Strong ocean/sea-ice contrasts observed in satellite-derived ice crystal number concentrations in Arctic boundary-layer clouds

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Abstract

The Arctic climate changes at a faster rate than the rest of the globe. Boundary-layer clouds may play an important role to this change. At temperatures below 0°C, mixed-phase clouds exist and their phase and longevity is influenced by the abundance of ice crystals, which in turn is a function of aerosols serving as ice nucleating particles (INPs). Previous in-situ studies suggested a local source of INPs due to biological activity over open ocean. Here we investigate ice crystal concentrations in clouds below 2km at a large scale, by exploiting a newly-developed dataset - DARDAR-Nice - retrieved from active satellite remote sensing. The dataset spans from 2006-2016. Contrary to previous expectation, we find that at a given latitude and temperature, there are more ice crystals over sea ice than over open ocean. This enhancement is particularly found in clouds south of 70°N, but also at temperatures between 0 and -10°C.

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Key Points:

- New ice microphysics from active satellite instruments enable large-scale analysis of Arctic boundary-layer clouds.
- Ice crystal numbers are enhanced over sea ice compared to open ocean at temperatures above -10°C.
- This difference is most pronounced in clouds south of 70°N through all the temperature range.

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Abstract

The Arctic climate changes at a faster rate than the rest of the globe. Boundary-layer clouds may play an important role to this change. At temperatures below 0°C, mixed-phase clouds exist and their phase and longevity is influenced by the abundance of ice crystals, which in turn is a function of aerosols serving as ice nucleating particles (INPs). Previous in-situ studies suggested a local source of INPs due to biological activity over open ocean. Here we investigate ice crystal concentrations in clouds below 2km at a large scale, by exploiting a newly-developed dataset - DARDAR-Nice - retrieved from active satellite remote sensing. The dataset spans from 2006-2016. Contrary to previous expectation, we find that at a given latitude and temperature, there are more ice crystals over sea ice than over open ocean. This enhancement is particularly found in clouds south of 70°N, but also at temperatures between 0 and -10°C.

Plain Language Summary

The Arctic region is particularly affected by climate change, since its warming is 2-3 times larger than global average during recent decades. One of the contributors to this "Arctic Amplification" may be the Arctic clouds and in particular the mixed phase type, where ice and supercooled liquid coexist at temperatures lower than 0°C. Aerosols play also a significant role in cloud formation, since without the presence of some effective particles, the ice crystals could not form at all in temperatures between 0 and roughly -40°C. In this study, we use a new satellite dataset which provides an important cloud quantity, the amount of ice crystals in the clouds. Although this dataset is limited to pure ice clouds, it can prove useful for understanding the behavior of Arctic clouds. What we find here is that Arctic low-level clouds show larger quantities of ice crystals over sea ice than over ocean and we think that this can be attributed to the amount and type of aerosols related to each surface. This finding contradicts a previous hypothesis, which stated that more ice crystals would possibly form over ocean because of the presence of highly ice effective aerosols there.

1 Introduction

Clouds constitute an important component of the climate system, since they influence both the radiative budget and the atmospheric water balance (Stephens, 2005), i.e. the energy and water cycles. Cloud formation is inherently connected to the presence of aerosols. In particular, cloud droplets as well as ice crystals form on aerosols that serve as cloud condensation nuclei (CCN) and ice nucleating particles (INPs), respectively. As a consequence, anthropogenic aerosol emissions exert an effective climate forcing due to aerosol-cloud interactions, contributing one of the largest uncertainties in our knowledge of anthropogenic climate change (Boucher et al., 2013; Szopa et al., 2021; Forster et al., 2021).

In the present study, we are especially interested in clouds forming in the Arctic region and their relation to INPs as potentially emitted from surface sources. Arctic clouds are involved in a complex interplay of processes and feedbacks and may indirectly play a role in the enhanced Arctic warming (Pithan & Mauritsen, 2014). This warming, about 2-3 times stronger than the rest of the world, is commonly referred to as Arctic Amplification (Serreze & Barry, 2011; Wendisch et al., 2017). Although much effort has already been invested during the previous decades, aiming to disentangle the causes of this phenomenon, its main contributions are not yet fully understood. As a result, more studies towards this direction are needed, to fill the knowledge gaps in our perspective of the complex Arctic climate system. Here we consider the idea that Arctic boundary layer clouds may change their radiative impact over time due to the influence of aerosol. Specifically, there are hypotheses that local sources of INPs in the Arctic may change due to

the global increase in atmospheric temperature and the corollary sea ice retreat, which will lead to increased aerosol emissions from the exposed ocean surfaces (Browse et al., 2014; Gilgen et al., 2018).

Several studies in the past have investigated ice clouds and the factors determining ice crystal formation. At very cold temperatures, below −38°C, ice crystals are formed homogeneously by the spontaneous freezing of haze particles or supercooled droplets. A prerequisite for homogeneous ice nucleation is sufficient supersaturation, which is generated by updrafts and decreasing temperature (Heymsfield et al., 2017). At warmer temperatures however, above −38°C, the ice crystal nucleation is also possible without the need of these large amounts of supersaturation, but instead with the aid of some ice-active aerosols, the INPs, which are able to nucleate ice heterogeneously. These aerosols are very important for the formation and lifetime of ice-containing clouds in the regime where without INPs present, supercooled liquid clouds would exist. Such clouds are prevalent in the Arctic boundary layer (Shupe et al., 2006; de Boer et al., 2009; Shupe, 2011; Morrison et al., 2012).

Depending on the temperature, different types of INPs are able to nucleate ice. According to Hoose and Möhler (2012), mineral dust is an important source of ice nuclei at lower temperatures, while some bioaerosols are highly effective in forming ice at relatively warm temperatures (roughly above −10°C). Biological INPs can be for example bacteria, fungi, pollen, lichen, viruses, phytoplankton or diatoms (Kanji et al., 2017), but also thawing permafrost has been recently found to be a potential source of biological INPs (Creamean et al., 2020). Moreover, it has been suggested by previous studies that a marine source of INPs can be associated with sea spray (DeMott et al., 2016) and is able to determine the ice nuclei concentrations in remote environments (Burrows et al., 2013; Wilson et al., 2015). In addition, McCluskey et al. (2017, 2018) linked sea spray aerosols in remote oceanic environments to organic material and suggested biological aerosols as important contributors to INP populations in these regions.

In the Arctic several measurement campaigns have taken place that included measurements of INPs. A source of highly ice-active INPs in Arctic water surfaces has been previously reported (Wilson et al., 2015; Irish et al., 2017, 2019). Creamean et al. (2018) performed measurements during spring 2017 in an Arctic oilfield location and found high concentrations of INPs, efficient at warm temperatures. Wex et al. (2019) using ground-based filter samples, also observed INPs which can nucleate ice at temperatures as warm as −5°C during the Arctic summer. The authors hinted at a potentially biogenic nature of INPs, although no explicit source was identified in this study. Hartmann et al. (2020) using airborne samples, found INPs with similar onset freezing temperatures, which originated from the Arctic winter marine boundary layer. Following on, Hartmann et al. (2021) found a correlation between Arctic INP populations in the sea surface microlayer and the air, suggesting a local biogenic marine source as the likely origin.

Some of the aforementioned studies suggest the presence of a potential marine source of particularly effective aerosols in the Arctic, possibly originating from the Arctic ocean. Nevertheless, other studies support that aspects of the sea ice surface are also capable of containing such particles. Irish et al. (2017) studied INPs in the sea surface microlayer and bulk seawater and found a negative correlation between salinity and INP presence, suggesting a possible relation to sea ice melting. According to Zeppenfeld et al. (2019) the sea surface microlayer of the marginal ice zone and that of melt ponds is indicative of high ice nucleating activity. In addition, other microorganisms such as ice algal aggregates or sea ice diatoms, which can accumulate in and below the sea ice and melt ponds and float into the water during the melting season (Assmy et al., 2013; Fernández-Méndez et al., 2014; Boetius et al., 2015) may also play an important role as INPs. As a result, even though there are clues on the marine origin of INPs, it is not yet quite clear whether they originate entirely from the ocean or they are also connected to sea ice melting processes.

Although the presence of INPs is critical to ice formation, it is not the only reason determining the ice crystal number in clouds. The atmospheric conditions play also an important role in transfering the particles into the cloud layer. There are several studies in the Arctic dealing with coupling conditions of clouds (Sotiropoulou et al., 2014; Gierens et al., 2020; Griesche et al., 2021) and turbulence (Egerer et al., 2021). Gierens et al. (2020) studied Arctic mixed-phase clouds and concluded that surface coupling is an important factor for their persistence and properties. Griesche et al. (2021) investigated Arctic clouds with regard to coupling conditions and found that more ice particles are detected in coupled clouds at warm temperatures. Thus, it seems that coupling and turbulence may have a large impact on the ice crystal nucleation and for this reason we decided to investigate also this aspect during this study.

A main question arising is whether such results from campaigns are valid at a large scale, i.e., whether one can find a widespread impact on the microphysical structure of Arctic clouds. Such a large-scale analysis became now possible thanks to a new dataset of satellite-retrieved ice crystal number concentrations (N_i) (Section 2.1; Sourdeval et al., 2018a), a key measure to link ice clouds to their aerosol environment. In this study, we are exploring the N_i in Arctic ice clouds over open ocean in comparison to sea ice, for the time-period 2006-2016. We are focusing on the temperature range where INPs are important (heterogeneous nucleation regime; 0 until −38°C) and at the lower part of the troposhere, in order to emphasize the relation to the surface aerosols. What is more, we are examining the atmospheric conditions that favor the transfer of surface aerosols to the base of the clouds to reduce uncertainty. Our goal is to provide some insight on the large-scale picture of boundary-layer ice clouds in the Arctic and the possible connection to the aerosols there.

2 Data

2.1 DARDAR-Nice

DARDAR-Nice is a dataset retrieving ice crystal number concentrations (N_i) and was based on the radar/lidar (DARDAR) algorithm developed by Delanoë and Hogan (2010). It combines lidar and radar information to extract the particle size distributions of ice crystals. Based on DARDAR, Sourdeval et al. (2018a) created a number concentrations retrieval product. Lidar measurements originate from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP; Winker et al., 2003) onboard CALIPSO satellite, while radar measurements come from the Cloud Profiling Radar (CPR; Stephens et al., 2018) onboard CloudSat. The data are provided with a 1.7 km footprint and a vertical resolution of 60 m. This product has been thoroughly evaluated against recent in-situ measurements in Sourdeval et al. (2018a) and Krämer et al. (2020) and was further used to investigate the controls on the N_i by Gryspeerdt et al. (2018).

For the purpose of this study we used Level-2 (L2) data, starting from June 2006 until the end of 2016. The data provide ice crystal number concentration profiles along the satellite's track, but also thermodynamic variables, e.g. temperature and pressure. These profiles originate from the ECMWF-AUX dataset, which contains EMCWF state variables interpolated into each CPR's vertical bin (Delanoë & Hogan, 2010). The data concern either pure ice clouds or mixed-phase, where ice is mixed with or below supercooled liquid water droplets. It should be noted that mixed-phase conditions, occuring mostly between 0 and -40°C, are considered to be less trustworthy and should be handled with great attention when comes to interpretation (Sourdeval et al., 2018a), because the lidar signal is quickly extinguished by liquid droplets and microphysical assumptions in the algorithm do not properly treat such mixtures. DARDAR-Nice reports ice crystal numbers by integrating the size distribution starting from two different lower cut-off sizes, distinguishing those that are larger than 5 µm and 100 µm (here denoted as $\mathrm{N}_{\mathrm{i},5\mu\mathrm{m}}$ and $\mathrm{N}_{\mathrm{i},100\mu\mathrm{m}}$), respectively. A larger cut-off size leads to a more reliable retrieval, but

fails to account for a large fraction of ice crystals in an air volume (Sourdeval et al., 2018a). In order to relate the number concentrations of ice crystals to the underlying surface, another satellite dataset which retrieves sea ice concentration was necessary, which will be further discussed in the next section.

2.2 AMSR-E/AMSR2 Sea Ice Concentration

The Arctic region comprises, beyond land and open ocean, a large portion of sea-ice covered surface. The sea ice itself is a variable quantity, being subject to seasonal and inter-annual changes, and depending on its thickness and position it can impact the Arctic radiative balance and Arctic environment, e.g. the water vapor and clouds (Wendisch et al., 2017). Here we make use of the product developed by the Institute of Environmental Physics (IUP) of the University of Bremen, which retrieves sea ice concentration on a daily basis for the Arctic region (Melsheimer & Spreen, 2019, 2020).

This dataset was based on the application of the ARTIST Sea Ice Algorithm (ASI; Spreen et al., 2008) on microwave radiometer data. It has a resolution of 6.25 km on a north polar stereographic grid and its time period overlaps with that of DARDAR-Nice. The data were obtained from two instruments; the AMSR-E (Advanced Microwave Scanning Radiometer for EOS) and its successor AMSR2. According to Melsheimer and Spreen (2019, 2020), despite the change of the measuring instrument in 2011-2012, all data have been processed in the exact same settings.

3 Method

3.1 Constraint of Clouds

In our analysis we are interested in ice clouds with tops below 2 km in order to account for low-level boundary layer clouds. When referring to ice clouds here, we mean ice crystals that were retrieved inside cloud layers, identified as consisting purely of ice by the DARDAR algorithm. The definition of an ice layer we use is the same as in DARDAR-Nice, namely a certain profile of number concentrations that is vertically separated by another one by at least 480 m. Ice coexisting with supercooled water or lying below it (denoted as mixed-phase in DARDAR-Nice) was excluded from our analysis, due to high uncertainty of those retrievals (Sourdeval et al., 2018a). In addition, a limitation exists at the lower part of the atmosphere due to the CloudSat radar's blind zone between 1.2 km and the ground surface (Maahn et al., 2014). As a consequence, we use retrievals that are only constrained by the lidar instrument below this height.

The focus of this study is on the ice phase of clouds and in particular on the heterogeneous nucleation regime, where the ice formation requires the presence of INPs in the atmosphere to form onto (above -38°C). For this reason, we classified the ice crystal numbers into four distinct subfreezing temperature classes of 10°C each. In this temperature range however, the N_{i} retrievals have to be carefully interpreted, due to the assumption of a monomodal particle size distribution in small ice crystals (Sourdeval et al., 2018a). In order to avoid sedimentation of ice crystals from higher levels of the cloud or even another higher cloud, our analysis contains only single-layer clouds. In this way, we aim to limit ice formation processes such as secondary ice production (Yano & Phillips, 2011; Field et al., 2017; Korolev & Leisner, 2020) or seeder-feeder processes. Moreover, following Gryspeerdt et al. (2018) we defined the cloud top as the upper 120 m of a certain profile.

3.2 Coupling/Decoupling

If there is an aerosol emission source at the surface, it is not certain to which degree some of the aerosols will eventually reach the cloud to nucleate ice. Atmospheric

conditions, and especially the stability of the atmospheric layer below the cloud, play a large role on the transfer of aerosols to the cloud base. A useful measure to determine the stability conditions is the potential temperature θ (Sotiropoulou et al., 2014; Gierens et al., 2020; Griesche et al., 2021). For its derivation we used the temperature and pressure information from the ECMWF-AUX data, included in the DARDAR-Nice dataset. For each profile we determined a stability index, which in turn was used to categorize clouds in coupled and decoupled cases. The index was calculated using a rather simple approach, as the θ-difference between the cloud base and the ground surface (Goren et al., 2018). Where this difference was almost zero (less than 0.5 K), then turbulence and vertical mixing are present in the boundary layer, leading to coupling between the cloud and the surface. On the other hand, where the difference was positive (greater than 0.5 K), the boundary layer is characterized by stability, the vertical motions are supressed and the cloud is decoupled from the surface. The 0.5 K limit was used to account for interpolation errors in the thermodynamic variables.

3.3 Analysis of the N_i

Different kinds of aerosols are emitted into the atmosphere, depending on the surface type (e.g. land, ocean). For this reason, we distinguished the ice crystal numbers with respect to the underlying surface. Two main surface types are discussed here; sea ice and open ocean. By sea ice we mean the area where sea ice concentration exceeds 80%, a limit which corresponds to the definition by the World Meteorological Organization as "close ice" (JCOMM Expert Team on Sea Ice, 2014), while open ocean refers to the ice-free area. Since the datasets used here were neither in the same spatial nor temporal resolution, it was necessary to perform spatial interpolation and time-averaging of N_i, to correspond with the AMSRE/2 resolutions.

The daily ice crystal concentrations related to each surface, were further analyzed in terms of seasonality and regionality. The Arctic was divided into five equal parallel zones (per 5°) north of 60°N and the time-period was sliced into two main seasons; cold (boreal winter, autumn) and warm (boreal spring, summer). It is important to note here, that the geographical area covered by each zone decreases with latitude. In addition, the region from 80° N northward only covers the latitudes up to around 82° N, due to the maximum possible latitude the satellite can reach in the polar regions.

From the daily N_i distributions, we determined the medians and their 95% confidence intervals. The confidence intervals were calculated using a nonparametric bootstrapping technique by using random resampling with replacement. Since we aim at a comparison of the mean behavior of the ice crystals over sea ice and ocean, in the following sections we present and discuss only the medians along with their confidence intervals, rather than the full N_i distributions.

4 Results

The results of the analysis are shown in Fig. 1. As documented earlier, albeit with a focus on high-altitude clouds (Sourdeval et al., 2018a), N_i is increasing as temperature decreases. The absolute numbers vary from $\mathcal{O}(1\mathrm{L}^{-1})$ above temperatures of −10°C to $\mathcal{O}(10\mathrm{L}^{-1})$. The concentrations within given temperature classes do not change very much with latitude.

Fig. 1 depicts ice crystal numbers over sea ice and open ocean. A larger concentration of ice crystals ($N_{\mathrm{i},5\mu\mathrm{m}}$) over sea ice is observed in the lower latitudes of the Arctic (60-70°N), forming a positive difference between sea ice and ocean in both seasons (cold & warm). This difference tends to increase with decreasing temperature during the warm season. A higher concentration over sea ice is not always observed at higher latitudes (north of 70°N). In warm temperatures (0 to −10°C) larger $N_{\mathrm{i},5\mu\mathrm{m}}$ still persist over

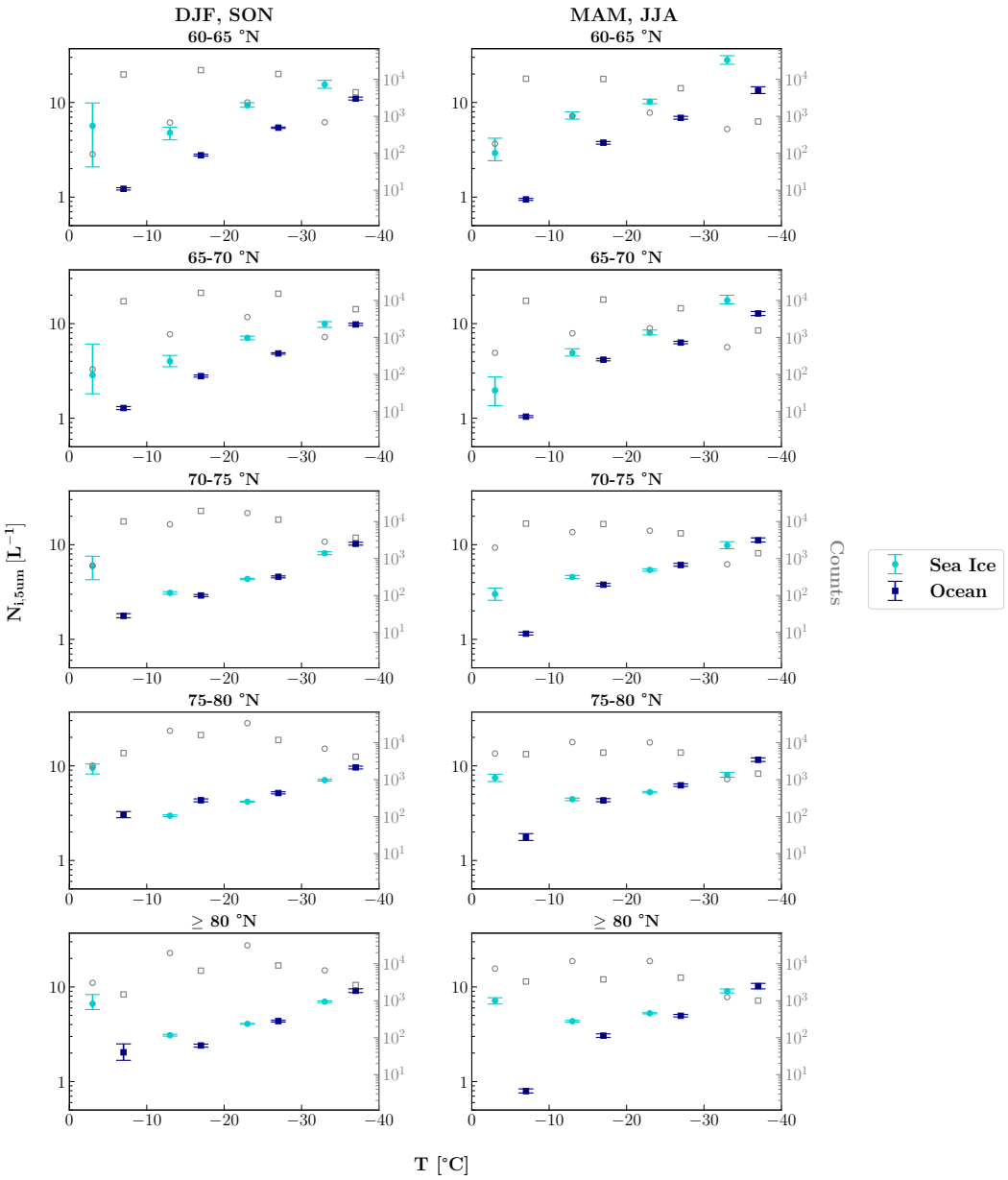

Figure 1. Medians of distributions of daily number concentrations of ice crystals that are larger than 5µm (in L^{-1}; left y-axis) as a function of temperature for the time period 2006-2016. Different colors indicate the surfaces over which these clouds exist; sea ice (in turquoise) and open ocean (in blue). Two different seasons are presented (cold: DJF, SON / warm: MAM, JJA) and five geographical regions (latitude belts). The 95% confidence intervals are displayed as the error bars. The numbers of samples used to calculate the medians are shown in grey (right y-axis).

sea ice, but these differences become smaller or even close to zero as the temperature drops and even sometimes a positive difference over ocean is formed.

Clouds separated based on coupling conditions are given in the Supporting Information. Coupled clouds (see Fig. S1) show a similar pattern through all the seasons and

regions. Decoupled clouds follow more or less the general tendency, with the exception of showing increased number concentrations over sea ice compared to open ocean at warmer temperatures, usually up to -20°C (see Fig. S2).

Larger ice crystals, with a minimum size of 100µm ($N_{\mathrm{i},100\mu\mathrm{m}}$), have a similar behavior with what presented in Fig. 1 (not shown). However, the concentrations of large ice crystals are - as expected - much lower. Furthermore, DARDAR-Nice detected also some cases of ice with some supercooled water existing lower into the same cloud layer. By incorporating those tops into the analysis - without using the actual mixed-phase layers - the main outcome didn't change very much (not shown). However, a distinction from what was presented above can be spotted at warmer temperatures (above -10°C), where the differences between N_{i} over sea ice and ocean are quite smaller.

5 Discussion and Conclusion

Ice nucleating particles, being the seed of the ice phase between 0 and roughly -40°C, is one of the factors controlling the ice crystal number in clouds. INP numbers are relatively low, albeit with a strong variability (10^{-6} to $10^{-1}\mathrm{L}^{-1}$) at warm temperatures (above -10°C), while increasing between -10 and -25°C (10^{-5} to $10^{2}\mathrm{L}^{-1}$) and especially below -25°C (10 to $10^{3}\mathrm{L}^{-1}$) (Petters & Wright, 2015). Wex et al. (2019), Hartmann et al. (2020) and Hartmann et al. (2021) report similar or lower numbers from measurements in the Arctic.

The DARDAR-Nice ice crystal concentrations analysed here are of the order of $1\,\mathrm{L}^{-1}$ at temperatures between 0°C and -10°C , at or above the upper top of the range by in situ measurements for INPs. In contrast, the number concentrations are comparable between -20 and -25°C. In colder temperatures (below -30°C) the values for N_{i} retrieved by DARDAR-Nice are even lower than the observed INPs.

It is also interesting that our analysis shows a tendency of more ice crystals over the sea ice than over the ocean, contradicting our previous expectation. This is seen quite consistently at warmer temperatures, independently of the season, region and coupling conditions, but it is particularly visible in the low latitudes of the Arctic. These regions concern mostly coastal areas, close to the sea ice edge and land, where the sea ice varies greatly throughout the year and the INP sources can be diverse. Towards northern latitudes, long-range transport of aerosols could also play a role where the concentrations are similar, but this still cannot explain why the concentrations over sea ice appear higher at the warmer temperature range.

There are three main reasons we could suspect as probable causes for this difference:

1. local source of INPs over sea ice
2. blowing of snow
3. secondary ice production

A local source of INPs could be related to the sea ice melting and refreezing processes. Melt ponds developing on sea ice can be responsible for the release of new particles into the atmosphere (Dall´Osto et al., 2017), with potentially high ice nucleating ability (Zeppenfeld et al., 2019). Such particles could contain biological/organic material that is considered as highly efficient INP. Frost flowers growing ontop of young sea ice may also result in the production of aerosols. However, since frost flowers are highly saline structures, the aerosols emitted are mostly sea salt aerosols (Rankin et al., 2000; Rankin & Wolff, 2003; Xu et al., 2016; Hara et al., 2017), that are not efficient INPs. Blowing snow from the ground has also been found to be responsible for seeding cold clouds (Vali et al., 2012; Geerts et al., 2011, 2015). At snow covered surfaces, such as the high

latitudes, under strong wind and updraft conditions, ice fractures may be able to reach low-level clouds and facilitate the nucleation process (Yang & Yau, 2008). Nevertheless, blowing snow is usually effective up to 1 m from the ground (Schmidt, 1982) and most of these studies consider mountainous regions and orographic clouds, where this mechanism is more crucial.

Secondary ice production over sea ice, e.g. ice break up due to collisions, could also play a role, in particular in low updraft variability and relatively warm temperature conditions (Sotiropoulou et al., 2020). Our analysis shows that when mixed-phase processes are present, the differences between ice crystal concentrations over sea ice and ocean are dampened at warm temperatures, which might mean that the secondary ice production is not so important in determining this difference. However, the data analysis alone does not allow to attribute the identified differences between sea ice and ocean presented above, calling for model studies that are planned for the future.

Even though these results give a first impression, from a large-scale perspective, of the ice crystal formation over different surfaces in the Arctic, there is still some uncertainty related to the cloud type studied here. This analysis is limited to boundary-layer ice clouds. The prevalent cloud type in the Arctic boundary layer are mixed-phase clouds with a supercooled liquid layer on top (Shupe & Intrieri, 2004; Shupe et al., 2006; Morrison et al., 2012). However, for such clouds N_i unfortunately cannot be studied even from the new active satellite remote sensing product. The clouds consisting purely of ice are still instructive for the problem studied here and are possibly at the decaying stage of their formation. Future observational and modelling studies are thus needed to corroborate the results presented here. However, our results suggest that in a warming world with retreating sea ice, boundary-layer clouds in the mixed-phase temperature regime will contain less ice crystals, with implications for local climate change.

Open Research

The DARDAR-Nice data used for the study of ice crystal number concentrations are available from the AERIS ICARE data center via `https://doi.org/10.25326/09` and can be provided upon request (Sourdeval et al., 2018b). The AMSR-E and AMSR2 ASI Sea Ice Concentration datasets used here to dinstinguish the Arctic surface conditions are publicly available through PANGAEA - Data Publisher for Earth & Environmental Science via `https://doi.org/10.1594/PANGAEA.919777` and `https://doi.pangaea.de/10.1594/PANGAEA.898399` respectively (Melsheimer & Spreen, 2019, 2020).

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Supporting Information for "Strong ocean/sea-ice contrasts observed in satellite-derived ice crystal number concentrations in Arctic boundary-layer clouds"

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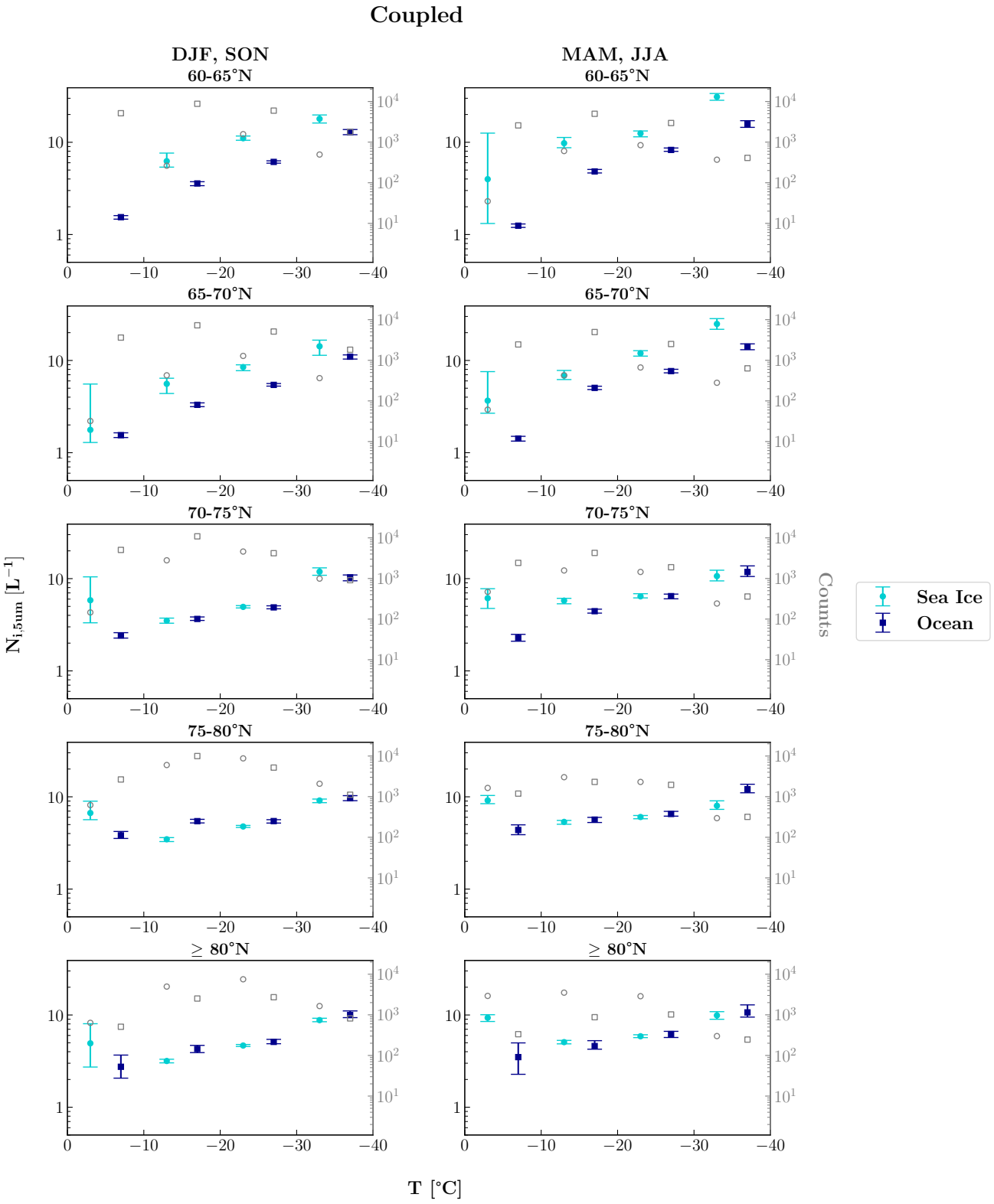

Figure S1. As in Figure 1, but for coupled clouds.

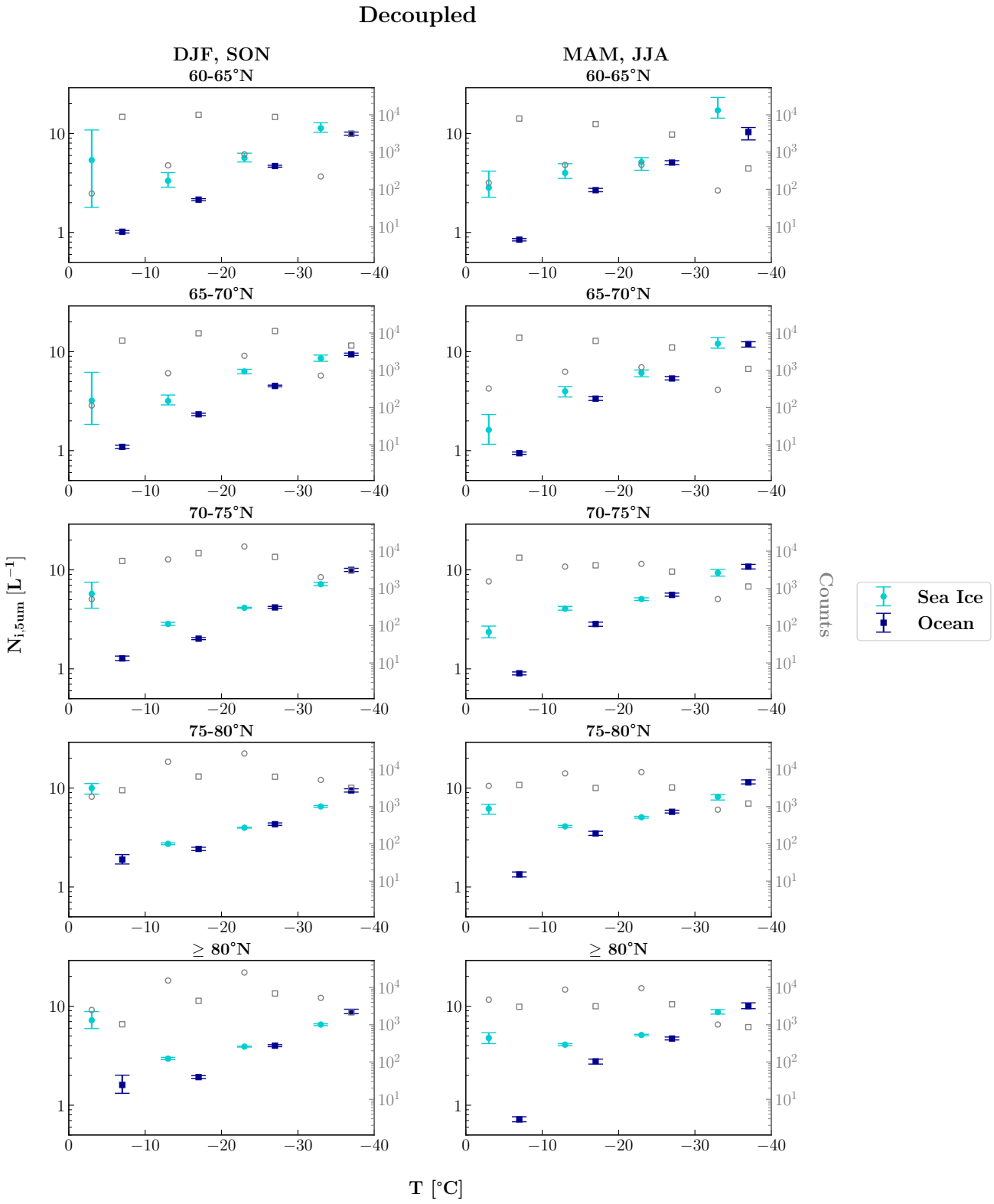

Figure S1. As in Figure 1, but for decoupled clouds.