terms of SWCF, LWCF, and net cloud forcing by 2.1, −1.0, and 1.1 W m⁻², respectively.

Through ice budget analysis, we found that SIP triggers a series of changes of ice-related processes, which ultimately change the liquid-ice partitioning and cloud radiative forcing. These findings highlight the importance of incorporating physical representations of SIP in GCMs.

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

**Data availability:** The model code is available at https://github.com/CESM-Development. The observation data of LWCF/SWCF is available at https://ceres.larc.nasa.gov/data/. The IWP data is available at https://cloudsat.atmos.colostate.edu/data. The LWP data is available at: https://climatedataguide.ucar.edu/climate-data/water-vapor-nvap.

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Reference for Supporting Information


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Figure 1. Annual zonal mean pressure-latitude cross sections of (a) secondary ice production rate (in unit of kg⁻¹s⁻¹), (b) primary ice production rate (in unit of kg⁻¹s⁻¹), and (c) ice multiplication denoted as $\log_{10}(\text{secondary ice production rate over primary ice production rate})$ from the SIP_CNT experiment. Black solid lines show the $-40^\circ\text{C}$, $-25^\circ\text{C}$ and $-10^\circ\text{C}$ isotherms, respectively.
Figure 2. Annual zonal-mean distributions of (a) liquid water path over ocean (in unit of g m$^{-2}$), (b) ice water path (in unit of g m$^{-2}$), (c) shortwave cloud forcing (in unit of W m$^{-2}$), and (d) longwave cloud forcing (in unit of W m$^{-2}$), (e) total cloud fraction (in unit of %), (f) high cloud fraction (in unit of %), (g) middle-level cloud fraction (in unit of %), and (h) low cloud fraction (in unit of %) from CTL (orange) and SIP_CNT (green) experiments, and observations (OBS, shown as gray dashed lines). Modeled IWP includes the snow component to be consistent with the observation, since cloud ice is not separated from snow in the observation. In order to fairly compare modeled cloud properties against satellite observations, the MODIS cloud simulator is used to calculate the modeled cloud fraction.
Figure 3. Global annual mean vertical profiles of rates (in unit of $10^{-10}$ kg kg$^{-1}$ s$^{-1}$) of (a) deposition of water vapor on cloud ice and snow, (b) autoconversion of cloud ice to snow, (c) accretion of cloud ice by snow, and (d) Wegener–Bergeron–Findeisen (WBF) process of cloud ice and snow from CTL (orange) and SIP_CNT (green) experiments.
Figure 4. Annual zonal mean pressure-latitude cross sections of primary ice production rate (first row, in unit of kg$^{-1}$s$^{-1}$), secondary ice production rate (second row, in unit of kg$^{-1}$s$^{-1}$), and ice multiplication denoted as Log$_{10}$(secondary ice production rate over primary ice production rate) (third row) from SIP_CNT (first column), SIP_D15 (second column), and SIP_N12 (third column) experiments. Black solid lines show the −40, −25, and −10°C isotherms.
Table 1. Global annual means of cloud liquid water path (LWP), ice water path (IWP), shortwave, longwave and net cloud forcings (SWCF, LWCF, and net CF), and low-, mid-, high-, and total cloud fractions from CTL and SIP_CNT experiments as well as absolute and relative differences (in parenthesis, %) between these two experiments.

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<td>-22.89</td>
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