Spaceborne evidence that ice-nucleating particles influence cloud phase

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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.

Spaceborne evidence that ice-nucleating particles influence cloud phase

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Key Points:

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- 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.

 $^{^{*}\}mathrm{Both}$ authors contributed equally to this work.

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11 Abstract

Mixed-phase clouds (MPCs), which consist of both supercooled cloud droplets and ice 12 crystals, play an important role in the Earth's radiative energy budget and hydrologi-13 cal cycle. In particular, the fraction of ice crystals in MPCs determines their radiative 14 effects, precipitation formation and lifetime. In order for ice crystals to form in MPCs, 15 ice-nucleating particles (INPs) are required. However, a large-scale relationship between 16 INPs and ice initiation in clouds has yet to be observed. By analyzing satellite obser-17 vations of the typical transition temperature (T^{*}) where MPCs become more frequent 18 than liquid clouds, we constrain the importance of INPs in MPC formation. We find that 19 over the Arctic and Southern Ocean, snow and sea ice cover significantly reduces T^{*}. This 20 indicates that the availability of INPs is essential in controlling cloud phase evolution 21 and that local sources of INPs in the high-latitudes play a key role in the formation of 22

²³ MPCs.

24 Plain Language Summary

Mixed-phase clouds (MPCs), which consist of both liquid droplets and ice crystals, 25 play an important role for the Earth's climate system. For example, the number of ice 26 crystals in MPCs determines how much sunlight is reflected by the cloud and how ef-27 ficiently the cloud can form precipitation. The formation of ice crystals in MPCs requires 28 a special subset of aerosol particles called ice-nucleating particles (INPs). INPs are re-29 quired for liquid cloud droplets to freeze at temperatures warmer than -36 °C. However, 30 a large-scale relationship between INPs and ice formation in clouds has yet to be observed. 31 Using satellite observations, we determine the transition temperature (T^*) where MPCs 32 become more frequent than liquid clouds and find that T^* is strongly dependent on snow 33 and sea ice cover over the Arctic and Southern Ocean. This indicates that sea ice and 34 snow cover act as a lid that inhibits the emission of INPs from the ocean. In a warm-35 ing world with retreating sea ice and snow cover, our results suggest that clouds in these 36 regions will contain ice crystal at warmer temperatures than previously estimated and 37 thus, have potential implications for future warming predictions. 38

39 1 Introduction

The amount of liquid and ice within MPCs influences precipitation formation, cloud 40 lifetime, and electrification (Cantrell & Heymsfield, 2005). Simultaneously, the thermo-41 dynamic phase composition controls the radiative properties of MPCs due to the differ-42 ent scattering properties between liquid water and ice. In a warming climate, MPCs are 43 believed to transition towards a state with more liquid water and a higher albedo, which 44 limits future warming (Bjordal et al., 2020; Zelinka et al., 2020). This cloud phase feed-45 back makes the accurate representation of ice crystal concentrations in MPCs in Earth 46 System Models (ESMs) essential for correctly predicting the future climate (Tan et al., 47 2016; Forster et al., 2021). But what controls the formation of ice and the thermody-48 namic phase composition in MPCs? 49

The importance of INPs for forming ice in MPCs is undisputed. Laboratory ex-50 periments show that pure water does not freeze without the presence of an INP until it 51 is supercooled to around -36 $^{\circ}\mathrm{C}.$ Therefore, field measurements including precipitation 52 sampling (Vali, 1971; Petters & Wright, 2015), airborne (Borys, 1989; Rogers et al., 2001; 53 Pratt et al., 2009; DeMott et al., 2010), ship (Wilson et al., 2015; Welti et al., 2020), and 54 mountaintop measurements (Lacher et al., 2017) have been conducted to investigate the 55 abundance of INPs, globally. These studies have found that INP concentrations can vary 56 by several orders of magnitude at a given temperature. This variability is partially ex-57 plained by the location and type of aerosol acting as INPs (Kanji et al., 2017). Close to 58 the Earth's major deserts, dust is the primary source of INPs, especially at temperatures 59 below -15 °C (Atkinson et al., 2013; Murray et al., 2012; Boose et al., 2016). Meanwhile, 60

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, 64 INP parametrizations have been developed to account for different aerosol species and 65 the observed variability from field and laboratory studies. When implemented into ESMs, 66 different INP parametrizations can have profound effects on both MPC optical proper-67 ties and lifetimes. However, when in situ ice crystal number concentrations are compared 68 69 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 70 al., 2017; Ramelli et al., 2021; Rangno & Hobbs, 2001). This would suggest that INPs 71 are not as important for ice crystal formation in MPCs as laboratory studies indicate. 72 The main explanation for this discrepancy is secondary ice production (SIP) (Korolev 73 & Leisner, 2020; Hallett & Mossop, 1974), which has been shown to rapidly increase the 74 concentration of ice crystals in MPCs through what has been described as a cascading 75 process. Nevertheless, the occurrence and efficiency of secondary ice processes is still an 76 area of open research. 77

Another and larger scale approach to assess the influence of INPs on MPCs has been 78 through the so-called supercooled liquid fraction (SLF, ratio of supercooled liquid to ice). 79 The SLF and ambient aerosol concentration (a proxy for INPs) comparisons show that 80 there is a correlation, but a weak dependence between dust aerosols and the SLF of MPCs 81 at a given temperature (Choi et al., 2010; Tan et al., 2014). However, the SLF is prone 82 to the influence of dynamics (vertical velocities), the Wegener-Bergeron-Findeisen pro-83 cess (Korolev, 2007), and secondary ice processes, thereby masking the importance of 84 INPs for the distribution of the cloud phase. 85

Therefore, with the exception of laboratory and modeling studies, direct evidence 86 of the importance of INPs on MPC formation and subsequent thermodynamic phase com-87 position has yet to be observed or quantified. Here we show that by using the transition 88 temperature from supercooled liquid clouds to MPCs, as observed by satellites, the in-89 fluence of INPs on the thermodynamic phase composition in MPCs can be disentangled. 90 In particular, we focus this analysis on the high-latitudes, where MPCs are abundant 91 (Korolev et al., 2017). Additionally, field studies indicate that local INP emissions have 92 a strong seasonal dependence in the Arctic (Wex et al., 2019; Tobo et al., 2019), provid-93 ing a unique opportunity to analyse the influence of differing INP concentrations on MPCs. 94 We find that this transition temperature is significantly suppressed over sea ice and snow, 95 confirming that INPs play a critical role in the evolution of cloud phase and that the INPs 96 in this region are primarily of a local nature. 97

⁹⁸ 2 Materials and Methods

Here we use satellite observations from CloudSat and the Cloud-Aerosol Lidar and 99 Infrared Pathfinder Satellite Observation (CALIPSO, Stephens et al., 2002; Winker et 100 al., 2010) to discriminate between single-layer liquid only (LO) and liquid-topped MPCs 101 (LTMPCs) and combine them with cloud top temperatures (CTTs) from atmospheric 102 reanalysis data to characterize the occurrence of both cloud types as a function of CTT 103 (see Figs. 1A and B). The warmest CTT when LTMPCs become more frequent than LO 104 clouds, is hereafter referred to as T^* (see Fig. 1C). We perform this analysis on a $5^{\circ}x5^{\circ}$ 105 grid (see Fig. 1D) for each season over 9 years (2006-2017). In this section, the calcula-106 tion of T^{*}, its significance, and the averaging procedure are described together with the 107 processing of the sea ice data used in the interpretation of the results. 108



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

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For the discrimination between LO clouds and LTMPCs, we use the data product 110 2B-CLDCLASS-LIDAR (Sassen et al., 2008), which combines observations from the cloud 111 profiling radar (CPR) on CloudSat (Stephens et al., 2002) and the Cloud-Aerosol Lidar 112 with Orthogonal Polarization (CALIOP) (Winker et al., 2007) on CALIPSO (Winker 113 et al., 2010). The 2B-CLDCLASS-LIDAR product utilizes the different sensitivities of 114 the radar and lidar to liquid droplets and ice crystals to determine the phase of a cloud 115 layer. The logics of the phase determination algorithm are based on a temperature de-116 pendent radar reflectivity threshold (Zhang et al., 2010), the integrated attenuated li-117 dar backscattering coefficient, and cloud base and top temperatures from atmospheric 118 reanalysis (Wang, 2019). This way, each individual cloud layer of the CPR profile gets 119 assigned a phase (variable CloudPhase: 'ice', 'mixed', or 'water'). Here we restrict our 120 analysis to single-layer clouds. The cloud phase information comes with a confidence level 121 assigned to the cloud phase (variable CloudPhaseConfidenceLevel). The confidence value 122 generally ranges from zero to 10, where 10 indicates the highest confidence level. While 123 it is not recommended to use data with a confidence level of five or lower, we further re-124 strict our analysis to cloud phase confidence levels of seven or higher. For each cloudy 125 profile, we retrieve the cloud top temperature (CTT) from the ECMWF-AUX dataset 126 that contains ancillary European Centre for Medium-Range Weather Forecast (ECMWF) 127 state variable data interpolated to each CPR vertical bin. For the definition of the dif-128 ferent cloud types, we include observations with CTTs below 270 K to stay away from 129 the temperature limits of the phase determination algorithm. We define LO clouds and 130 MPCs based on the CloudPhase variable of 'water' or 'mixed', respectively. For the def-131 inition of LTMPCs, we further use the Water_layer_top variable from the 2B-CLDCLASS-132 LIDAR product, which indicates the location of a possible water layer in MPCs. We de-133 fine LTMPCs as all MPCs where the Water_layer_top is within 3 vertical radar bins 134 (90 m) of the cloud top height (variable CloudLayerTop). The CTT of all LO and LTMPC 135 are combined into $5^{\circ}x5^{\circ}$ grid cells over the entire globe. Then the CTTs of the LO and 136 LTMPCs are binned into 2°C temperature bins by season for the years 2006 to 2017. 137

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.

¹⁴³ 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):

$$SE = \frac{\sigma}{\sqrt{100}} \tag{1}$$

¹⁵⁰ From the SE, the 99% confidence interval (CI) can be calculated as:

$$CI = 2.58 \cdot SE \tag{2}$$

Grid cells are classified as insignificant if the CI is larger than 0.5 °C (corresponding to

¹⁵² a T^{*} confined to about 1 bin during bootstrapping) and are excluded from the analy-

 $_{153}$ sis. 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.

157 2.3 Sea ice data

The sea ice concentration data is from the Institute of Environmental Physics (IUP). 158 University of Bremen, based on the ARTIST Sea Ice (ASI) algorithm (Spreen et al., 2008). 159 The ASI retrieval is applied to microwave radiometer data of the AMSR-E (Advanced 160 Microwave Scanning Radiometer for EOS) on the Aqua satellite and AMSR2 (Advanced 161 Microwave Scanning Radiometer 2) on GCOM-W1 sensors, which were reprocessed in 162 2018 for both platforms with the same parameters. The sea ice edge as visible in Fig. 2 163 is calculated using the following steps: (1) Retrieval of the dates for the Arctic/Antarctic 164 sea ice maximum/minimum for each year (Grosfeld et al., 2016) and calculation of the 165 multi-year average of the sea ice maxima/minima from these days. If the maximum/minimum 166 occurred in March/September, we used the last day of the respective season (February/August) 167 in the calculation. (2) The sea ice edge is defined where the sea ice concentration is at 168 least 15%. (3) Re-gridding of the sea ice data on a regular 0.25°x0.25° grid using bilin-169 ear interpolation. 170

171 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°x5° 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.

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

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

Table 1. Area-weighted averages of significant T* (in °C) for different regions and seasons. The masks used for averaging are displayed in Fig. S2.

In contrast, during the winter months (DJF) the T^* over the sea ice region (green 198 line in Fig. 2) drops to -24 °C, while over open ocean it slightly decreases to -16 °C (Fig. 2B, 199 compare Tab. 1). Similarly, during the winter months snow cover reduces T^{*} to -23 °C 200 over land. Thus, the largest seasonal differences in T^{*} are observed in regions covered 201 by snow and sea ice during the winter (Fig. 2C). Previous ship (Bigg, 1996; Bigg & Leck, 202 2001) and coastal (Creamean et al., 2018) measurements also observed a dependence of 203 the INP concentration on the extent of snow and sea ice coverage, with a decrease and 204 increase in INP concentration during the Fall freeze up and Spring thaw, respectively. 205 A reduction in wintertime INP concentration was also observed at an inland Arctic lo-206 cation in Alaska (Borys, 1983) and a Boreal Forest in Finland (Schneider et al., 2021). 207 Similarly, Wex et al. (2019) observed an increase in INPs during the snow-free summer 208 months and a decrease during the winter months at four different measurement locations 209 in the Arctic. Their back trajectory analysis showed that the highest INP concentrations 210 were associated with air mass interaction with snow-free terrain and open water, while 211 the lowest concentrations came from the sea ice and snow. 212

Airborne Arctic INP measurements (Borys, 1989; Hartmann et al., 2020) also ob-213 served a decrease in INP concentration over sea ice and snow cover. The only exception 214 was over open leads in the sea ice (Rogers et al., 2001; Hartmann et al., 2020; Curry et 215 al., 2000), which further indicates that sea ice inhibits the emissions of INPs. This lack 216 of available biological INPs has also been used to explain the lower temperatures required 217 to observe MPCs in the Arctic relative to the midlatitudes and tropics (Costa et al., 2017). 218 Furthermore, Griesche et al. (2021) observed a decrease in the frequency of ice contain-219 ing clouds when they were decoupled from the ocean surface, also indicating that ma-220 rine INPs are essential for ice formation in Arctic clouds. 221

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-226 F, Tab. 1), it is apparent that T^* is -15 °C during the Austral summer (DJF) and ho-227 mogeneous over the entire region. This is consistent with summertime ship measurements 228 conducted in the Southern Ocean, where INPs were typically observed at temperatures 229 above -14 °C (Welti et al., 2020; C. McCluskey et al., 2018) and their concentration only 230 varied by about one order of magnitude at -15 °C (Welti et al., 2020). When comparing 231 to the Arctic, the summertime T^{*} in the Southern Ocean is about 2 °C cooler. This may 232 in part be due to higher biological activity in the Arctic Ocean (McCluskey et al., 2019; 233 Irish et al., 2017) and a larger land area where glacial out wash can be emitted and act 234 as a local episodic INP source (Sanchez-Marroquin et al., 2020; Tobo et al., 2019; Ri-235 naldi et al., 2021). Meanwhile, during the winter (JJA), a similar relationship between 236

sea ice coverage and T^{*} emerges (Fig. 2E). The T^{*} over the sea ice region drops to -27 °C, 237 while over open ocean regions the T^* slightly decreases to -17 °C. This indicates that the 238 sea ice acts to inhibit the emission of INPs and directly impacts cloud phase over the 239 Southern Ocean as well. It is well known that the ocean is an important source of INPs 240 in the Southern Ocean (Schnell, 1977; Burrows et al., 2013), which is far from the Earth's 241 deserts (DeMott et al., 2016; C. McCluskey et al., 2018). Indeed, INP observations from 242 South Pole were significantly lower than at a coastal site (Belosi et al., 2014) and increased 243 when airmass back trajectories originated from the coast (Ardon-Dryer et al., 2011). Mod-244 elling studies have shown that replacing dust-based with marine-based INP parametriza-245 tions greatly improves the representation of clouds over the Southern Ocean (Vergara-246 Temprado et al., 2018; Frey & Kay, 2018). This further provides evidence that the South-247 ern Ocean is the primary source of INPs over this region and when it is covered with sea 248 ice, fewer INPs are emitted. Therefore, our results indicate that the decrease in T^{*} over 249 the sea ice is a result of the sea ice acting as a lid that inhibits the emission of INPs from 250 the Southern Ocean and, in turn, hinders the initiation of MPCs in this region. 251

Previous remote sensing observations of cloud phase over the Southern Ocean have 252 also observed a spatial pattern in the occurrence of MPCs (e.g., Mace et al., 2020, 2021) 253 with a maximum in the vicinity of the so-called Antarctic Polar Front (APF, Freeman 254 & Lovenduski, 2016). Mace et al. (2021) attributed this relationship to potentially en-255 hanced vertical updrafts in convective clouds over the APF due to warmer sea surface 256 temperatures, which would loft ice crystals from lower layers of clouds to their top where 257 a lidar-depolarization based cloud classification algorithm (Mace et al., 2020) would clas-258 sify them as mixed-phase. Additionally, they highlight that these enhanced updrafts would 259 act as a production zone for larger cloud droplets, which have been shown to be more 260 efficient for SIP (Lauber et al., 2018; Keinert et al., 2020). Although we cannot rule out 261 the importance of SIP on the classification of a cloud as LTMPC, through the combined 262 use of lidar and radar observations in the 2B-CLDCLASS-LIDAR product we can re-263 duce the importance of ice crystals being lofted to cloud top for the classification of LTM-264 PCs. Regardless of the importance of SIP in producing ice in LTMPCs, primary ice formed 265 on INPs is still required and thus, T^{*} is representative of when INPs are responsible for 266 controlling the cloud phase over the SO. Furthermore, when comparing T^{*} values with 267 the location of the APF in the summertime (see Fig. 3 in Freeman & Lovenduski, 2016) 268 there is no clear dependence, indicating that the observed seasonal variability in T^* is 269 associated with the sea ice extent and its ability to inhibit INP emissions. 270

With this in mind, it is important to note that there are well-documented differ-271 ences in high-latitude clouds over the open ocean and sea ice due to differences in sur-272 face fluxes (e.g. heat and moisture) and thermodynamic structure (Palm et al., 2010; Eirund 273 et al., 2019; Sotiropoulou et al., 2016; Young et al., 2017). However, to our knowledge 274 these differences would not lead to LO clouds occurring more frequently at colder tem-275 peratures over sea ice than over open water as we observe. Furthermore, when compar-276 ing the cloud top heights and occurrence of LO and LTMPCs over the Southern Ocean 277 (see Fig. S3), we find that they overlap and occur at the same heights in the troposphere, 278 regardless of whether they form over open ocean or sea ice. Therefore, the observed vari-279 ability in T^{*} can only be explained by the variability in the efficiency and concentration 280 of INPs present during the onset of ice formation and MPC initiation. 281

²⁸² 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 impor tance 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 298 the cloud-phase feedback (Murray et al., 2021; Prenni et al., 2007), which has so far been 299 projected to limit warming over the Southern Ocean (Forster et al., 2021; Zelinka et al., 300 2020; Bjordal et al., 2020; Tan et al., 2016). The warming-induced INP increase could 301 weaken, or even reverse, the projected increase in LO clouds with warming. This would 302 have major implications for both the magnitude and sign of the Southern Ocean cloud 303 feedback, thus shaping the climate evolution of the region itself and ultimately, the fu-304 ture climate of the entire planet. Figure 3 shows the T^{*} averaged by latitude over the 305 Southern Ocean. The stark contrast in T^{*} over sea ice and open ocean indicates that 306 in a warming world with sea ice retreat, T^{*} over formerly ice covered regions will increase 307

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.

³¹⁴ 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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Supporting Information for "Spaceborne evidence that ice-nucleating particles influence cloud phase"

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Figure S1. 99% confidence interval (CI) of T* based on bootstrapping (N = 100) for the Arctic (Panel A: summer JJA, Panel B: winter DJF) and Antarctica (Panel C: summer DJF, Panel D: winter JJA). Grid cells with an insignificant T* value are hatched in blue (CI ≥ 0.5 °C). Grid cells with no data are marked with dots.



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Figure S2. Masks for the area-weighted averaging of significant T* values for different regions and seasons. These masks are used in the calculation of the values in Table S1. For Arctic: (A) summer (JJA) ocean, (B) summer (JJA) land, (C) winter (DJF) sea ice, (D) winter (DJF) open ocean, (E) winter (DJF) land. For Antarctica: (F) summer (DJF) ocean, (G) winter (JJA) sea ice, (H) winter (JJA) open ocean.

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Figure S3. Frequency of occurrence of LO (blue) and LTMPCs (red) as a function of cloud top height summed over 5°-latitudinal bands with a midpoint at 50°S, 55°S, 60°S, 65°S, and 70°S for Austral Summer (DJF, panels: A, C, E, G, I) and winter (JJA, panels: B, D, F, H, J) normalized to all measured single-layer clouds in the given season.