

Spaceborne evidence that ice-nucleating particles influence cloud phase

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Key Points:

- Ice-nucleating particles (INPs) control ice formation in high-latitude clouds.
- Sea ice and snow inhibit the local emission of INPs, which directly influences cloud phase in the Arctic and Southern Ocean.
- This has implications for the predicted negative cloud phase feedback with future warming and the associated sea ice and snow cover loss.

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Abstract

Mixed-phase clouds (MPCs), which consist of both supercooled cloud droplets and ice crystals, play an important role in the Earth's radiative energy budget and hydrological cycle. In particular, the fraction of ice crystals in MPCs determines their radiative effects, precipitation formation and lifetime. In order for ice crystals to form in MPCs, ice-nucleating particles (INPs) are required. However, a large-scale relationship between INPs and ice initiation in clouds has yet to be observed. By analyzing satellite observations of the typical transition temperature (T^*) where MPCs become more frequent than liquid clouds, we constrain the importance of INPs in MPC formation. We find that over the Arctic and Southern Ocean, snow and sea ice cover significantly reduces T^* . This indicates that the availability of INPs is essential in controlling cloud phase evolution and that local sources of INPs in the high-latitudes play a key role in the formation of MPCs.

Plain Language Summary

Mixed-phase clouds (MPCs), which consist of both liquid droplets and ice crystals, play an important role for the Earth's climate system. For example, the number of ice crystals in MPCs determines how much sunlight is reflected by the cloud and how efficiently the cloud can form precipitation. The formation of ice crystals in MPCs requires a special subset of aerosol particles called ice-nucleating particles (INPs). INPs are required for liquid cloud droplets to freeze at temperatures warmer than -36°C . However, a large-scale relationship between INPs and ice formation in clouds has yet to be observed. Using satellite observations, we determine the transition temperature (T^*) where MPCs become more frequent than liquid clouds and find that T^* is strongly dependent on snow and sea ice cover over the Arctic and Southern Ocean. This indicates that sea ice and snow cover act as a lid that inhibits the emission of INPs from the ocean. In a warming world with retreating sea ice and snow cover, our results suggest that clouds in these regions will contain ice crystal at warmer temperatures than previously estimated and thus, have potential implications for future warming predictions.

1 Introduction

The amount of liquid and ice within MPCs influences precipitation formation, cloud lifetime, and electrification (Cantrell & Heymsfield, 2005). Simultaneously, the thermodynamic phase composition controls the radiative properties of MPCs due to the different scattering properties between liquid water and ice. In a warming climate, MPCs are believed to transition towards a state with more liquid water and a higher albedo, which limits future warming (Bjorndal et al., 2020; Zelinka et al., 2020). This cloud phase feedback makes the accurate representation of ice crystal concentrations in MPCs in Earth System Models (ESMs) essential for correctly predicting the future climate (Tan et al., 2016; Forster et al., 2021). But what controls the formation of ice and the thermodynamic phase composition in MPCs?

The importance of INPs for forming ice in MPCs is undisputed. Laboratory experiments show that pure water does not freeze without the presence of an INP until it is supercooled to around -36°C . Therefore, field measurements including precipitation sampling (Vali, 1971; Petters & Wright, 2015), airborne (Borys, 1989; Rogers et al., 2001; Pratt et al., 2009; DeMott et al., 2010), ship (Wilson et al., 2015; Welti et al., 2020), and mountaintop measurements (Lacher et al., 2017) have been conducted to investigate the abundance of INPs, globally. These studies have found that INP concentrations can vary by several orders of magnitude at a given temperature. This variability is partially explained by the location and type of aerosol acting as INPs (Kanji et al., 2017). Close to the Earth's major deserts, dust is the primary source of INPs, especially at temperatures below -15°C (Atkinson et al., 2013; Murray et al., 2012; Boose et al., 2016). Meanwhile,

in more remote regions and at higher temperatures, biological sources such as sea spray aerosol are believed to be the most important source of INPs (Burrows et al., 2013; Schnell & Vali, 1975; Wilson et al., 2015; C. S. McCluskey et al., 2018; Irish et al., 2019).

Based on the fundamental importance of INPs for ice crystal formation in MPCs, INP parametrizations have been developed to account for different aerosol species and the observed variability from field and laboratory studies. When implemented into ESMs, different INP parametrizations can have profound effects on both MPC optical properties and lifetimes. However, when in situ ice crystal number concentrations are compared with INP concentrations, they seldomly agree (Mignani et al., 2019) and ice crystal concentrations often exceed INP concentrations by several orders of magnitude (Ladino et al., 2017; Ramelli et al., 2021; Rangno & Hobbs, 2001). This would suggest that INPs are not as important for ice crystal formation in MPCs as laboratory studies indicate. The main explanation for this discrepancy is secondary ice production (SIP) (Korolev & Leisner, 2020; Hallett & Mossop, 1974), which has been shown to rapidly increase the concentration of ice crystals in MPCs through what has been described as a cascading process. Nevertheless, the occurrence and efficiency of secondary ice processes is still an area of open research.

Another and larger scale approach to assess the influence of INPs on MPCs has been through the so-called supercooled liquid fraction (SLF, ratio of supercooled liquid to ice). The SLF and ambient aerosol concentration (a proxy for INPs) comparisons show that there is a correlation, but a weak dependence between dust aerosols and the SLF of MPCs at a given temperature (Choi et al., 2010; Tan et al., 2014). However, the SLF is prone to the influence of dynamics (vertical velocities), the Wegener-Bergeron-Findeisen process (Korolev, 2007), and secondary ice processes, thereby masking the importance of INPs for the distribution of the cloud phase.

Therefore, with the exception of laboratory and modeling studies, direct evidence of the importance of INPs on MPC formation and subsequent thermodynamic phase composition has yet to be observed or quantified. Here we show that by using the transition temperature from supercooled liquid clouds to MPCs, as observed by satellites, the influence of INPs on the thermodynamic phase composition in MPCs can be disentangled. In particular, we focus this analysis on the high-latitudes, where MPCs are abundant (Korolev et al., 2017). Additionally, field studies indicate that local INP emissions have a strong seasonal dependence in the Arctic (Wex et al., 2019; Tobo et al., 2019), providing a unique opportunity to analyse the influence of differing INP concentrations on MPCs. We find that this transition temperature is significantly suppressed over sea ice and snow, confirming that INPs play a critical role in the evolution of cloud phase and that the INPs in this region are primarily of a local nature.

2 Materials and Methods

Here we use satellite observations from CloudSat and the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO, Stephens et al., 2002; Winker et al., 2010) to discriminate between single-layer liquid only (LO) and liquid-topped MPCs (LTMPs) and combine them with cloud top temperatures (CTTs) from atmospheric reanalysis data to characterize the occurrence of both cloud types as a function of CTT (see Figs. 1A and B). The warmest CTT when LTMPs become more frequent than LO clouds, is hereafter referred to as T^* (see Fig. 1C). We perform this analysis on a $5^\circ \times 5^\circ$ grid (see Fig. 1D) for each season over 9 years (2006-2017). In this section, the calculation of T^* , its significance, and the averaging procedure are described together with the processing of the sea ice data used in the interpretation of the results.

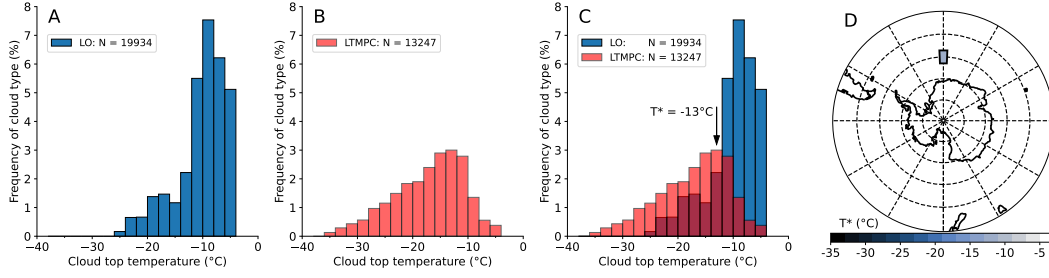


Figure 1. Frequency of cloud types (in %) with respect to cloud top temperature (bin width: 2 °C) for the 5°x5° grid cell centered at 60°S and 0°E combining 9 austral summer seasons (DJF, 2006-2009 and 2012-2016). (A) for liquid-only clouds, (B) for liquid-top mixed-phase clouds, (C) The combination of LO and LTMPC frequency distributions yields an exemplary T^* of -13°, °C.

2.1 Satellite data and definition of cloud regimes

For the discrimination between LO clouds and LTMPCs, we use the data product 2B-CLDCLASS-LIDAR (Sassen et al., 2008), which combines observations from the cloud profiling radar (CPR) on CloudSat (Stephens et al., 2002) and the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) (Winker et al., 2007) on CALIPSO (Winker et al., 2010). The 2B-CLDCLASS-LIDAR product utilizes the different sensitivities of the radar and lidar to liquid droplets and ice crystals to determine the phase of a cloud layer. The logics of the phase determination algorithm are based on a temperature dependent radar reflectivity threshold (Zhang et al., 2010), the integrated attenuated lidar backscattering coefficient, and cloud base and top temperatures from atmospheric reanalysis (Wang, 2019). This way, each individual cloud layer of the CPR profile gets assigned a phase (variable `CloudPhase`: 'ice', 'mixed', or 'water'). Here we restrict our analysis to single-layer clouds. The cloud phase information comes with a confidence level assigned to the cloud phase (variable `CloudPhaseConfidenceLevel`). The confidence value generally ranges from zero to 10, where 10 indicates the highest confidence level. While it is not recommended to use data with a confidence level of five or lower, we further restrict our analysis to cloud phase confidence levels of seven or higher. For each cloudy profile, we retrieve the cloud top temperature (CTT) from the ECMWF-AUX dataset that contains ancillary European Centre for Medium-Range Weather Forecast (ECMWF) state variable data interpolated to each CPR vertical bin. For the definition of the different cloud types, we include observations with CTTs below 270 K to stay away from the temperature limits of the phase determination algorithm. We define LO clouds and MPCs based on the `CloudPhase` variable of 'water' or 'mixed', respectively. For the definition of LTMPCs, we further use the `Water_layer_top` variable from the 2B-CLDCLASS-LIDAR product, which indicates the location of a possible water layer in MPCs. We define LTMPCs as all MPCs where the `Water_layer_top` is within 3 vertical radar bins (90 m) of the cloud top height (variable `CloudLayerTop`). The CTT of all LO and LTMPC are combined into 5°x5° grid cells over the entire globe. Then the CTTs of the LO and LTMPCs are binned into 2°C temperature bins by season for the years 2006 to 2017.

For each 5°x5° grid cell, the LO and LTMPC observations are normalized by the total number of single-layer observations (LO, MPCs, ice-only) in the given cell. Finally, T^* is then defined as the warmest CTT bin, where LTMPCs are more frequent than LO clouds and where at least 10 LTMPCs were observed within that given season summed over the nine year period.

2.2 Significance of T^*

To test the robustness and significance of T^* , its calculation is repeated 100 times using random sampling with replacement (bootstrapping) of the original observations of both LO and LTMPCs for each grid cell and each season. The significance of T^* is estimated based on the distribution of the T^* values from the bootstrapped calculations by calculating the standard error (SE) from the standard deviation σ of the bootstrapped T^* and the number of bootstrapped T^* values (100):

$$SE = \frac{\sigma}{\sqrt{100}} \quad (1)$$

From the SE, the 99 % confidence interval (CI) can be calculated as:

$$CI = 2.58 \cdot SE \quad (2)$$

Grid cells are classified as insignificant if the CI is larger than 0.5°C (corresponding to a T^* confined to about 1 bin during bootstrapping) and are excluded from the analysis. Most grid cells analysed within this study show very robust T^* values (see Fig. S1).

For the difference in T^* (Summer - Winter) in Fig. 2C and F, the grid cells where the sum of the summer and winter CIs is larger than the absolute value of the T^* difference (min/max error propagation) are treated as insignificant.

2.3 Sea ice data

The sea ice concentration data is from the Institute of Environmental Physics (IUP), University of Bremen, based on the ARTIST Sea Ice (ASI) algorithm (Spren et al., 2008). The ASI retrieval is applied to microwave radiometer data of the AMSR-E (Advanced Microwave Scanning Radiometer for EOS) on the Aqua satellite and AMSR2 (Advanced Microwave Scanning Radiometer 2) on GCOM-W1 sensors, which were reprocessed in 2018 for both platforms with the same parameters. The sea ice edge as visible in Fig. 2 is calculated using the following steps: (1) Retrieval of the dates for the Arctic/Antarctic sea ice maximum/minimum for each year (Grosfeld et al., 2016) and calculation of the multi-year average of the sea ice maxima/minima from these days. If the maximum/minimum occurred in March/September, we used the last day of the respective season (February/August) in the calculation. (2) The sea ice edge is defined where the sea ice concentration is at least 15 %. (3) Re-gridding of the sea ice data on a regular $0.25^\circ \times 0.25^\circ$ grid using bilinear interpolation.

2.4 Averaging of T^*

We perform area-weighted averaging of T^* for the different regions and seasons based on the sea ice concentrations and land/ocean masks (from ASI data set) that we re-gridded on the $5^\circ \times 5^\circ$ grid of T^* using bilinear interpolation. Further, grid cells with insignificant T^* values are excluded from the averaging. The resulting masks used for the calculation of the average T^* values in Table 1 are displayed in Figure S2.

3 Results and discussion

T^* is based on the underlying principle that as the CTT of LO clouds cool, the initiation of ice becomes more likely as a larger fraction of aerosols can act as INPs (Fletcher, 1962; Meyers et al., 1992) and therefore, the probability of observing LTMPCs increases. As shown by the exemplary histograms in Fig. 1, the observed frequency of LTMPCs increases at colder temperatures, as expected, and exceeds that of LO clouds at -13°C . Therefore, in this example the T^* of -13°C represents the typical temperature at which INPs act to alter the cloud phase for this region.

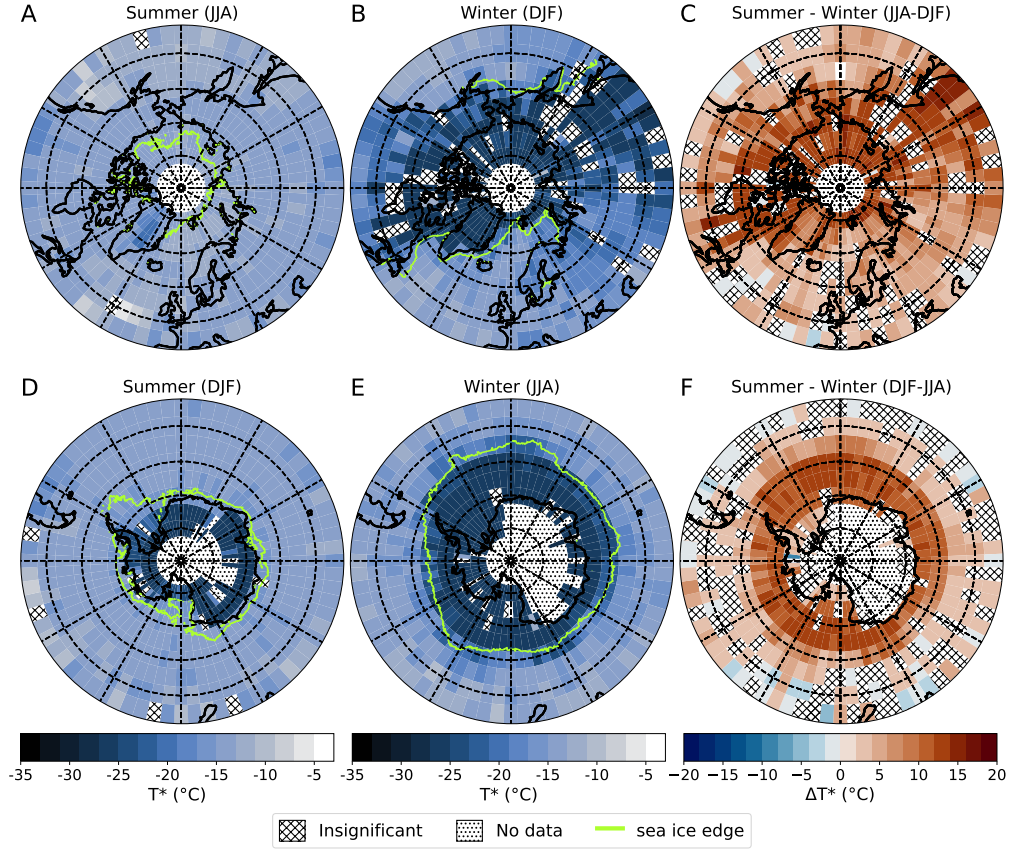


Figure 2. Seasonal T^* over the Arctic and the Southern Ocean based on observations between 2006-2017. Grid cells where T^* calculations are insignificant (on a 99 % confidence level) are hatched, while dotted areas have no data. The green line shows the average minimum/maximum sea ice edge between 2006-2017, defined as the 15% sea ice concentration line for the given season. Arctic T^* during Summer (JJA), Winter T^* (DJF) and the difference between Summer and Winter (Summer minus Winter) are shown in panel A,B, and C, respectively. Similarly, the Southern Ocean T^* during Austral Summer (DJF), Winter (JJA) and the difference between Summer and Winter (Summer minus Winter) are shown in panel D,E and F, respectively.

When calculating T^* over the Arctic as a function of sea and land mask (see Tab. 1), we find that during the summer time (JJA), T^* is -13°C over the Ocean and -14°C over land (Fig. 2A). At these temperature, biological INPs are expected to be dominant (Kanji et al., 2017) and indeed field studies have shown that INPs are primarily biological during the Arctic summer (Tobo et al., 2019; Creamean et al., 2019). T^* is homogeneous over both the land and the ocean and this suggests that the abundance of INPs and their efficiency is rather similar throughout the region. The only exception is over the Greenland ice sheet (lower T^* values, see Fig. 2A) where due to its high altitude and frequently cold temperatures, INPs are expected to be washed out during transport to this area (Stopelli et al., 2015) or relatively INP depleted air from the free troposphere (Lacher et al., 2018) descends over the ice sheet (Guy et al., 2021). Consistently, field measurements have shown that INP concentrations are lower over the Greenland ice sheet than elsewhere during the Arctic summertime (Wex et al., 2019).

Table 1. Area-weighted averages of significant T^* (in $^{\circ}\text{C}$) for different regions and seasons. The masks used for averaging are displayed in Fig. S2.

Arctic	ocean	land	sea ice
Summer (JJA)	-13	-14	-
Winter (DJF)	-16	-23	-24
Antarctica	ocean	land	sea ice
Summer (DJF)	-15	-	-
Winter (JJA)	-17	-	-27

In contrast, during the winter months (DJF) the T^* over the sea ice region (green line in Fig. 2) drops to -24°C , while over open ocean it slightly decreases to -16°C (Fig. 2B, compare Tab. 1). Similarly, during the winter months snow cover reduces T^* to -23°C over land. Thus, the largest seasonal differences in T^* are observed in regions covered by snow and sea ice during the winter (Fig. 2C). Previous ship (Bigg, 1996; Bigg & Leck, 2001) and coastal (Creamean et al., 2018) measurements also observed a dependence of the INP concentration on the extent of snow and sea ice coverage, with a decrease and increase in INP concentration during the Fall freeze up and Spring thaw, respectively. A reduction in wintertime INP concentration was also observed at an inland Arctic location in Alaska (Borys, 1983) and a Boreal Forest in Finland (Schneider et al., 2021). Similarly, Wex et al. (2019) observed an increase in INPs during the snow-free summer months and a decrease during the winter months at four different measurement locations in the Arctic. Their back trajectory analysis showed that the highest INP concentrations were associated with air mass interaction with snow-free terrain and open water, while the lowest concentrations came from the sea ice and snow.

Airborne Arctic INP measurements (Borys, 1989; Hartmann et al., 2020) also observed a decrease in INP concentration over sea ice and snow cover. The only exception was over open leads in the sea ice (Rogers et al., 2001; Hartmann et al., 2020; Curry et al., 2000), which further indicates that sea ice inhibits the emissions of INPs. This lack of available biological INPs has also been used to explain the lower temperatures required to observe MPCs in the Arctic relative to the midlatitudes and tropics (Costa et al., 2017). Furthermore, Griesche et al. (2021) observed a decrease in the frequency of ice containing clouds when they were decoupled from the ocean surface, also indicating that marine INPs are essential for ice formation in Arctic clouds.

These studies are in agreement with our findings, that sea ice and snow cover significantly reduce T^* . Thus, the combination of previous INP studies with the T^* metric presented here demonstrates the large-scale influence INPs have on the formation of MPCs in the Arctic.

When calculating T^* over the Southern Ocean and separating by season (Figs. 2D-F, Tab. 1), it is apparent that T^* is -15°C during the Austral summer (DJF) and homogeneous over the entire region. This is consistent with summertime ship measurements conducted in the Southern Ocean, where INPs were typically observed at temperatures above -14°C (Welti et al., 2020; C. McCluskey et al., 2018) and their concentration only varied by about one order of magnitude at -15°C (Welti et al., 2020). When comparing to the Arctic, the summertime T^* in the Southern Ocean is about 2°C cooler. This may in part be due to higher biological activity in the Arctic Ocean (McCluskey et al., 2019; Irish et al., 2017) and a larger land area where glacial out wash can be emitted and act as a local episodic INP source (Sanchez-Marroquin et al., 2020; Tobo et al., 2019; Rinaldi et al., 2021). Meanwhile, during the winter (JJA), a similar relationship between

sea ice coverage and T^* emerges (Fig. 2E). The T^* over the sea ice region drops to -27°C , while over open ocean regions the T^* slightly decreases to -17°C . This indicates that the sea ice acts to inhibit the emission of INPs and directly impacts cloud phase over the Southern Ocean as well. It is well known that the ocean is an important source of INPs in the Southern Ocean (Schnell, 1977; Burrows et al., 2013), which is far from the Earth's deserts (DeMott et al., 2016; C. McCluskey et al., 2018). Indeed, INP observations from South Pole were significantly lower than at a coastal site (Belosi et al., 2014) and increased when airmass back trajectories originated from the coast (Ardon-Dryer et al., 2011). Modelling studies have shown that replacing dust-based with marine-based INP parametrizations greatly improves the representation of clouds over the Southern Ocean (Vergara-Temprado et al., 2018; Frey & Kay, 2018). This further provides evidence that the Southern Ocean is the primary source of INPs over this region and when it is covered with sea ice, fewer INPs are emitted. Therefore, our results indicate that the decrease in T^* over the sea ice is a result of the sea ice acting as a lid that inhibits the emission of INPs from the Southern Ocean and, in turn, hinders the initiation of MPCs in this region.

Previous remote sensing observations of cloud phase over the Southern Ocean have also observed a spatial pattern in the occurrence of MPCs (e.g., Mace et al., 2020, 2021) with a maximum in the vicinity of the so-called Antarctic Polar Front (APF, Freeman & Lovenduski, 2016). Mace et al. (2021) attributed this relationship to potentially enhanced vertical updrafts in convective clouds over the APF due to warmer sea surface temperatures, which would loft ice crystals from lower layers of clouds to their top where a lidar-depolarization based cloud classification algorithm (Mace et al., 2020) would classify them as mixed-phase. Additionally, they highlight that these enhanced updrafts would act as a production zone for larger cloud droplets, which have been shown to be more efficient for SIP (Lauber et al., 2018; Keinert et al., 2020). Although we cannot rule out the importance of SIP on the classification of a cloud as LTMPC, through the combined use of lidar and radar observations in the 2B-CLDCLASS-LIDAR product we can reduce the importance of ice crystals being lofted to cloud top for the classification of LTMPCs. Regardless of the importance of SIP in producing ice in LTMPCs, primary ice formed on INPs is still required and thus, T^* is representative of when INPs are responsible for controlling the cloud phase over the SO. Furthermore, when comparing T^* values with the location of the APF in the summertime (see Fig. 3 in Freeman & Lovenduski, 2016) there is no clear dependence, indicating that the observed seasonal variability in T^* is associated with the sea ice extent and its ability to inhibit INP emissions.

With this in mind, it is important to note that there are well-documented differences in high-latitude clouds over the open ocean and sea ice due to differences in surface fluxes (e.g. heat and moisture) and thermodynamic structure (Palm et al., 2010; Eirund et al., 2019; Sotiropoulou et al., 2016; Young et al., 2017). However, to our knowledge these differences would not lead to LO clouds occurring more frequently at colder temperatures over sea ice than over open water as we observe. Furthermore, when comparing the cloud top heights and occurrence of LO and LTMPCs over the Southern Ocean (see Fig. S3), we find that they overlap and occur at the same heights in the troposphere, regardless of whether they form over open ocean or sea ice. Therefore, the observed variability in T^* can only be explained by the variability in the efficiency and concentration of INPs present during the onset of ice formation and MPC initiation.

4 Atmospheric implications

The apparent relationship between the suppressed T^* and sea ice and snow cover (Fig. 2), which is well documented as a region with reduced INP concentrations (Wex et al., 2019; Bigg & Leck, 2001; Creamean et al., 2019), indicates that INPs play a critical role in the initiation of MPCs. Furthermore, this relationship provides additional evidence that INPs in the high-latitudes are primarily of a local origin. Ultimately, through the use of T^* , we highlight the global relevance of INPs on MPC formation, confirming

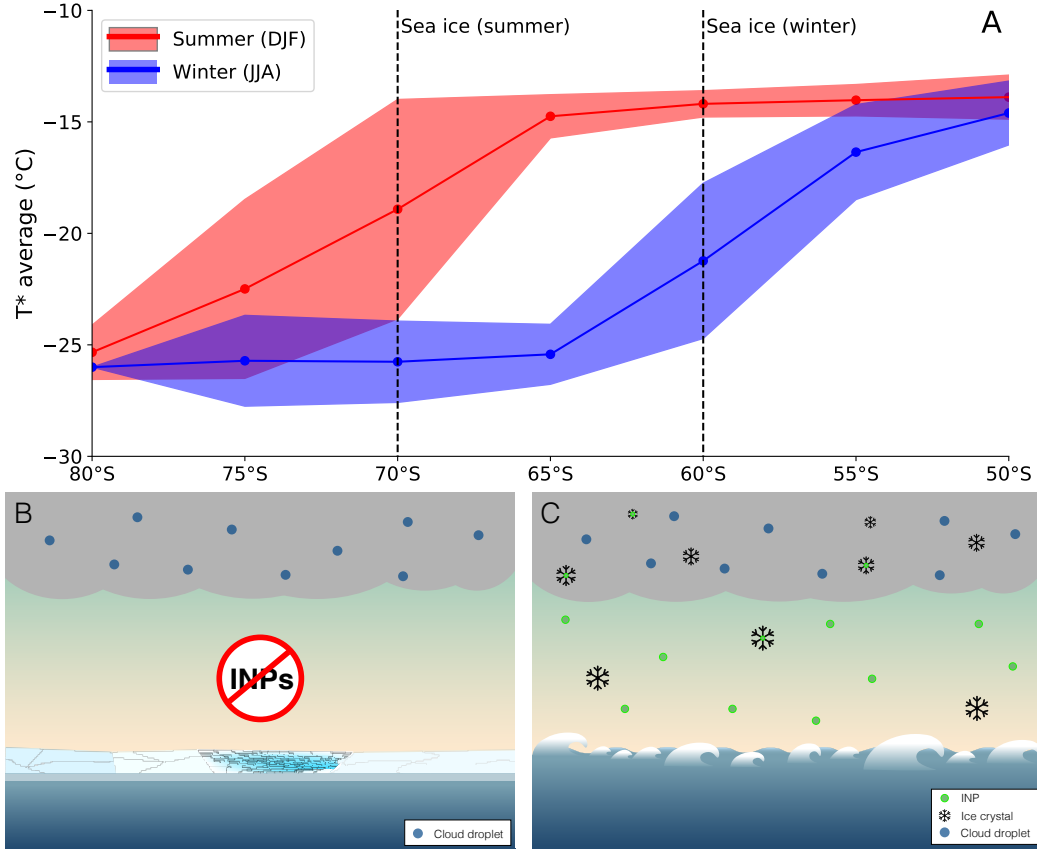


Figure 3. (A) Latitudinal average (line) and standard deviation (fill) of T^* during Austral summer (red) and winter (blue) over the Southern Ocean as a northward cross section from 80°S to 50°S. (B-C) Conceptual overview of how sea ice cover influences INP sources and, consequently, ice formation in MPCs.

laboratory studies dating back to the 1940s (Vonnegut, 1947) that showed the importance of INPs for MPC formation.

Based on these findings, we conclude that differing INP parametrizations are required over ice/snow-covered and ice/snow-free portions of the high-latitudes to account for the observed seasonal variations in the MPC transition temperature, T^* . This is especially important for future climate projections, where in a warming climate, sea ice and snow cover are projected to decrease (Fox-Kemper et al., 2021) and therefore, although temperatures will rise, INPs may become more abundant due to newly available source regions (i.e. ice and snow free areas).

An increase in the abundance of high-latitude INPs could have profound effects on the cloud-phase feedback (Murray et al., 2021; Prenni et al., 2007), which has so far been projected to limit warming over the Southern Ocean (Forster et al., 2021; Zelinka et al., 2020; Bjordal et al., 2020; Tan et al., 2016). The warming-induced INP increase could weaken, or even reverse, the projected increase in LO clouds with warming. This would have major implications for both the magnitude and sign of the Southern Ocean cloud feedback, thus shaping the climate evolution of the region itself and ultimately, the future climate of the entire planet. Figure 3 shows the T^* averaged by latitude over the Southern Ocean. The stark contrast in T^* over sea ice and open ocean indicates that in a warming world with sea ice retreat, T^* over formerly ice covered regions will increase

to -15 °C. As the ice covered regions currently have a T^* of approximately -25 °C, this would suggest that a warming of 10 °C would be required to significantly offset the formation of MPCs over future ice free regions of the Southern Ocean. Therefore, without a detailed quantification of the seasonal nature of INPs in the high-latitudes and subsequent inclusion in ESMs, the influence of the negative cloud phase-feedback on buffering future warming will remain uncertain.

5 Open Research

The standard CloudSat (Stephens et al., 2002) and CALIPSO (Winker et al., 2010) data products (version R05) used in this study (2B-CLDCLASS-LIDAR, ECMWF-AUX) were downloaded from the CloudSat Data Processing Center’s (at Cooperative Institute for Research in the Atmosphere, Colorado State University, Fort Collins) website (<http://www.cloudsat.cira.colostate.edu>).

The sea ice concentration data is from the Institute of Environmental Physics (IUP), University of Bremen, based on the ARTIST Sea Ice (ASI) algorithm (Spreen et al., 2008). The daily data sets (Melsheimer & Spreen, 2020b, 2020a, 2019b, 2019a) were downloaded for the years 2006-2017 from the data publisher PANGAEA. The dates for the Arctic and Antarctic sea ice maximum/minimum for each year were retrieved from <https://www.meereisportal.de> (Grosfeld et al., 2016).

The code used to analyse the satellite data will be made available on a public GitHub repository pending final publication of this manuscript.

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