Highly active ice-nucleating particles at the summer North Pole

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Abstract

The amount of ice versus supercooled water in clouds defines their radiative properties and role in climate feedbacks. Hence, knowledge of the concentration of ice-nucleating particles (INPs) is needed. Generally, the concentrations of INP is found to be very low in remote marine locations allowing clouds to persist in a supercooled state. However, little is known about the INP population in clouds at and around the summertime North Pole. We had expected that concentrations of INPs at the North Pole would have been very low given the distance from open ocean and terrestrial sources coupled with effective wet scavenging processes. Here we show that during summer 2018 (August and September) high concentrations of biological INPs (active at >-20°C) were present at the North Pole. In fact, INP concentrations were sometimes as high as those recorded in mid-latitude locations strongly impacted by highly active biological INPs, in strong contrast to the Southern Ocean. Furthermore, using a balloon borne sampler we demonstrated that INP concentrations were often different at the surface versus higher in the boundary layer where clouds form. Back trajectory analysis suggests that there were strong sources of INPs near the Russian coast, possibly associated with wind-driven sea spray production, whereas the pack ice, open leads, and the marginal ice zone were not sources of highly active INPs. These findings suggest that primary ice production, and therefore Arctic climate, is sensitive to transport from locations such as the Russian coast that are already experiencing marked climate change.

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Key Points:

- The concentration of ice-nucleating particles at the North Pole in summer 2018 was amongst the highest anywhere in the world.
- These biological ice-nucleating particles were derived from the Russian seas and perhaps associated with wind-driven sea spray.
- The concentration of ice-nucleating particles at the surface was often different to that higher in the boundary layer where clouds form.

Abstract

The amount of ice versus supercooled water in clouds defines their radiative properties and role in climate feedbacks. Hence, knowledge of the concentration of ice-nucleating particles (INPs) is needed. Generally, the concentrations of INP is found to be very low in remote marine locations allowing clouds to persist in a supercooled state. However, little is known about the INP population in clouds at and around the summertime North Pole. We had expected that concentrations of INPs at the North Pole would have been very low given the distance from open ocean and terrestrial sources coupled with effective wet scavenging processes. Here we show that during summer 2018 (August and September) high concentrations of biological INPs (active at >-20°C) were present at the North Pole. In fact, INP concentrations were sometimes as high as those recorded in mid-latitude locations strongly impacted by highly active biological INPs, in strong contrast to the Southern Ocean. Furthermore, using a balloon borne sampler we demonstrated that INP concentrations were often different at the surface versus higher in the boundary layer where clouds form. Back trajectory analysis suggests that there were strong sources of INPs near the Russian coast, possibly associated with wind-driven sea spray production, whereas the pack ice, open leads, and the marginal ice zone were not sources of highly active INPs. These findings suggest that primary ice production, and therefore Arctic climate, is sensitive to transport from locations such as the Russian coast that are already experiencing marked climate change.

Plain Language Summary

Clouds play a critical role in Earth's climate, both reflecting incoming sunlight and trapping outgoing heat energy. Hence, even small errors in the representation of clouds in climate models can lead to uncertainty in predictions of, for example, sea ice extent. In the Arctic, clouds often exist below 0°C and cloud water droplets can exist in a supercooled liquid state. In the absence of a special class of particle that can trigger ice formation in droplets, ice-nucleating particles (INPs), supercooled water droplets can cool well below -35°C before spontaneously freezing. Hence, the presence of INPs can reduce the amount of supercooled water in clouds, making them less reflective with a shorter lifetime. Based on our knowledge of INPs in other remote oceans, we expected very low INP concentrations in the central Arctic. However, we have shown that there are high concentrations of biological INPs in the summertime North Pole. Furthermore, these INPs come from the seas off the coast of Russia, a region already experiencing strong climate change. It is possible that these sources may become even more important as the Arctic becomes increasingly ice-free, causing changes in Arctic clouds and further changes in climate.

1 Introduction

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The Arctic climate is strongly influenced by ubiquitous low-level mixed-phase clouds [Kay and L'Ecuyer, 2013; Tjernstrom *et al.* , 2012; Vüllers *et al.* , 2021]. The radiative effect of these clouds is influenced by the amount of ice and supercooled water they contain, which depends on an intricate balance of dynamical and microphysical processes [Morrison *et al.* , 2012]. Realistic representation of these processes is needed to correct model biases in the amount of supercooled liquid in mixed-phase clouds and reduce uncertainty in feedbacks [Tan and Storelvmo, 2019].

A rare subset of the total aerosol particle population, ice-nucleating particles (INPs), can induce primary ice production in Arctic mixed-phase clouds when immersed in supercooled cloud droplets [Murray*et al.* , 2012]. In the summertime, Arctic marine atmospheric boundary layer temperatures are usually much warmer than those required for homogeneous freezing ([?] –35 degC) [Herbert *et al.* , 2015], hence heterogeneous nucleation on INPs determines the production of ice in clouds, at least in the absence of ice precipitating from overlying clouds [Vassel *et al.* , 2019]. Numerous INP types that can induce nucleation over a large range of temperatures have been identified [Hoose and Mohler, 2012; Kanji *et al.* , 2017; Murray*et al.* , 2012]. However, the sources and ice-nucleating properties of INPs in the Arctic, especially the central Arctic (>80deg N), are poorly defined.

INP measurements have been made around the periphery of the Arctic circle from locations close to, or on, land, but relatively few measurements have been made in the summertime central Arctic Ocean (see compilations in [Welti *et al.* , 2020] and [Murray *et al.* , 2021]. Recent research suggests that there are significant terrestrial sources of Arctic INPs including glacial dust from Svalbard [Tobo *et al.* , 2019] and Iceland [Sanchez-Marroquin *et al.* , 2020], terrestrial biological aerosol from boreal forests [Schneider *et al.* , 2021], and even particles released from thawing permafrost [Creamean *et al.* , 2020]. There is also a plethora of other high latitude dust sources that have not been investigated in terms of their ice-nucleating ability [Bullard*et al.* , 2016]. Marine biogenic INPs emitted from the sea surface through bubble bursting are also thought to contribute to the INP population of the oceanic high-latitudes [Bigg, 1996; Bigg and Leck, 2001; Hartmann *et al.* , 2020a; Hartmann *et al.* , 2021; Ickes *et al.* , 2020; Irish *et al.* , 2017; Wilson *et al.* , 2015]. Sea spray is thought to produce relatively low INP concentrations, but in the absence of other INP types it can dominate the INP population [McCluskey *et al.* , 2018a; Vergara-Temprado*et al.* , 2017].

Ground level observations at several land-based sites around the Arctic throughout the seasonal cycle showed the highest (but variable) INP concentrations during spring, summer and autumn and the lowest concentrations in winter [Wex *et al.* , 2019]. These measurements suggest that there are marine and terrestrial INP sources around the Arctic, but it is unclear how important these sources are for clouds over the summertime central Arctic Ocean. Based on back trajectory analysis of INP measurements in the central Arctic, Bigg [1996] suggested that there was an open ocean source of INPs active at –15 degC. Later, Bigg and Leck [2001] suggested the pack ice edge and bubble bursting in local leads throughout the pack ice can serve as a source of INPs. Indeed, it has been shown that there is a reservoir of INPs in the seas around the Arctic [Creamean *et al.* , 2019; Hartmann *et al.* , 2021; Irish *et al.* , 2017; Wilson *et al.* , 2015] and INP concentrations in the central Arctic decrease during the transition from Arctic summer to autumn, possibly due to the reduced availability of ice-free marine sources [Bigg and Leck, 2001].

While it is clear that there are strong sources of INPs in the lower Arctic environment ([?]80degN), it is not clear if these INPs are transported to the central Arctic. The prevailing view is that aerosol within the summertime high Arctic boundary layer experiences little effect from long-range transport [Kupiszewski *et al.* , 2013], and with few sources of primary aerosol in the central Arctic Ocean, sources such as local leads may be important [Bigg and Leck, 2001]. However, it has also been suggested that aerosol particles can be transported from lower latitudes into the central Arctic boundary layer either through boundary layer transport or entrainment from the free troposphere [Igel *et al.* , 2017; Morrison *et al.* , 2012; Schmale *et al.* , 2021].

The structure of the Arctic summertime boundary layer is complex (Figure 1). The boundary layer is typically several hundred meters to over a kilometer deep, but often consists of two distinct layers: the surface mixed layer and the cloud mixed layer. These two layers are each well mixed, but separated by a

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decoupling layer at ˜100 m to 300 m that prevents efficient transport between them [Brooks *et al.* , 2017]. Hence, measurements at the surface are not necessarily representative of those in the cloud mixed layer.

Here, we present measurements of INP concentrations close to the North Pole both in and above the surface mixed layer. The measurements were made during the Microbiology-Ocean-Cloud-Coupling in the High Arctic (MOCCHA) campaign, which took place throughout August and September 2018 on the Swedish icebreaker Oden. Measurements took place while Oden was on route to the North Pole as well as when it was moored to an ice floe in the inner pack ice and drifting passively between 88-90degN. Samples were collected for INP analysis at both ship level (in the surface mixed layer) and using a balloon-borne sampler in the cloud mixed layer. We use backward trajectories alongside other measurements to suggest that the source of the most active INPs reaching the North Pole is outside of the pack ice, and near the Arctic coast of Russia.

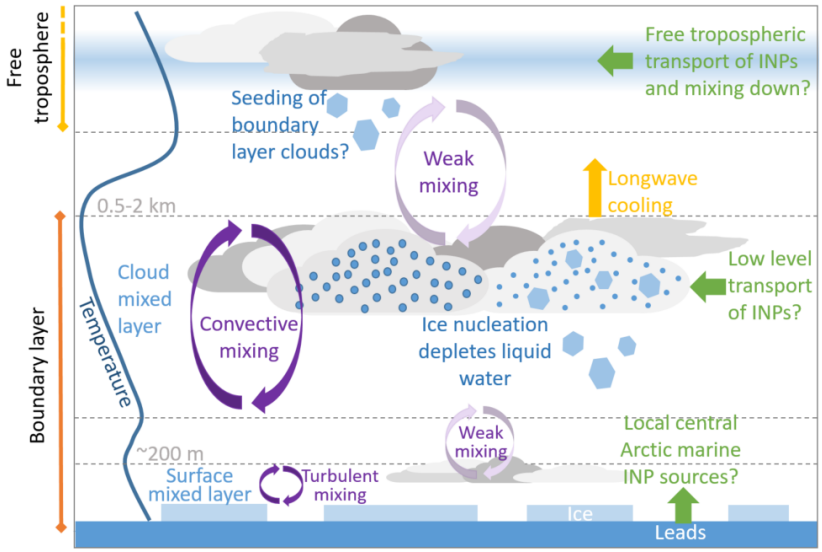

Figure 1. Central Arctic boundary layer structure and potential sources of INPs. The lowest part of the boundary layer (surface mixed layer) is often decoupled from the rest of the boundary layer [Brooks *et al.* , 2017]. In this paper we report INP measurements both in the surface mixed layer and in the cloud mixed layer and use these measurements to infer information about the dominant sources of INPs in the central Arctic boundary layer.

2 Methods

To determine the INP concentration spectra relevant for mixed-phase clouds in the central Arctic, 48 days of sampling were conducted aboard the Swedish icebreaker Oden during Arctic summertime and into the early freeze-up period (August and September). Filter samples were collected and analysed during the journey towards the North Pole from Svalbard whilst ice breaking, and whilst moored to an ice floe. The dates for the respective periods are: Marginal ice zone (MIZ) 02/08/18-03/08/18, Clean-air station 10/08/18-11/08/18, Ice-breaking 03/08/18-16/08/18, Ice floe 16/08/18-15/09/18, Ice-breaking 15/09/18-19/09/18, MIZ 19/09/18.

2.1 Aerosol sampling from the ship and the balloon borne platform

For the ship-based aerosol sampling, filters (0.4 µm pore size, polycarbonate, Nuclepore Track-Etched Membrane Filters, Whatman) were collected by subsampling from a heated whole-air inlet at a flow rate of 9 L min-1 (standard temperature and pressure). The inlet was mounted on the 4th deck of the ship, 25 m above

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mean sea level. This type of filter has been used previously for INP sampling and has a low background INP count and high particle recovery rates [Adams *et al.* , 2020; O'Sullivan *et al.* , 2018; Sanchez-Marroquin *et al.* , 2021]. In addition, these filters collect aerosol across the full atmospheric size distribution with high efficiency, despite having pores of 0.4 μm (smaller aerosol particles are efficiently lost to the filter surface through diffusional processes) [Adams *et al.* , 2020].

Aerosol samples from a balloon-borne sampler, the selective-height aerosol research kit (SHARK) [Porter *et al.* , 2020], were collected above the surface mixed layer. All inlets were covered until sampling was started via a radio signal from the ground. Two cascade impactors (100 L min-1, MSP Model 128, TSI, USA and 9 L min-1 Sioutas, SKC Ltd., UK) sampled aerosol. See [Porter *et al.* , 2020] for details of the size bins and how data from SHARK are treated. A radiosonde (S1H2-R, Windsond, Sweden) was used to measure the temperature, pressure and relative humidity. In order to choose an appropriate altitude for sampling, the radiosonde was constantly operating to provide information to the user on the ground about the SHARK altitude and boundary-layer temperature and humidity structure as the SHARK was ascending. In addition, sampling was paused if the relative humidity increased above 80 %, and was stopped completely before the SHARK was brought back down.

2.2 INP analysis

Filters were analysed for INP content as soon as possible after sampling, usually within 1 - 12 h of being removed from the inlet. The filter samples were not frozen before offline INP analysis, due to concerns this may affect the INP activity, but were stored at +4 °C. Performing the analysis on ship soon after sampling minimised the chances (risks?) of changes in the INPs on the filter since storage at any temperature is expected to affect the activity of the samples [Beall *et al.* , 2020]. The aerosol particles on the filters were washed into either 5 or 10 mL of ultra-pure water (Millipore Alpha-Q, with a resistivity of 18 MΩ cm at 25 °C) to suspend the collected aerosol particles. These particle suspensions were then pipetted to form an array of 1 μL droplets on a cold stage, the Microlitre Nucleation by Immersed Particle Instrument, μL-NIPI [Whale*et al.* , 2015]. The uL-NIPI is a standard INP measurement instrument that has been benchmarked alongside a range of other INP instruments during a number of intercomparison studies [DeMott*et al.* , 2018]. The cold stage cooled at a controlled rate of 1 °C min-1 until all droplets had frozen, and the freezing events were recorded in order to determine the concentration of INPs with respect to the volume of air that had been sampled through the inlet. Heat sensitivity of the collected INP samples was determined by heat treatment, where subsamples of the particle suspensions in 50 mL conical centrifuge tubes were immersed in a water bath at 100 °C for 30 min, before being reanalysed using the μL-NIPI [Daily *et al.* , 2021].

The INP concentration data presented here is shown with the contribution from the background accounted for. The background influence on the INP concentration was determined by collating the differential nucleus concentrations for water and handling blanks, and subtracting this from the sample differential nucleus concentrations. The differential concentrations were then summed to produce the cumulative INP spectra [Sanchez-Marroquin *et al.* , 2021].

2.3 Other measurements at ship level

To evaluate the concentration of dimethyl sulfide (DMS), filter samples of DMS were collected and analysed onboard. Equivalent black carbon (eBC) concentrations were obtained from a multi-angle absorption photometer (MAAP, Model 5012, Thermo Fisher Scientific Inc.). Particle size distribution measurements were made continuously using an aerosol spectrometer (WELAS 2300HP, Palas GmbH) for particles of size 0.15 - 9.65 μm, and a differential mobility particle sizer (DMPS) with a custom-built medium Vienna-type differential mobility analyzer (DMA) with a mixing condensation particle counter (MCPC, Model 1720, Brechtel Manufacturing Inc.) for particles of size 10–921 nm.

An ion chromatography system (ICS-2000, Thermo Fischer Scientific, previously Dionex) was used to determine the chemical composition of the samples. Using certain standards, the concentration of chloride, nitrate, sulphate, mesylate, methane sulfonic acid, sodium, ammonium, potassium, magnesium, calcium in the sample were determined from the ion chromatograms. A synthetic sample (QC Rainwater Standard,

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Inorganic Ventures, USA) was used to estimate the random percentrage error, which is up to 3 %. More details on the method can be found in Leck and Svensson [2015].

2.4 Prevention of ship stack pollution

Combustion products in a ship's exhaust may influence INP populations [Thomson *et al.* , 2018]. In order to ensure that the INP concentrations measured were not affected by the ship stack emissions, rigorous sampling procedures were put in place. The aerosol sampling inlets faced the ship's bow and the ship was manoeuvred to face into the wind whenever the wind direction changed, which minimised the probability of sampling ship stack emissions. In addition, an auto-stop for the inlet pumps was operated if aerosol concentrations increased suddenly (which would be indicative of sampling the ship stack plume), halting the sampling until aerosol size distributions returned to normal. As a precaution, the direction and speed of the wind was monitored closely, and sampling was stopped when there was a chance that the wind might introduce ship stack to the sampled aerosol. Finally, sampling was stopped if any activity that could produce aerosol was planned, including the movement of the ship, ice coring, and helicopter flights (this involved the operators being on call 24 hours a day to respond to any potential contamination). Smoking of cigarettes was also only allowed in certain areas of the ship, to ensure there was no influence on aerosol sampling.

2.5 Backward trajectories

In order to define the potential origin of measured INPs, backward trajectories of the air reaching the sampling location was conducted. The 10-day (only 7 days of which are used here) back trajectories were calculated using the Lagrangian analysis tool LAGRANTO [Sprenger and Wernli, 2015] with wind fields from 3-hourly operational ECMWF analyses, interpolated to a regular grid with 0.5° horizontal resolution on the 137 model levels. The trajectory data contains the hourly positions (longitude, latitude, pressure) along the trajectory. To focus on the segments of the trajectories that can potentially be affected by surface aerosol emissions, the trajectories are only included when they were within the model boundary layer. Additionally, removal of aerosol by precipitation, which may remove the signature of upwind aerosol sources via wet deposition, has been considered by removing all the trajectory points before the precipitation event (using a threshold of 0.1 mm h-1). The overall relationship with origin is unchanged by the addition of this filter, which indicates that the results were insensitive to precipitation events.

3 Results and Discussion

3.1 Ice-nucleating particle concentrations within the surface mixed layer

We first present our INP concentrations derived from samples collected on the ship, which was within the surface mixed layer (Figure 2a). The concentrations of INPs measured in the surface mixed layer were highly variable, and ranged from $< 6 \times 10^{-3}$ INP L^{-1} to 2 INP L^{-1} at -15 degC. This resulted in INP activation temperatures ranging from -9 to -30 degC for a concentration of 0.1 INP L^{-1}. This is clearly contrary to what we expected in this remote location based on measurements in other remote oceanic loactions around the world. For example, in the Southern Ocean, INP concentrations are systematically at the low end of what we observe here [McCluskey *et al.* , 2018a; Murray *et al.* , 2021; Welti *et al.* , 2020].

The vast majority of INP measurements made in the Arctic were made on land or at least some distance from the Pole. A summary of these measurements is given Figure 2b. These measurements clearly show that there are strong sources of INP between around 65 to 80degN [Hartmann*et al.* , 2021; Sanchez-Marroquin *et al.* , 2020; Tobo*et al.* , 2019; Wex *et al.* , 2015]. Our measurements demonstrate that the INP concentrations can also sporadically be very high in the pack at the North Pole. Previous measuremets at close to the Noth Pole also reveal substantial variability in INP concentrations active at -15degC [Bigg, 1996; Bigg and Leck, 2001]. Our results indicate sporadically higher concentrations than those results and also demonstrate that the concentration of INP can be in exces of 0.1 L^{-1} at temperatures up to around -10degC. We come back to the question of where these highly active INP come from later in the paper.

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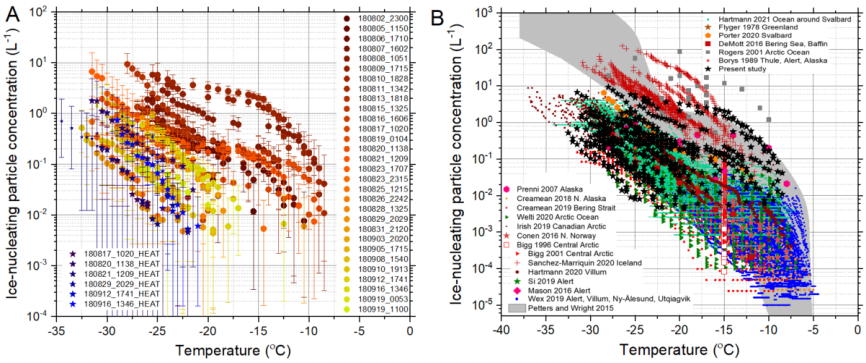

Figure 2. Surface mixed layer INP concentrations throughout the campaign. a) The number of INPs per litre of air sampled was calculated using data from offline droplet freezing experiments, conducted within hours of the samples being taken. The spectra shown in blues represent samples that were heated to close to 100 °C for 30 min. Background values were subtracted from the data. Sampling times varied from 6 h to 3 days and were taken using a heated whole air inlet on the 4th deck (25 m above mean sea level) of the Oden Icebreaker. Temperature uncertainties (not shown) for the droplet freezing experiments were estimated to be ±0.4 °C. The format of the key is YYMMDD_hhmm; these correspond to the start time of the filter sample in the 24-hour time format; the start and end times are in Table S1. b) The data from this study are presented alongside literature data for ground, ship and aircraft-based campaigns around the Arctic [Bigg, 1996; Bigg and Leck, 2001; Borys, 1989; Conen*et al.* , 2016; Creamean *et al.* , 2019; Creamean *et al.* , 2018; DeMott *et al.* , 2016; Flyger and Heidam, 1978; Hartmann *et al.* , 2020b; Hartmann *et al.* , 2021; Irish*et al.* , 2019; Mason *et al.* , 2016; Porter *et al.* , 2020; Prenni *et al.* , 2007; Rogers *et al.* , 2001; Sanchez-Marroquin *et al.* , 2020; Si *et al.* , 2019; Wex*et al.* , 2019] and a compilation derived from precipitation samples [Petters and Wright, 2015].

It is is remarkable to note that the highest concentrations measured here at the North Pole are as high as the highest INP concentration reported in INP rich environments such as the mid-latitude terrestrial environment [O'Sullivan *et al.* , 2018; Petters and Wright, 2015], despite having much lower aerosol concentrations. Overall, our INP measurements indicate that the INP concentration spectra within the high Arctic surface mixed boundary layer can be extremely variable, perhaps far more variable than anywhere else on Earth.

In order to test for the presence of proteinaceous biological ice-nucleating material, we heated the most active sample suspensions to close to 100 °C [Daily *et al.* , 2021]. The activity of these samples was always reduced, with all of the activity above -20 degC being removed (Figure 2a and Figure S1). Hence, it appears that the most active INPs sampled close to the North Pole were most likely of biological origin. Atmospheric INP at lower latitudes were also found to be heat sensitive [Hartmann *et al.* , 2021]. Together, this indicates that proteinaceous biological INP are important in the Artic.

The time series in Figure 3 shows the temperature at which a concentration of 0.1 INP L^{-1} was measured ($T_{[INP]=0.1}$), and highlights the variability of INP concentrations at the North Pole throughout August and September of 2018. The first peak in ice-nucleating activity was observed during a period in which the ship was breaking ice prior to being moored to an ice-floe (i.e. prior to 16th August). It is reasonable to question whether the very high INP concentrations observed during the ice-breaking period resulted from the ice-breaking itself. Ice-breaking involved frequent backward and forward motions, hence there is the potential for sampling ship emissions (such as ship stack emissions, detailed in the methodology) and aerosol resulting from ice-breaking and the disruption of the sea surface. We manually stopped sampling if there were activities planned that would affect sampling (such as helicopter flights) in addition to using a pollution control system that stopped the flow through the filters when aerosol concentrations increased rapidly in a manner associated with sampling a ship plume, and also when the wind was not from the correct direction (from forward of the ship's superstructure). Despite the precautions taken to eliminate these sources of

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contamination, we cannot completely exclude the possibility of contamination. However, there was a pause in ice-breaking when a clean air station was established (10th August, we sampled for 6 hours) that coincided with high INP concentrations. At this clean air station the ship was moored facing into the wind, with ship-based aerosol sources aft of the aerosol inlets. Hence, we were confident that sampling of ship pollution and ice-breaking aerosol were eliminated, increasing confidence that these high values in this period were indeed representative of the central Arctic Ocean. In addition, there was also a period of very high ice-nucleating activity a few days after the ice-floe station had been established and the ship was pointing into the wind, demonstrating that there were very active INPs that were not related to ice-breaking or ship emissions. The time series also highlights two distinct periods. The period up to the 23rd August was characterised by variable but often very high INP concentrations, whereas the period after this was characterised by much lower INP concentrations. We examine the back trajectories associated with these different periods later in the paper.

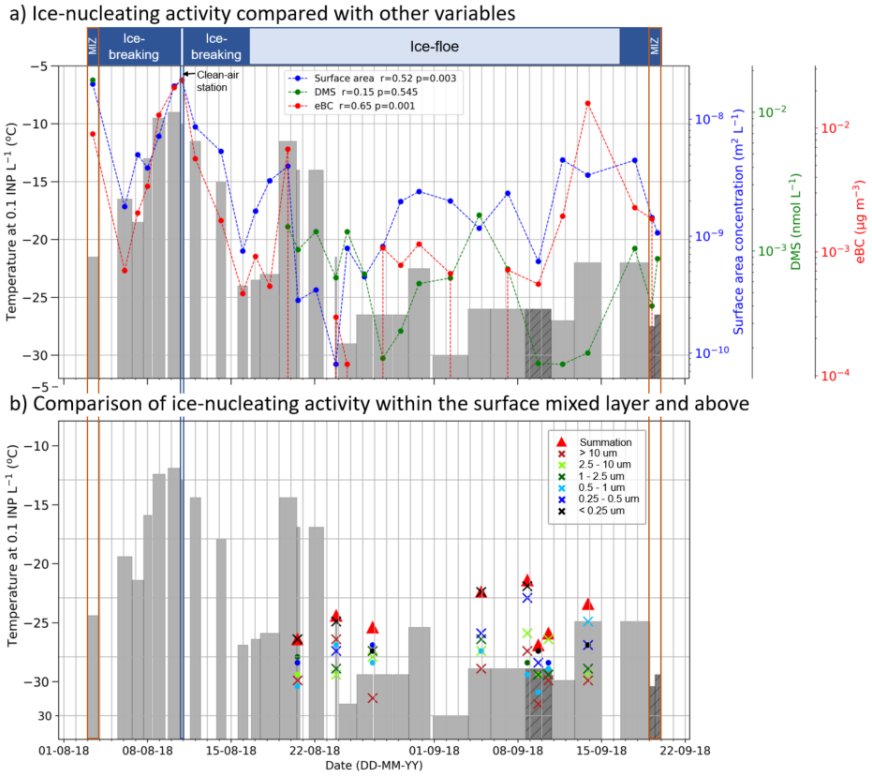

Figure 3. Time series showing the ice-nucleating activity (expressed as the temperature at which a concentration of 0.1 INP L^{-1}($T_{[INP]=0.1}$)) throughout the campaign alongside DMS, aerosol surface area and eBC. In both panels a and b, the tops of the grey bars represent $T_{[INP]=0.1}$ in the surface mixed layer (i.e. at ship level, 20 m above mean sea level), with the width of the bar representing the period over which air was sampled. The hatched grey bars are limiting values (where droplet freezing was indistinguishable from the control experiments). a) The time series of the daily average surface area of aerosol per litre (blue), the dimethyl sulfide (DMS) concentration (green) and the equivalent black carbon (eBC) concentrations (red) measured in the aerosol are shown. b) The values of $T_{[INP]=0.1}$ measured above the surface mixed layer (using the SHARK balloon-borne sampler) are shown against those at taken at ship level (grey bars). The red triangles are the $T_{[INP]=0.1}$ for the summed INP concentrations across all size categories (comparable to the measurements at ship level), while the crosses indicate the $T_{[INP]=0.1}$ associated with each size category (circles indicate limiting values. The dates for the respective periods are: MIZ 02/08/18-03/08/18, Clean-air station 10/08/18-11/08/18, Ice-breaking 03/08/18-16/08/18, Ice floe 16/08/18-15/09/18, Ice-breaking

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15/09/18-19/09/18, MIZ 19/09/18.

We also show the ice-nucleating activity in the form of ice-active sites per unit surface area (n_s) in Figure 4. The variable n_s provides a means of comparing the activity of aerosol on a per unit surface area basis. It is striking that the activity of the samples in the central Arctic are often much more active than aerosol over the Southern Ocean [McCluskey *et al.* , 2018a] or from the north Atlantic [McCluskey *et al.* , 2018b]. This shows the aerosol in this location are much more ice-active than aerosol in other remote marine environments.

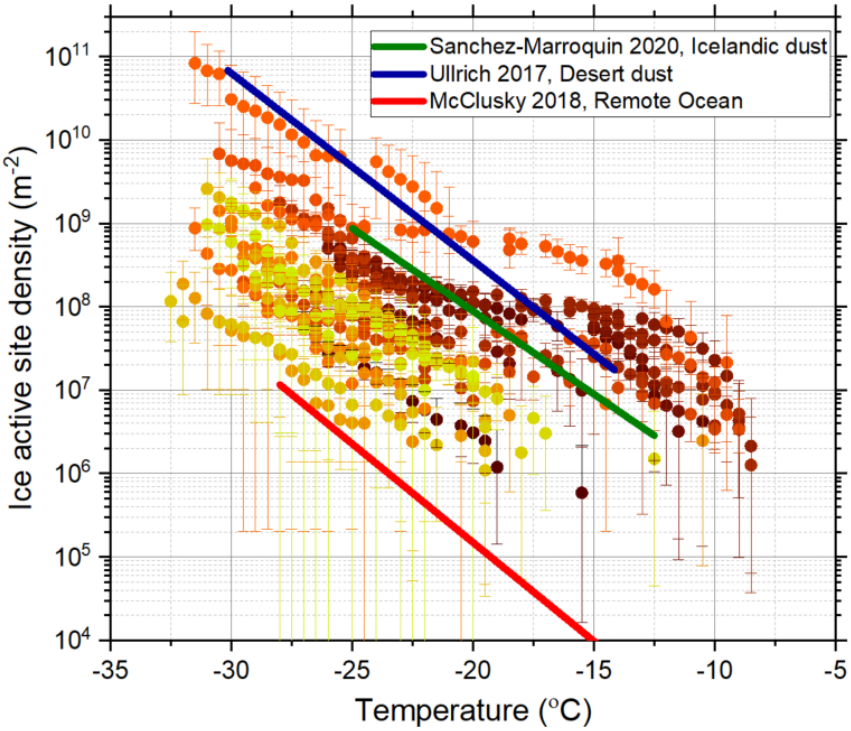

Figure 4. The ice-active site density (ns) for aerosol sampled during the cruise (see Figure 2 for key to colours) compared to $n_s$parameterisations for desert dust [Ullrich *et al.* , 2017], Icelandic dust [Sanchez-Marroquin *et al.* , 2020] and for aerosol from the North Atlantic [McCluskey *et al.* , 2018b] that is also consistent with values for aerosol over the Southern Ocean [McCluskey *et al.* , 2018a].

3.2 Ice-nucleating particle concentrations above the surface mixed layer

Eight flights with a balloon-borne size-resolved aerosol sampler were conducted while the *Oden* was at the ice station (see SI for flight details). This sampler, known as the selective height aerosol research kit (SHARK) was lofted to a defined height using a tethered balloon system operated from the sea ice and aerosol samples were collected into multiple size bins from below 0.25 μm to 10 μm with an additional stage for particles larger than 10 μm (with a poorly defined upper limit) [Porter *et al.* , 2020]. During these flights, we used a live link to the on-board temperature and humidity measurements to ensure that we sampled above the surface mixed layer and thus in air decoupled from the surface, but within the boundary layer (i.e. in the cloud mixed layer). Hence, the flights occurred at 390 m to 600 m altitude while the ship was on the ice-floe station and we sampled for 3 to 6 hours. In addition, we paused sampling when the RH was more than 80 % to avoid sampling biases associated with hygroscopically swollen aerosol and we also avoided sampling while the SHARK was enveloped in cloud.

Given the surface mixed layer is often decoupled from the rest of the boundary layer, these measurements in principle allow us to compare INP concentrations within and above the surface mixed layer. The values of T

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$_{[INP]=0.1}$ are shown in Figure 2b, whereas the INP spectra are shown in Fig SI2 for the INP summed across all the particle size bins. It should be borne in mind that, for practical reasons, the sampling durations on the ship and on the SHARK were not the same, however it is still possible to draw conclusions from this comparison. There is evidence that there are substantial differences between the INP concentrations in the surface mixed layer compared to above it. For example, on the 5th and 8th September the $T_{[INP]=0.1}$ (summed across all sizes) was around -18 to -19 °C above the surface mixed layer, whereas it was below –26 °C within it. However, on the 20th August, $T_{[INP]=0.1}$ was around –23 °C above the surface mixed layer, but –14 °C within it. On all three days, radiometer and radiosonde temperature profiles confirmed that the surface mixed layer was decoupled from the rest of the boundary layer (Table S2). In contrast, on 13th September, the surface mixed-layer was mainly coupled and the INP concentrations within and above the surface mixed layer were similar. However, on two other days (23rd August and 10th September) the activity in the surface mixed layer and above were similar even though it was decoupled. Overall, out of the eight SHARK samples collected above the surface mixed layer, there was one SHARK sample that had much lower ice-nucleating activity than that in the surface mixed layer, three samples with higher activity, three with similar activity and one that was ambiguous (due to both samples being close to the baseline of detection). This is consistent with the air at the surface sometimes being coupled to the cloud mixed layer, allowing transport of aerosol throughout the boundary layer, but at other times the measurements at the surface are not representative of those above the surface mixed layer.

The size-resolved INP activity ($T_{[INP]=0.1}$) is also shown in Figure 3b. In many locations around the world, supermicron aerosol dominate the INP population [Porter *et al.* , 2020]. However, contrary to what might be expected, the smallest size ranges of < 0.25 μm contributed the most INPs on five out of the eight flights, with the 2.5 to 10 μm and 0.5 to 1 μm bins both contributing the most on one flight each. Inspection of the corresponding INP spectra associated with each bin (Fig SI2) revealeds that the particles < 0.25 μm made a pronounced contribution to the INP population on the 23rd August and the 8th and 9th September. The other flights produced data mainly in the baseline for all sizes.

The coarse mode (>2.5 μm diameter) has a relatively short lifetime in the Arctic boundary layer, being removed effectively by wet scavenging processes [Leck and Svensson, 2015]. Hence, it is perhaps not so surprising that the fine mode aerosol (< 0.25 μm) appears to be so important in this region for the INP population. While INPs are typically thought of as being the larger particles in a size distribution [Mason *et al.* , 2016; Porter *et al.* , 2020], there are INPs that fall into the < 0.25 μm size range that are also very active. For example, film droplet aerosol resulting from wave breaking are produced in a range of sizes centred around 100-200 nm and are often rich in organic material [O'Dowd*et al.* , 2004] that is known to include small ice-nucleating entities [Schnell and Vali, 1975; Wilson *et al.* , 2015]. Alternatively, ice-nucleating macromolecules from terrestrial biological sources internally mixed with other aerosol particles might fall into this size range [O'Sullivan *et al.* , 2016; O'Sullivan *et al.* , 2015; Pummer *et al.* , 2015] and it has been proposed that fungal material, some of which is known to act as an INP [O'Sullivan*et al.* , 2015], can fragment to form nanoparticles [Lawler*et al.* , 2020].

3.3 Correlation between INP concentrations and dimethyl sulfide, equivalent black carbon and aerosol surface area

To investigate possible sources of the INPs we detected over the central Arctic Ocean, we have correlated the ice-nucleating activity of the aerosol with: i) dimethyl sulfide (DMS), a product of marine biological activity, particularly in the marginal ice zone (MIZ, the transitional zone between open sea and dense ice) [Leck and Persson, 1996]; ii) equivalent black carbon (eBC), based on aerosol absorption at 637 nm; and iii) aerosol surface area, derived from size distribution measurements. We present the time series for aerosol particle surface area, DMS and eBC concentrations, as well as the Pearson's $r$coefficient between ice-nucleating activity and each quantity in Figure 3a.

DMS is found in the marine atmosphere, originating from the metabolites of some marine algae [Leck and Persson, 1996; Lohmann and Leck, 2005]. Hence, the presence of DMS indicates that an air mass has origins in a location rich in biological activity, which may also be expected to correlate with marine biological INP

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sources. DMS is thought to be relatively short-lived in the atmosphere, with a lifetime on the order of 1-3 days [Kerminen and Leck, 2001; Khan *et al.* , 2016]. Therefore, it is a useful indicator for the interaction of air masses with the MIZ at the outer edge of the pack ice region, and possibly the open leads or melt ponds within the pack ice if they were producing DMS at that time.

The concentration of DMS during the cruise was highest in the outbound 24-hour MIZ station (2^{nd}-3^{rd}August), where the ship was close to open water, but was variable whilst in the pack ice (Figure 3), and remained relatively low in the inbound MIZ station (19^{th} September). The data in Fig. 2 clearly shows that there is no obvious correlation between DMS and INP activity ($r = 0.15$) suggesting that MIZ marine biogenic sources exerted little influence on the measured INP concentrations.

Equivalent BC (eBC) is a quantity derived from aerosol absorption and is the equivalent black carbon mass concentration needed to produce the observed absorption. Other aerosol types such as dust, brown carbon or other organic aerosol might also produce absorption, thus potentially contaminating the small signal we observed. However, absorption by BC is much stronger at 637 nm than other materials, hence the signal is most likely dominated by BC. BC is produced through a range of combustion processes, including biomass burning, wildfires and fossil fuel combustion, which are all remote from the central Arctic. Other potential contributors to the absorption signal, such as dust or brown carbon are also remote from the central Arctic. Rigorous procedures were in place to ensure that BC (and other aerosol) from the ship stack did not affect measurements (see methods for details). Therefore, eBC is used here as an indicator of long-range transport. The literature indicates that BC is a relatively ineffective ice nucleator under mixed-phase cloud conditions [Adams *et al.* , 2020; Chen *et al.* , 2018; Schill *et al.* , 2020; Vergara-Temprado *et al.* , 2018a], hence we would not necessarily interpret a positive correlation as an indication of ice nucleation by BC. However, combustion processes are thought to be a source of ice-nucleating aerosol, even if BC itself is not an effective INP [Barry *et al.* , 2021; Jahn *et al.* , 2020; Umo *et al.* , 2015]. Thus, a correlation between BC and INP concentrations would indicate that aerosol particles transported along with BC from outside the central Arctic Ocean nucleate ice. Wildfires around the Arctic are a potential source of BC, and we note that during the cruise there were persistent Siberian wildfires, as can be seen using the NASA Worldview satellite imagery tool (e.g. clear skies on the 14^{th} August reveal widespread fires; https://worldview.earthdata.nasa.gov/). There is also industry, shipping and mining along the Arctic coast of Russia, but the role of gas flaring is thought to be a particularly important source of BC [Stohl *et al.* , 2013].

The overall correlation between eBC and INP concentration ($r = 0.65$) is much stronger than for DMS. In fact, the eBC concentration appears to track the INP concentration in Figure 3 until the 27^{th} August, after which the INP concentrations stay relatively low whilst the eBC remains highly variable. The decoupling of eBC and INPs later in the campaign indicates that distant sources of BC are not always related to distant sources of highly active INPs.

The surface area concentration of the bulk aerosol follows a similar trend to the eBC concentrations, but with a slightly weaker correlation with the INP activity ($r = 0.52$). This indicates that the variability in INP concentrations at the North Pole is not simply driven by aerosol surface area, rather that some specific component(s) of the aerosol population are ice-active and these particles are most likely associated with specific sources at latitudes further south than the MIZ.

3.4 Trajectory analysis of aerosol collected in the central Arctic

Backward trajectories from the sampling location near the surface are presented in Figure 5a. We only show points that are in in the boundary layer and for a maximum of 7 days. The figure shows a clear relationship between the origin of the aerosol – considered here as the boundary layer points along the trajectories - and the measured INP concentrations. The origin of the air with the most active INPs is around the Russian Arctic coast including the Barents, Kara and Laptev Seas. Out of the 30 filter runs, those filters with the highest INP activity (the top 20 % of filters; -9 °C [?] $T_{[INP]=0.1}$ > -13.5 degC) sampled air masses originating over the Barents and Kara Seas. The next seven highest (23 %; -13.5 degC [?] $T_{[INP]=0.1}$> -22 degC) filters, in terms of INP activity, sampled air originating from over the Laptev and East Siberian Seas. The next

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six filters with lower INP activity (20 % of filters; -22 degC [?] $T_{[INP]=0.1}$ > -25 degC) sampled air that originated off the eastern coast of Greenland from over both the pack ice and open ocean. The 11 filters with the lowest INP activity (bottom 37 % of filters; -25 degC [?] $T_{[INP]=0.1}$ [?] -30 degC) all sampled air which mostly originated from the pack ice adjacent to North America (also see Figure SI3).

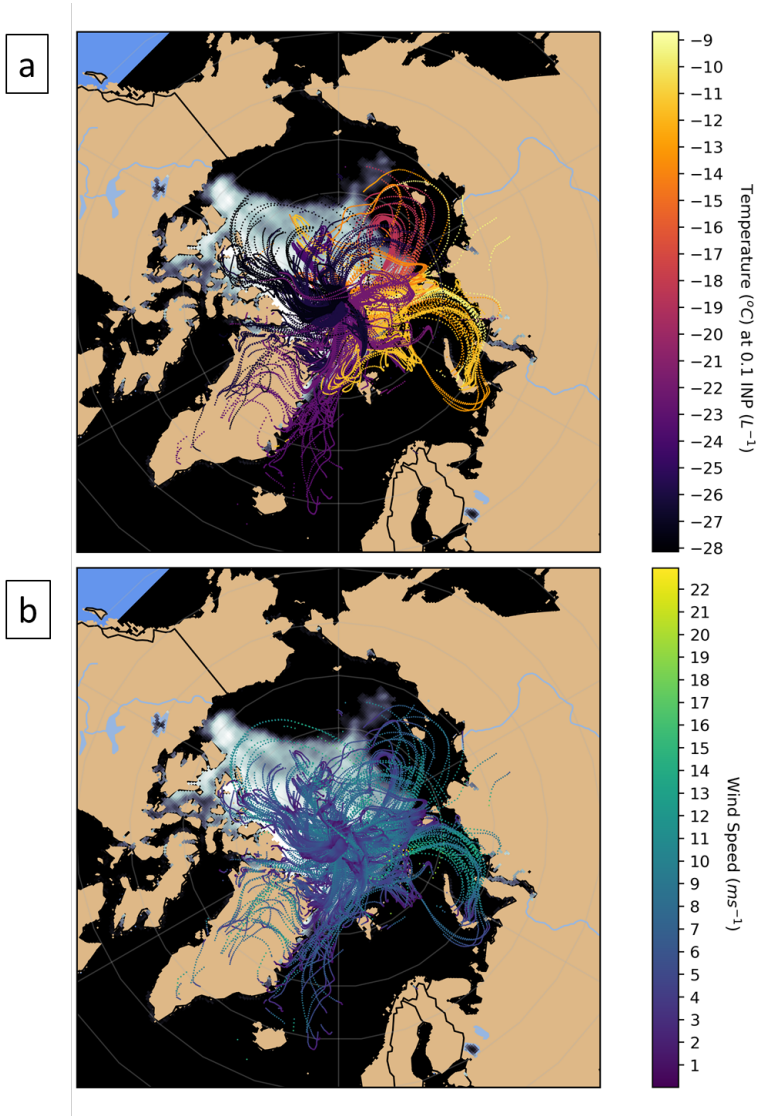

Figure 5. Backward trajectories over 7 days, starting at the ship location, for the INP samples taken throughout the campaign. Trajectories were launched every hour during the sampling period, and each point represents an hour in time along the back trajectory. The starting height for the trajectories was 32 m above mean sea level. Any points along the trajectories which were above the model boundary layer were removed, and any points along the trajectory that preceded precipitation events (>0.1 mm h-1) were removed. Hence, any potential sources of INP in the boundary layer are neglected if they occurred prior to a precipitation event (we assume precipitation removes INPs) a) The colour of the trajectories represents the temperature at which 0.1 INP L-1 was measured for that sampling period. b) The colour of the trajectories represents the wind speed for each point along the trajectory. The sea ice extent is from the NASA National

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Snow and Ice Data Center [Maslanik and Stroeve, 1999].

It is striking that the trajectories with the lowest INP concentrations spent most of the preceding seven days over the pack ice and to some extent over the MIZ. These results indicate that, during this campaign, open leads, sea ice and the MIZ were weak sources of INPs, in conflict with previous suggestions [Bigg and Leck, 2001; Hartmann *et al.* , 2020a].

The highest ice-nucleating activities from sampled aerosol originating along the Russian coast were also correlated with high wind speeds along the trajectories, i.e., in the region of the aerosol origin (Figure 5b). This, together with the heat tests and INP size information, point to a wind-driven marine biological source of INPs associated with organic-rich film droplet sea spray aerosol. There were trajectories with high wind speeds over the North American continent, the pack ice and the coast of Greenland, but the ice-nucleating activity for these was not greatly enhanced. Hence, our results are consistent with a strong source of highly active INPs in the coastal marine waters of northern Russia which were aerosolised during windy conditions. Marine waters elsewhere in the world are thought to produce aerosol with relatively low ice nucleating activities [Vergara-Temprado *et al.* , 2017] (see Figure 4); however, our results suggest that the shallow seas off the Russian coast might be relatively strong sources of highly active INPs. Composition analysis of the aerosol during the peak ice nucleation activity on the 11^{th} August and the 19^{th} August is consistent with a marine source for many of these aerosol particles (samples were rich in Na, Cl and sulfate; see Table S3). Hence, the question is why the aerosol from near the coast of Russia is so much more active than aerosol derived from other marine locations such as the North Atlantic and the Southern Ocean [McCluskey *et al.* , 2018a; McCluskey *et al.* , 2018b].

A major difference to the North Atlantic and Southern Ocean is that the shallow seas off the coast of northern Russia are strongly influenced by riverine input from Russia that is rich in organic material, silt and nutrients [Ahmed *et al.* , 2020; Juhls *et al.* , 2019]. In fact, much of the dissolved organic matter in the Arctic Ocean is derived from river input [Juhls *et al.* , 2019] and the discharge of these rivers (and the amount of dissolved organic carbon flowing in the sea via rivers) is increasing [Ahmed *et al.* , 2020; Juhls *et al.* , 2020]. It has been shown that melting permafrost, which is known to enter river water [Juhls *et al.* , 2020], harbors copious quantities of warm temperature INPs [Creamean*et al.* , 2020]. Hence, it is possible that the highest INP concentrations we detected at the North Pole were derived from marine waters rich in terrestrially derived ancient biological INPs. Alternatively, ocean biology fertilised by the nutrient rich waters on the continental shelf may produce more INPs than are present in remote marine locations. A measurement campaign to quantify the INP content of the waters off the coast of Russia is clearly required.

Some of the back trajectories that had the highest INP concentrations passed over islands in the Barents and Kara Seas, including Svalbard, Franz Josef Land, Novaya Zemlya and Severnaya Zemlya. Many of these locations have been identified as poorly defined dust sources [Bullard*et al.* , 2016] and dust from Svalbard has been shown to contain biological ice-nucleating materials [Tobo *et al.* , 2019]. However, in a further analysis of the back trajectory data (Figure 6), we find that there was little to no correlation with time spent over land, whereas the ice-nucleating activity increased with the time the air parcels spent over open ocean. This implies that the sources of INP were associated with the marine environment. Having said this, we cannot rule out relatively small island point sources being important sources of INPs.

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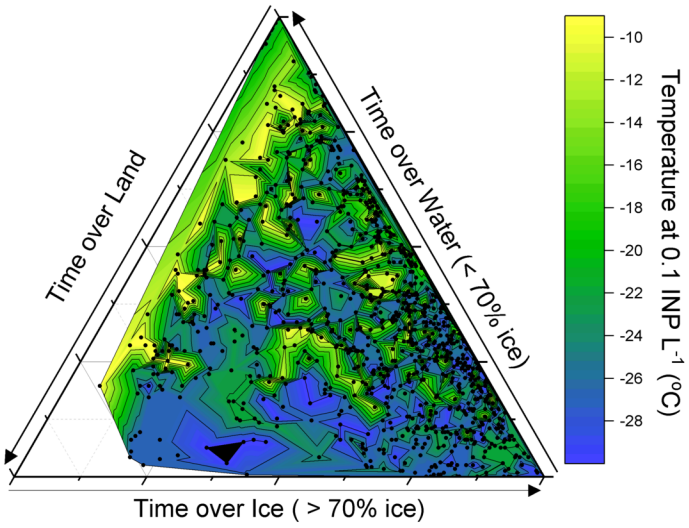

Figure 6. The time that air masses spent over water, ice and land derived using back trajectory analysis. The colour represents the temperature at which an INP concentration of 0.1 INP L-1 was reached. The black dots correspond to individual trajectories. The same filters as in Figure 5 were applied.

Overall, the evidence indicates that there is a strong source of biogenic INPs in the Barents, Kara and Laptev Seas off the Russian coast that can be sporadically transported to the central Arctic Ocean. There was high wind along the trajectories off the Russian coast which would be consistent with both the production of INPs in sea spray from these organic-rich seas or dust combined with terrestrial biogenic material from the various islands in this region. We also note that a recent study found that very active INP were produced in the Chukchi sea (near Alaska) under high wave conditions [Inoue *et al.* , 2021]. Furthermore, we note that these potential sources are sensitive to a changing climate, with river discharge, permafrost melt and organic matter input into the ocean from the major Russian rivers increasing in a warmer world [Jahn *et al.* , 2020]. In addition, the removal of ice from the Arctic could also expose marine and terrestrial sources of ice-nucleating aerosol around the Arctic Ocean, where they can then be aerosolised by the action of wind [Schmale *et al.* , 2021].

3.5 Implications for ice production in boundary layer central Arctic mixed-phase clouds

In this section, we assess whether the measured INP concentrations are high enough to initiate a transition from liquid-dominated clouds to ice-dominated clouds. Model simulations indicate that on the order of one ice crystal per litre of air is required to remove the bulk of liquid water from an Arctic cloud, whereas lower concentrations still reduce the liquid water path [Stevens *et al.* , 2018; Vergara-Temprado *et al.* , 2018b]. Hence, we use our INP measurements, both at ship level in the surface mixed layer and those from the SHARK in the cloud mixed layer, to estimate the concentration of INPs that become active at the ambient temperature of the atmosphere ($[\mathrm{INP}]_{\mathrm{ambient}}$).

The quantity $[\mathrm{INP}]_{\mathrm{ambient}}$ combines the atmospheric temperature profiles from radiosonde measurements with the corresponding INP spectra as a rough indicator of primary ice crystal production. This is a crude analysis and a full cloud model would be required to represent ice crystal formation and sedimentation as well as INP recycling and latent heat release, to predict ice crystal concentration given an INP spectrum, but it does give an indication of what the measured INP spectra might mean for primary ice production in

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clouds. The highest $[INP]_{ambient}$ will be at the cold points, i.e. the top of the surface mixed layer and the top of the boundary layer (top of the cloud mixed layer), hence this is where we focus this analysis. We assume that ship level measurements are representative of the $[INP]_{ambient}$ at the top of the surface mixed layer and those from SHARK are representative of the top of the boundary layer, the assumption being that these respective layers are individually well-mixed.

The temperature minima within the main boundary layer were determined from radiosonde profiles, which were made every 6 hours throughout the entire cruise [Vüllers *et al.* , 2021]. The calculated $[INP]_{ambient}$ for the duration of the cruise are shown in Figure 7, where the minimum temperature at the top of the cloud mixed layer (main boundary layer) and the top of the surface mixed layer is indicated for each filter period. In many cases, the temperature of the atmosphere was higher than the highest INP measurement, hence we are only able to report upper limits to $[INP]_{ambient}$ in these cases.

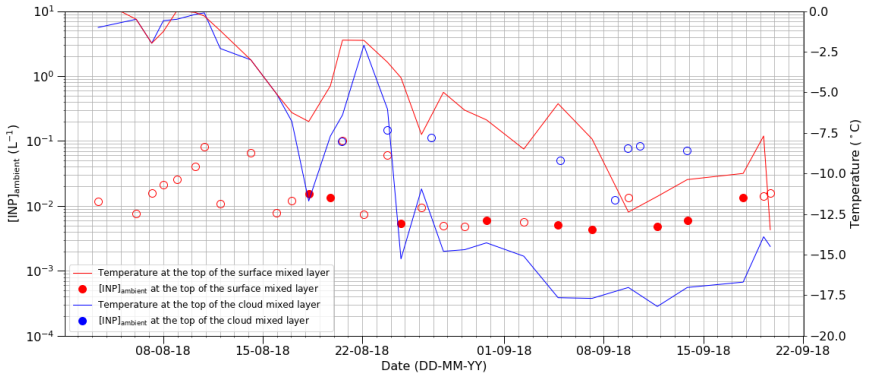

Figure 7. Time series showing [INP]ambient for both the measurements taken at ship height (within the surface mixed layer) and using the SHARK (within the cloud mixed layer). The temperature of the mixed layers is shown alongside these measurements. When an INP spectrum did not extend to the atmospheric temperature (i.e. where the highest temperature at which an INP concentration was reported was below the atmospheric temperature), the [INP] value associated with the highest temperature is provided as an upper limit to [INP]ambient (open symbols).

Generally, $[INP]_{ambient}$ was typically below about 0.1 INP L^{-1} at the top of both the surface mixed layer and the top of the cloud mixed layer. The periods of high INP concentration before the 23rd August coincide with periods of higher ambient inversion temperatures, whereas later in the campaign the opposite is the case, which results in a relatively invariant $[INP]_{ambient}$. Whether this is a coincidence, or if INP concentrations are correlated with ambient temperature, is unclear from this limited dataset. However, there may be a physical mechanism behind this apparent correlation. Transport of air to the North Pole from sources further afield will likely result in multiple cycles of cloud formation and dissipation in any one air mass, hence INP active at above the ambient temperature of those clouds will likely activate and be removed via precipitation (bearing in mind that the majority of INPs relevant for mixed-phase clouds only activate to ice in the presence of water droplets [Murray *et al.* , 2012]). While transport through the boundary layer likely removes INP active above the lowest temperatures experienced by an air parcel, further cooling subsequent to the measurement time will lead to primary ice production. Hence, the air masses sampled before the 23rd August with high INP concentrations have a great potential for primary ice production if or when these air masses become colder downwind of the sampling location. In the absence of local sources, the INP spectrum over the central Arctic Ocean must therefore be determined by a combination of the characteristics of the upwind sources and the cloud temperature that these air parcels experience on transport.

The relatively low $[INP]_{ambient}$ values suggest that clouds not influenced by seeding from above should be mixed-phase, i.e. contain a substantial proportion of liquid water with some ice crystals. Observations of the

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phase of clouds during this campaign are discussed in [Vüllers *et al.* , 2021]. Overall, the fraction of single layer clouds (where seeding from above is unlikely) throughout the troposphere that were mixed-phase was relatively constant throughout the campaign: mixed phase frequency ˜20-30 %, ice cloud frequency ˜10-30 % and liquid clouds were generally infrequent; see Figure 15 in [Vüllers *et al.* , 2021]), despite the strong decrease in temperature during the campaign. Similarly, in the bottom few kilometres the frequency of occurrence of mixed-phase clouds were often between 25% and 50%, with ice clouds about half as frequent and liquid only clouds much less frequent throughout the campaign. Overall, the observations of cloud phase for single-layer clouds reported by [Vüllers *et al.* , 2021] are qualitatively consistent with our relatively invariant $[INP]_{ambient}$ measurements.

Clouds were multi-layered around 50 % of the time, with many situations identified where seeding of ice from higher, colder clouds into lower clouds might occur. Clouds were regularly observed up to around 8 or 9 km, where temperatures [Vüllers *et al.* , 2021] were low enough for homogeneous freezing [Herbert *et al.* , 2015], and frequently occurred in the mid-troposphere where heterogeneous nucleation on INPs was most likely important. These higher clouds were often in the free troposphere where our boundary layer INP measurements are not necessarily relevant. Indeed, aerosol and INPs in the free troposphere may have different sources to those in the boundary layer. In order to obtain a more complete picture of primary ice production in clouds in the central Arctic, INP measurements in the free troposphere in this region would be needed and should be a target of future campaigns.

4 Summary and conclusions

Arctic mixed-phase and supercooled clouds play a crucial role in Arctic climate, but the processes that dictate their characteristics are poorly understood. Here, we show that INP concentrations at 88 - 90°N are extremely variable, and throughout the MOCCHA campaign between the 1st of August 2018 and the 18th of September 2018 the temperature at which 0.1 INP L^{-1}was reached varied between -9 degC and -30 degC. The highest 20% of observed INP activity is related to air masses originating in the ice-free ocean environment off the Russian coast, while the lowest 37 % of observations related to air masses which originated and circled over the pack ice north of Canada for most of the 7-day back trajectory. Trajectories of air with intermediate INP activity also originated over the ice-free ocean. These results indicate a strong dependence of the measured INP concentration on the origin of the air with pack ice, open leads, and the MIZ apparently being weak sources of INP, whereas ice-free oceans, especially those near the Russian coast when wind speeds were high, were a significant source.

The heat sensitivity of the most active INPs indicates the INP to be proteinaceous, biogenic origin. This, together with the trajectory analysis, indicates that there are strong biogenic sources of INP in the shallow seas over the Russian continental shelf. The ice-nucleating activity of the aerosol at the North Pole derived from off the coast of Russia is much greater than that for sea spray aerosol in remote oceans (such as the Southern Ocean [McCluskey *et al.* , 2018a] or the North Atlantic [McCluskey *et al.* , 2018b]). This may indicate the marine waters off Russia are very rich in ice-nucleating material, perhaps related to the substantial riverine input, or alternatively the islands in this region may be sources of biogenic INPs. More work is needed to define what the key sources are along the Russian coast and to see if similar sources exist elsewhere around the Arctic and Antarctic.

By making measurements of INP spectra both above and within the surface mixed layer of decoupled boundary layers, we found that surface measurements were often not representative of the INPs in the cloud mixed layer. Hence, measurements at altitude, within the cloud mixed layer, are necessary in order to define primary ice production in Arctic mixed-phase clouds. In addition, our measurements allowed us to estimate the INP concentration active at the temperature of the top of the surface mixed layer and also at the top of the boundary layer. This revealed that, despite massive variability in INP spectra, the INP concentration at ambient temperature was typically less than 0.1 L^{-1}, which is consistent with remote sensing observations that indicate the persistence of mixed-phase clouds (in the absence of seeding of ice from above). We also recommend future studies focus on INP measurements throughout the free troposphere where primary ice production may lead to seeding of ice in lower level clouds.

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Overall, it is striking that INP concentrations at the summertime North Pole vary from some of the lowest measured anywhere in the world, to as high as the highest INP concentrations in terrestrial locations rich in biological INPs such as in the UK [O'Sullivan *et al.* , 2018]. Since these INPs are transported from the seas off the Russian coast, they may be sensitive to changes in climate. In particular, reduced sea, land ice and permafrost may open up more sources for more of the year around the Arctic, which may increase the future strength (and may already have done so) of the sources of INPs that are important for mixed-phase clouds in the central Arctic. More work needs to be undertaken to understand how climate change may affect INP sources around the periphery of the Arctic and how this may influence Arctic clouds and feedback on Arctic climate.

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Competing interests

All other authors declare they have no competing interests.

Data Availability Statement

All data will be made available via the Research Data Leeds Repository and the Bolin Centre Database.

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Highly active ice-nucleating particles at the summer North Pole

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Key Points:

- The concentration of ice-nucleating particles at the North Pole in summer 2018 was amongst the highest anywhere in the world.
- These biological ice-nucleating particles were derived from the Russian seas and perhaps associated with wind-driven sea spray.
- The concentration of ice-nucleating particles at the surface was often different to that higher in the boundary layer where clouds form.

Abstract

The amount of ice versus supercooled water in clouds defines their radiative properties and role in climate feedbacks. Hence, knowledge of the concentration of ice-nucleating particles (INPs) is needed. Generally, the concentrations of INP is found to be very low in remote marine locations allowing clouds to persist in a supercooled state. However, little is known about the INP population in clouds at and around the summertime North Pole. We had expected that concentrations of INPs at the North Pole would have been very low given the distance from open ocean and terrestrial sources coupled with effective wet scavenging processes. Here we show that during summer 2018 (August and September) high concentrations of biological INPs (active at >-20°C) were present at the North Pole. In fact, INP concentrations were sometimes as high as those recorded in mid-latitude locations strongly impacted by highly active biological

INPs, in strong contrast to the Southern Ocean. Furthermore, using a balloon borne sampler we demonstrated that INP concentrations were often different at the surface versus higher in the boundary layer where clouds form. Back trajectory analysis suggests that there were strong sources of INPs near the Russian coast, possibly associated with wind-driven sea spray production, whereas the pack ice, open leads, and the marginal ice zone were not sources of highly active INPs. These findings suggest that primary ice production, and therefore Arctic climate, is sensitive to transport from locations such as the Russian coast that are already experiencing marked climate change.

Plain Language Summary

Clouds play a critical role in Earth's climate, both reflecting incoming sunlight and trapping outgoing heat energy. Hence, even small errors in the representation of clouds in climate models can lead to uncertainty in predictions of, for example, sea ice extent. In the Arctic, clouds often exist below 0°C and cloud water droplets can exist in a supercooled liquid state. In the absence of a special class of particle that can trigger ice formation in droplets, ice-nucleating particles (INPs), supercooled water droplets can cool well below -35°C before spontaneously freezing. Hence, the presence of INPs can reduce the amount of supercooled water in clouds, making them less reflective with a shorter lifetime. Based on our knowledge of INPs in other remote oceans, we expected very low INP concentrations in the central Arctic. However, we have shown that there are high concentrations of biological INPs in the summertime North Pole. Furthermore, these INPs come from the seas off the coast of Russia, a region already experiencing strong climate change. It is possible that these sources may become even more important as the Arctic becomes increasingly ice-free, causing changes in Arctic clouds and further changes in climate.

1 Introduction

The Arctic climate is strongly influenced by ubiquitous low-level mixed-phase clouds [Kay and L'Ecuyer, 2013; Tjernstrom *et al.*, 2012; Vüllers *et al.*, 2021]. The radiative effect of these clouds is influenced by the amount of ice and supercooled water they contain, which depends on an intricate balance of dynamical and microphysical processes [Morrison *et al.*, 2012]. Realistic representation of these processes is needed to correct model biases in the amount of supercooled liquid in mixed-phase clouds and reduce uncertainty in feedbacks [Tan and Storelvmo, 2019].

A rare subset of the total aerosol particle population, ice-nucleating particles (INPs), can induce primary ice production in Arctic mixed-phase clouds when immersed in supercooled cloud droplets [Murray *et al.*, 2012]. In the summertime, Arctic marine atmospheric boundary layer temperatures are usually much warmer than those required for homogeneous freezing (–35 °C) [Herbert *et al.*, 2015], hence heterogeneous nucleation on INPs determines the production of ice in clouds, at least in the absence of ice precipitating from overlying clouds [Vassel *et al.*, 2019]. Numerous INP types that can induce nucleation over a large

range of temperatures have been identified [Hoose and Möhler, 2012; Kanji *et al.*, 2017; Murray *et al.*, 2012]. However, the sources and ice-nucleating properties of INPs in the Arctic, especially the central Arctic (>80° N), are poorly defined.

INP measurements have been made around the periphery of the Arctic circle from locations close to, or on, land, but relatively few measurements have been made in the summertime central Arctic Ocean (see compilations in [Welti *et al.*, 2020] and [Murray *et al.*, 2021]. Recent research suggests that there are significant terrestrial sources of Arctic INPs including glacial dust from Svalbard [Tobo *et al.*, 2019] and Iceland [Sanchez-Marroquin *et al.*, 2020], terrestrial biological aerosol from boreal forests [Schneider *et al.*, 2021], and even particles released from thawing permafrost [Creamean *et al.*, 2020]. There is also a plethora of other high latitude dust sources that have not been investigated in terms of their ice-nucleating ability [Bullard *et al.*, 2016]. Marine biogenic INPs emitted from the sea surface through bubble bursting are also thought to contribute to the INP population of the oceanic high-latitudes [Bigg, 1996; Bigg and Leck, 2001; Hartmann *et al.*, 2020a; Hartmann *et al.*, 2021; Ickes *et al.*, 2020; Irish *et al.*, 2017; Wilson *et al.*, 2015]. Sea spray is thought to produce relatively low INP concentrations, but in the absence of other INP types it can dominate the INP population [McCluskey *et al.*, 2018a; Vergara-Temprado *et al.*, 2017].

Ground level observations at several land-based sites around the Arctic throughout the seasonal cycle showed the highest (but variable) INP concentrations during spring, summer and autumn and the lowest concentrations in winter [Wex *et al.*, 2019]. These measurements suggest that there are marine and terrestrial INP sources around the Arctic, but it is unclear how important these sources are for clouds over the summertime central Arctic Ocean. Based on back trajectory analysis of INP measurements in the central Arctic, Bigg [1996] suggested that there was an open ocean source of INPs active at –15 °C. Later, Bigg and Leck [2001] suggested the pack ice edge and bubble bursting in local leads throughout the pack ice can serve as a source of INPs. Indeed, it has been shown that there is a reservoir of INPs in the seas around the Arctic [Creamean *et al.*, 2019; Hartmann *et al.*, 2021; Irish *et al.*, 2017; Wilson *et al.*, 2015] and INP concentrations in the central Arctic decrease during the transition from Arctic summer to autumn, possibly due to the reduced availability of ice-free marine sources [Bigg and Leck, 2001].

While it is clear that there are strong sources of INPs in the lower Arctic environment (80°N), it is not clear if these INPs are transported to the central Arctic. The prevailing view is that aerosol within the summertime high Arctic boundary layer experiences little effect from long-range transport [Kupiszewski *et al.*, 2013], and with few sources of primary aerosol in the central Arctic Ocean, sources such as local leads may be important [Bigg and Leck, 2001]. However, it has also been suggested that aerosol particles can be transported from lower latitudes into the central Arctic boundary layer either through boundary layer transport or entrainment from the free troposphere [Igel *et al.*, 2017; Morrison

et al., 2012; Schmale *et al.*, 2021].

The structure of the Arctic summertime boundary layer is complex (Figure 1). The boundary layer is typically several hundred meters to over a kilometer deep, but often consists of two distinct layers: the surface mixed layer and the cloud mixed layer. These two layers are each well mixed, but separated by a decoupling layer at ~100 m to 300 m that prevents efficient transport between them [Brooks *et al.*, 2017]. Hence, measurements at the surface are not necessarily representative of those in the cloud mixed layer.

Here, we present measurements of INP concentrations close to the North Pole both in and above the surface mixed layer. The measurements were made during the Microbiology-Ocean-Cloud-Coupling in the High Arctic (MOCCHA) campaign, which took place throughout August and September 2018 on the Swedish icebreaker Oden. Measurements took place while Oden was on route to the North Pole as well as when it was moored to an ice floe in the inner pack ice and drifting passively between 88-90°N. Samples were collected for INP analysis at both ship level (in the surface mixed layer) and using a balloon-borne sampler in the cloud mixed layer. We use backward trajectories alongside other measurements to suggest that the source of the most active INPs reaching the North Pole is outside of the pack ice, and near the Arctic coast of Russia.

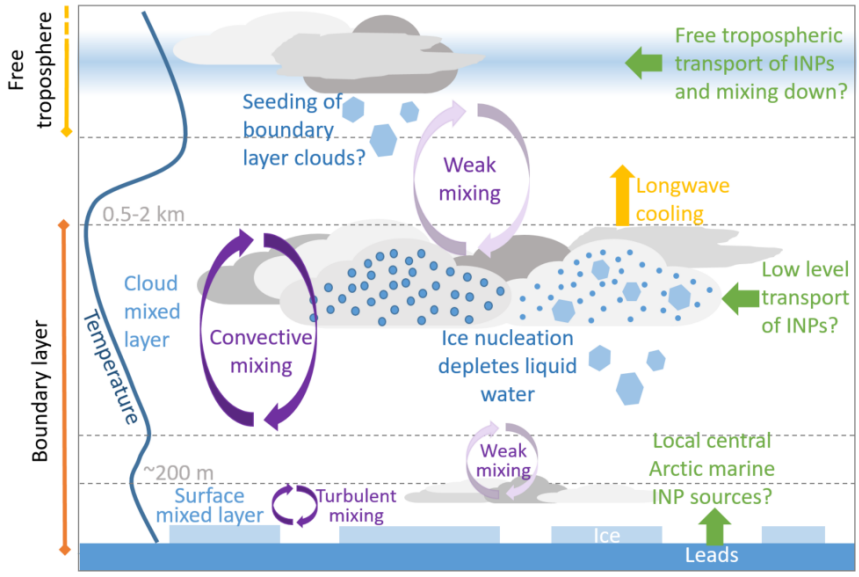

Figure 1. Central Arctic boundary layer structure and potential sources of INPs. The lowest part of the boundary layer (surface mixed layer) is often decoupled from the rest of the boundary layer [Brooks *et al.*, 2017]. In this paper we report INP measurements both in the surface mixed layer and in the cloud mixed layer and use these measurements to infer information about the

dominant sources of INPs in the central Arctic boundary layer.

2 Methods

To determine the INP concentration spectra relevant for mixed-phase clouds in the central Arctic, 48 days of sampling were conducted aboard the Swedish icebreaker Oden during Arctic summertime and into the early freeze-up period (August and September). Filter samples were collected and analysed during the journey towards the North Pole from Svalbard whilst ice breaking, and whilst moored to an ice floe. The dates for the respective periods are: Marginal ice zone (MIZ) 02/08/18-03/08/18, Clean-air station 10/08/18-11/08/18, Ice-breaking 03/08/18-16/08/18, Ice floe 16/08/18-15/09/18, Ice-breaking 15/09/18-19/09/18, MIZ 19/09/18.

2.1 Aerosol sampling from the ship and the balloon borne platform

For the ship-based aerosol sampling, filters (0.4 m pore size, polycarbonate, Nuclepore Track-Etched Membrane Filters, Whatman) were collected by sub-sampling from a heated whole-air inlet at a flow rate of 9 L min-1 (standard temperature and pressure). The inlet was mounted on the 4th deck of the ship, 25 m above mean sea level. This type of filter has been used previously for INP sampling and has a low background INP count and high particle recovery rates [Adams *et al.*, 2020; O'Sullivan *et al.*, 2018; Sanchez-Marroquin *et al.*, 2021]. In addition, these filters collect aerosol across the full atmospheric size distribution with high efficiency, despite having pores of 0.4 µm (smaller aerosol particles are efficiently lost to the filter surface through diffusional processes) [Adams *et al.*, 2020].

Aerosol samples from a balloon-borne sampler, the selective-height aerosol research kit (SHARK) [Porter *et al.*, 2020], were collected above the surface mixed layer. All inlets were covered until sampling was started via a radio signal from the ground. Two cascade impactors (100 L min-1, MSP Model 128, TSI, USA and 9 L min-1 Sioutas, SKC Ltd., UK) sampled aerosol. See [Porter *et al.*, 2020] for details of the size bins and how data from SHARK are treated. A radiosonde (S1H2-R, Windsond, Sweden) was used to measure the temperature, pressure and relative humidity. In order to choose an appropriate altitude for sampling, the radiosonde was constantly operating to provide information to the user on the ground about the SHARK altitude and boundary-layer temperature and humidity structure as the SHARK was ascending. In addition, sampling was paused if the relative humidity increased above 80 %, and was stopped completely before the SHARK was brought back down.

2.2 INP analysis

Filters were analysed for INP content as soon as possible after sampling, usually within 1 - 12 h of being removed from the inlet. The filter samples were not frozen before offline INP analysis, due to concerns this may affect the INP activity, but were stored at +4 °C. Performing the analysis on ship soon after sampling minimised the chances (risks?) of changes in the INPs on the filter

since storage at any temperature is expected to affect the activity of the samples [Beall *et al.*, 2020]. The aerosol particles on the filters were washed into either 5 or 10 mL of ultra-pure water (Millipore Alpha-Q, with a resistivity of 18 MΩ cm at 25 °C) to suspend the collected aerosol particles. These particle suspensions were then pipetted to form an array of 1 µL droplets on a cold stage, the Microlitre Nucleation by Immersed Particle Instrument, µL-NIPI [Whale *et al.*, 2015]. The uL-NIPI is a standard INP measurement instrument that has been benchmarked alongside a range of other INP instruments during a number of intercomparison studies [DeMott *et al.*, 2018]. The cold stage cooled at a controlled rate of 1 °C min-1 until all droplets had frozen, and the freezing events were recorded in order to determine the concentration of INPs with respect to the volume of air that had been sampled through the inlet. Heat sensitivity of the collected INP samples was determined by heat treatment, where subsamples of the particle suspensions in 50 mL conical centrifuge tubes were immersed in a water bath at 100 °C for 30 min, before being reanalysed using the µL-NIPI [Daily *et al.*, 2021].

The INP concentration data presented here is shown with the contribution from the background accounted for. The background influence on the INP concentration was determined by collating the differential nucleus concentrations for water and handling blanks, and subtracting this from the sample differential nucleus concentrations. The differential concentrations were then summed to produce the cumulative INP spectra [Sanchez-Marroquin *et al.*, 2021].

2.3 Other measurements at ship level

To evaluate the concentration of dimethyl sulfide (DMS), filter samples of DMS were collected and analysed onboard. Equivalent black carbon (eBC) concentrations were obtained from a multi-angle absorption photometer (MAAP, Model 5012, Thermo Fisher Scientific Inc.). Particle size distribution measurements were made continuously using an aerosol spectrometer (WELAS 2300HP, Palas GmbH) for particles of size 0.15 - 9.65 µm, and a differential mobility particle sizer (DMPS) with a custom-built medium Vienna-type differential mobility analyzer (DMA) with a mixing condensation particle counter (MCPC, Model 1720, Brechtel Manufacturing Inc.) for particles of size 10–921 nm.

An ion chromatography system (ICS-2000, Thermo Fischer Scientific, previously Dionex) was used to determine the chemical composition of the samples. Using certain standards, the concentration of chloride, nitrate, sulphate, mesylate, methane sulfonic acid, sodium, ammonium, potassium, magnesium, calcium in the sample were determined from the ion chromatograms. A synthetic sample (QC Rainwater Standard, Inorganic Ventures, USA) was used to estimate the random percentrage error, which is up to 3 %. More details on the method can be found in Leck and Svensson [2015].

2.4 Prevention of ship stack pollution

Combustion products in a ship's exhaust may influence INP populations [Thomson *et al.*, 2018]. In order to ensure that the INP concentrations measured were

not affected by the ship stack emissions, rigorous sampling procedures were put in place. The aerosol sampling inlets faced the ship's bow and the ship was manoeuvred to face into the wind whenever the wind direction changed, which minimised the probability of sampling ship stack emissions. In addition, an auto-stop for the inlet pumps was operated if aerosol concentrations increased suddenly (which would be indicative of sampling the ship stack plume), halting the sampling until aerosol size distributions returned to normal. As a precaution, the direction and speed of the wind was monitored closely, and sampling was stopped when there was a chance that the wind might introduce ship stack to the sampled aerosol. Finally, sampling was stopped if any activity that could produce aerosol was planned, including the movement of the ship, ice coring, and helicopter flights (this involved the operators being on call 24 hours a day to respond to any potential contamination). Smoking of cigarettes was also only allowed in certain areas of the ship, to ensure there was no influence on aerosol sampling.

2.5 Backward trajectories

In order to define the potential origin of measured INPs, backward trajectories of the air reaching the sampling location was conducted. The 10-day (only 7 days of which are used here) back trajectories were calculated using the Lagrangian analysis tool LAGRANTO [Sprenger and Wernli, 2015] with wind fields from 3-hourly operational ECMWF analyses, interpolated to a regular grid with 0.5° horizontal resolution on the 137 model levels. The trajectory data contains the hourly positions (longitude, latitude, pressure) along the trajectory. To focus on the segments of the trajectories that can potentially be affected by surface aerosol emissions, the trajectories are only included when they were within the model boundary layer. Additionally, removal of aerosol by precipitation, which may remove the signature of upwind aerosol sources via wet deposition, has been considered by removing all the trajectory points before the precipitation event (using a threshold of 0.1 mm h-1). The overall relationship with origin is unchanged by the addition of this filter, which indicates that the results were insensitive to precipitation events.

3 Results and Discussion

3.1 Ice-nucleating particle concentrations within the surface mixed layer

We first present our INP concentrations derived from samples collected on the ship, which was within the surface mixed layer (Figure 2a). The concentrations of INPs measured in the surface mixed layer were highly variable, and ranged from $< 6 \times 10^{-3}$ INP L^{-1} to 2 INP L^{-1} at -15 °C. This resulted in INP activation temperatures ranging from -9 to -30 °C for a concentration of 0.1 INP L^{-1}. This is clearly contrary to what we expected in this remote location based on measurements in other remote oceanic loactions around the world. For example, in the Southern Ocean, INP concentrations are systematically at the low end of what we observe here [McCluskey *et al.*, 2018a; Murray *et al.*, 2021; Welti *et al.*, 2020].

The vast majority of INP measurements made in the Arctic were made on land or at least some distance from the Pole. A summary of these measurements is given Figure 2b. These measurements clearly show that there are strong sources of INP between around 65 to 80°N [Hartmann *et al.*, 2021; Sanchez-Marroquin *et al.*, 2020; Tobo *et al.*, 2019; Wex *et al.*, 2015]. Our measurements demonstrate that the INP concentrations can also sporadically be very high in the pack at the North Pole. Previous measuremets at close to the Noth Pole also reveal substantial variability in INP concentrations active at -15°C [Bigg, 1996; Bigg and Leck, 2001]. Our results indicate sporadically higher concentrations than those results and also demonstrate that the concentration of INP can be in exces of 0.1 L^{-1} at temperatures up to around -10°C. We come back to the question of where these highly active INP come from later in the paper.

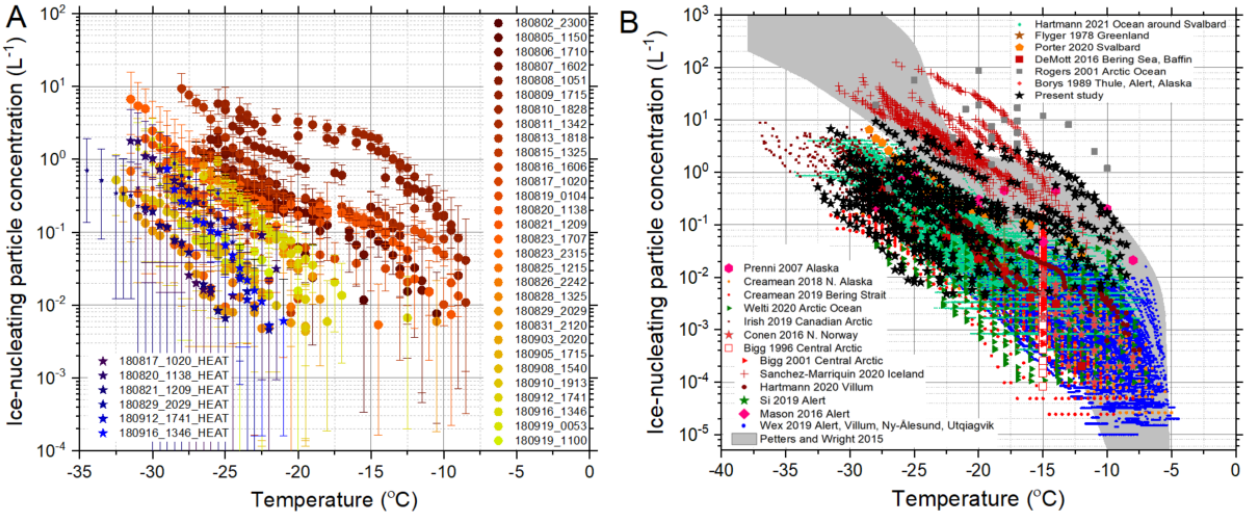

Figure 2. Surface mixed layer INP concentrations throughout the campaign. a) The number of INPs per litre of air sampled was calculated using data from offline droplet freezing experiments, conducted within hours of the samples being taken. The spectra shown in blues represent samples that were heated to close to 100 °C for 30 min. Background values were subtracted from the data. Sampling times varied from 6 h to 3 days and were taken using a heated whole air inlet on the 4th deck (25 m above mean sea level) of the Oden Icebreaker. Temperature uncertainties (not shown) for the droplet freezing experiments were estimated to be ±0.4 °C. The format of the key is YYMMDD_hhmm; these correspond to the start time of the filter sample in the 24-hour time format; the start and end times are in Table S1. b) The data from this study are presented alongside literature data for ground, ship and aircraft-based campaigns around the Arctic [Bigg, 1996; Bigg and Leck, 2001; Borys, 1989; Conen *et al.*, 2016; Creamean *et al.*, 2019; Creamean *et al.*, 2018; DeMott *et al.*, 2016; Flyger and Heidam, 1978; Hartmann *et al.*, 2020b; Hartmann *et al.*, 2021; Irish *et al.*, 2019; Mason *et al.*, 2016; Porter *et al.*, 2020; Prenni *et al.*, 2007; Rogers *et al.*, 2001; Sanchez-

Marroquin *et al.*, 2020; Si *et al.*, 2019; Wex *et al.*, 2019] and a compilation derived from precipitation samples [Petters and Wright, 2015].

It is is remarkable to note that the highest concentrations measured here at the North Pole are as high as the highest INP concentration reported in INP rich environments such as the mid-latitude terrestrial environment [O'Sullivan *et al.*, 2018; Petters and Wright, 2015], despite having much lower aerosol concentrations. Overall, our INP measurements indicate that the INP concentration spectra within the high Arctic surface mixed boundary layer can be extremely variable, perhaps far more variable than anywhere else on Earth.

In order to test for the presence of proteinaceous biological ice-nucleating material, we heated the most active sample suspensions to close to 100 °C [Daily *et al.*, 2021]. The activity of these samples was always reduced, with all of the activity above −20 °C being removed (Figure 2a and Figure S1). Hence, it appears that the most active INPs sampled close to the North Pole were most likely of biological origin. Atmospheric INP at lower latitudes were also found to be heat sensitive [Hartmann *et al.*, 2021]. Together, this indicates that proteinaceous biological INP are important in the Artic.

The time series in Figure 3 shows the temperature at which a concentration of 0.1 INP L^{-1} was measured ($T_{[INP]=0.1}$), and highlights the variability of INP concentrations at the North Pole throughout August and September of 2018. The first peak in ice-nucleating activity was observed during a period in which the ship was breaking ice prior to being moored to an ice-floe (i.e. prior to 16^{th} August). It is reasonable to question whether the very high INP concentrations observed during the ice-breaking period resulted from the ice-breaking itself. Ice-breaking involved frequent backward and forward motions, hence there is the potential for sampling ship emissions (such as ship stack emissions, detailed in the methodology) and aerosol resulting from ice-breaking and the disruption of the sea surface. We manually stopped sampling if there were activities planned that would affect sampling (such as helicopter flights) in addition to using a pollution control system that stopped the flow through the filters when aerosol concentrations increased rapidly in a manner associated with sampling a ship plume, and also when the wind was not from the correct direction (from forward of the ship's superstructure). Despite the precautions taken to eliminate these sources of contamination, we cannot completely exclude the possibility of contamination. However, there was a pause in ice-breaking when a clean air station was established (10^{th} August, we sampled for 6 hours) that coincided with high INP concentrations. At this clean air station the ship was moored facing into the wind, with ship-based aerosol sources aft of the aerosol inlets. Hence, we were confident that sampling of ship pollution and ice-breaking aerosol were eliminated, increasing confidence that these high values in this period were indeed representative of the central Arctic Ocean. In addition, there was also a period of very high ice-nucleating activity a few days after the ice-floe station had been established and the ship was pointing into the wind, demonstrating that there were very active INPs that were not related to ice-breaking or ship

emissions. The time series also highlights two distinct periods. The period up to the 23rd August was characterised by variable but often very high INP concentrations, whereas the period after this was characterised by much lower INP concentrations. We examine the back trajectories associated with these different periods later in the paper.

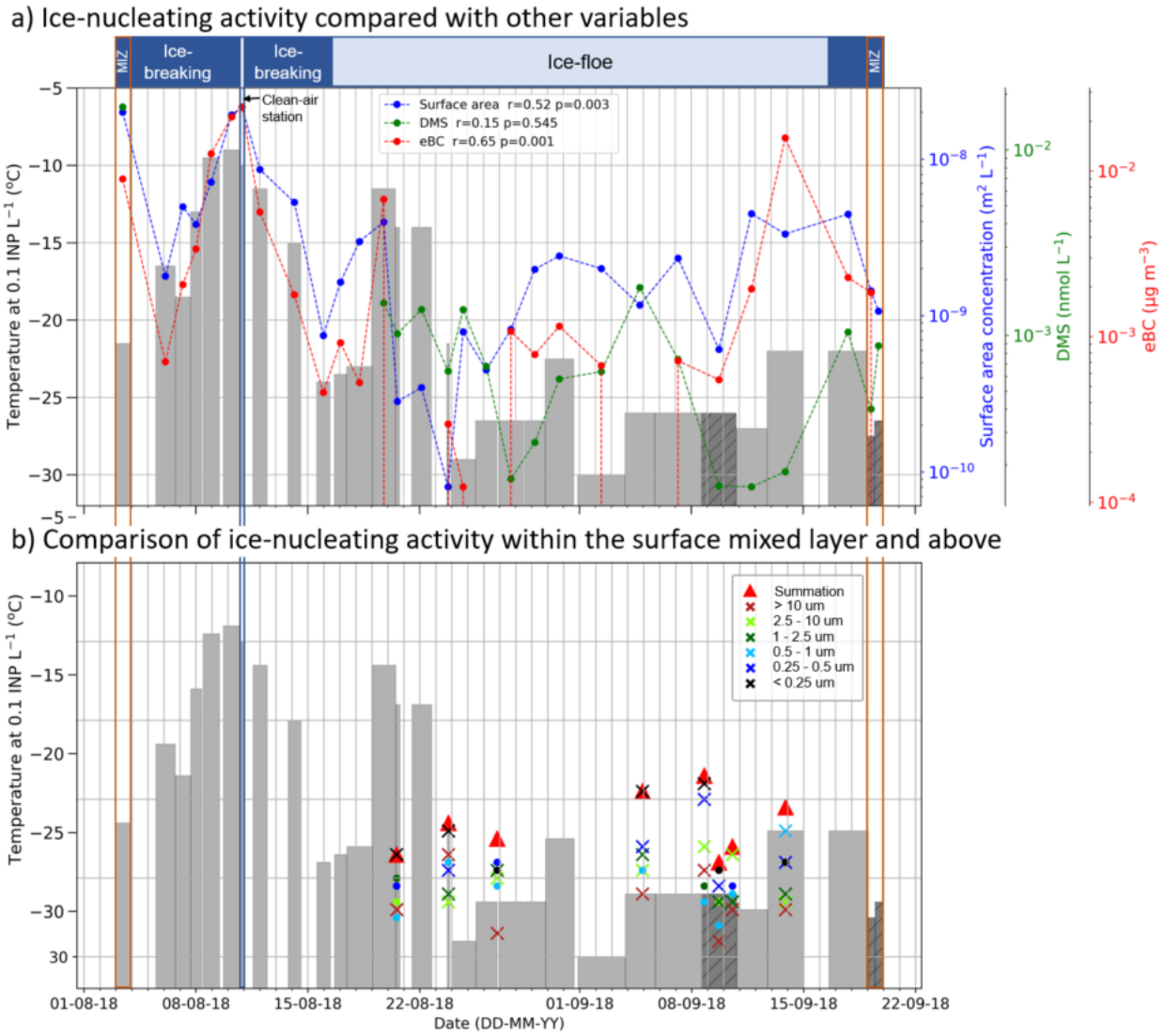

Figure 3. Time series showing the ice-nucleating activity (expressed as the temperature at which a concentration of 0.1 INP L^{-1} ($T_{[INP]=0.1}$)) throughout the campaign alongside DMS, aerosol surface area and eBC. In both panels a and b, the tops of the grey bars represent $T_{[INP]=0.1}$ in the surface mixed layer (i.e. at

ship level, 20 m above mean sea level), with the width of the bar representing the period over which air was sampled. The hatched grey bars are limiting values (where droplet freezing was indistinguishable from the control experiments). a) The time series of the daily average surface area of aerosol per litre (blue), the dimethyl sulfide (DMS) concentration (green) and the equivalent black carbon (eBC) concentrations (red) measured in the aerosol are shown. b) The values of $T_{[\mathrm{INP}]=0.1}$ measured above the surface mixed layer (using the SHARK balloon-borne sampler) are shown against those at taken at ship level (grey bars). The red triangles are the $T_{[\mathrm{INP}]=0.1}$ for the summed INP concentrations across all size categories (comparable to the measurements at ship level), while the crosses indicate the $T_{[\mathrm{INP}]=0.1}$ associated with each size category (circles indicate limiting values. The dates for the respective periods are: MIZ 02/08/18-03/08/18, Clean-air station 10/08/18-11/08/18, Ice-breaking 03/08/18-16/08/18, Ice floe 16/08/18-15/09/18, Ice-breaking 15/09/18-19/09/18, MIZ 19/09/18.

We also show the ice-nucleating activity in the form of ice-active sites per unit surface area (n_s) in Figure 4. The variable n_s provides a means of comparing the activity of aerosol on a per unit surface area basis. It is striking that the activity of the samples in the central Arctic are often much more active than aerosol over the Southern Ocean [McCluskey *et al.*, 2018a] or from the north Atlantic [McCluskey *et al.*, 2018b]. This shows the aerosol in this location are much more ice-active than aerosol in other remote marine environments.

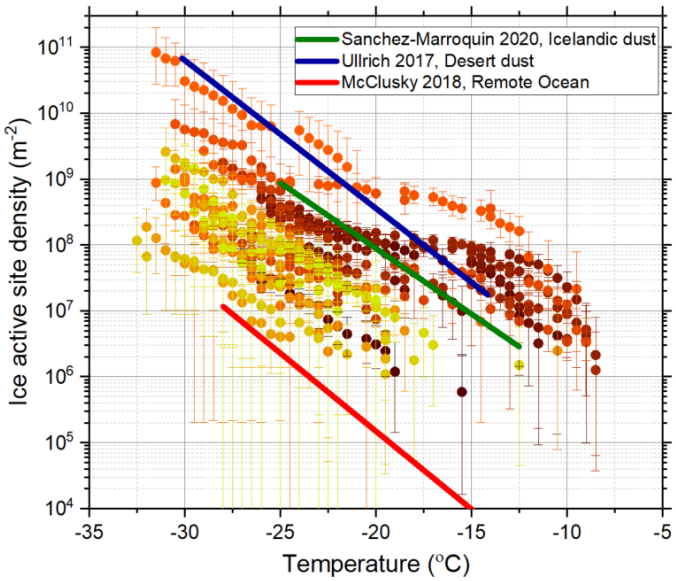

Figure 4. The ice-active site density (ns) for aerosol sampled during the cruise (see Figure 2 for key to colours) compared to n_s parameterisations for desert dust [Ullrich *et al.*, 2017], Icelandic dust [Sanchez-Marroquin *et al.*, 2020] and for aerosol from the North Atlantic [McCluskey *et al.*, 2018b] that is also consistent with values for aerosol over the Southern Ocean [McCluskey *et al.*, 2018a].

3.2 Ice-nucleating particle concentrations above the surface mixed layer

Eight flights with a balloon-borne size-resolved aerosol sampler were conducted while the *Oden* was at the ice station (see SI for flight details). This sampler, known as the selective height aerosol research kit (SHARK) was lofted to a defined height using a tethered balloon system operated from the sea ice and aerosol samples were collected into multiple size bins from below 0.25 µm to 10 µm with an additional stage for particles larger than 10 µm (with a poorly defined upper limit) [Porter *et al.*, 2020]. During these flights, we used a live link to the on-board temperature and humidity measurements to ensure that we sampled above the surface mixed layer and thus in air decoupled from the surface, but within the boundary layer (i.e. in the cloud mixed layer). Hence, the flights occurred at 390 m to 600 m altitude while the ship was on the ice-floe station and we sampled for 3 to 6 hours. In addition, we paused sampling when the RH was more than 80 % to avoid sampling biases associated with hygroscopically swollen aerosol and we also avoided sampling while the SHARK was enveloped in cloud.

Given the surface mixed layer is often decoupled from the rest of the boundary layer, these measurements in principle allow us to compare INP concentrations within and above the surface mixed layer. The values of $T_{[\mathrm{INP}]=0.1}$ are shown in Figure 2b, whereas the INP spectra are shown in Fig SI2 for the INP summed across all the particle size bins. It should be borne in mind that, for practical reasons, the sampling durations on the ship and on the SHARK were not the same, however it is still possible to draw conclusions from this comparison. There is evidence that there are substantial differences between the INP concentrations in the surface mixed layer compared to above it. For example, on the 5^{th} and 8^{th} September the $T_{[\mathrm{INP}]=0.1}$ (summed across all sizes) was around −18 to −19 °C above the surface mixed layer, whereas it was below –26 °C within it. However, on the 20^{th} August, $T_{[\mathrm{INP}]=0.1}$ was around –23 °C above the surface mixed layer, but –14 °C within it. On all three days, radiometer and radiosonde temperature profiles confirmed that the surface mixed layer was decoupled from the rest of the boundary layer (Table S2). In contrast, on 13^{th} September, the surface mixed-layer was mainly coupled and the INP concentrations within and above the surface mixed layer were similar. However, on two other days (23^{rd} August and 10th September) the activity in the surface mixed layer and above were similar even though it was decoupled. Overall, out of the eight SHARK samples collected above the surface mixed layer, there was one SHARK sample that had much lower ice-nucleating activity than that in the surface mixed layer, three samples with higher activity, three with similar activity and one that was ambiguous (due to both samples being close to the baseline of detection). This is consistent with the air at the surface sometimes being coupled to the cloud mixed layer, allowing transport of aerosol throughout the boundary layer, but at other times the measurements at the surface are not representative of those above the surface mixed layer.

The size-resolved INP activity ($T_{[\mathrm{INP}]=0.1}$) is also shown in Figure 3b. In many

locations around the world, supermicron aerosol dominate the INP population [Porter *et al.*, 2020]. However, contrary to what might be expected, the smallest size ranges of < 0.25 m contributed the most INPs on five out of the eight flights, with the 2.5 to 10 m and 0.5 to 1 m bins both contributing the most on one flight each. Inspection of the corresponding INP spectra associated with each bin (Fig SI2) revealeds that the particles < 0.25 m made a pronounced contribution to the INP population on the 23$^{\text{rd}}$ August and the 8$^{\text{th}}$ and 9$^{\text{th}}$ September. The other flights produced data mainly in the baseline for all sizes.

The coarse mode (>2.5 µm diameter) has a relatively short lifetime in the Arctic boundary layer, being removed effectively by wet scavenging processes [Leck and Svensson, 2015]. Hence, it is perhaps not so surprising that the fine mode aerosol (< 0.25 m) appears to be so important in this region for the INP population. While INPs are typically thought of as being the larger particles in a size distribution [Mason *et al.*, 2016; Porter *et al.*, 2020], there are INPs that fall into the < 0.25 m size range that are also very active. For example, film droplet aerosol resulting from wave breaking are produced in a range of sizes centred around 100-200 nm and are often rich in organic material [O'Dowd *et al.*, 2004] that is known to include small ice-nucleating entities [Schnell and Vali, 1975; Wilson *et al.*, 2015]. Alternatively, ice-nucleating macromolecules from terrestrial biological sources internally mixed with other aerosol particles might fall into this size range [O'Sullivan *et al.*, 2016; O'Sullivan *et al.*, 2015; Pummer *et al.*, 2015] and it has been proposed that fungal material, some of which is known to act as an INP [O'Sullivan *et al.*, 2015], can fragment to form nanoparticles [Lawler *et al.*, 2020].

3.3 Correlation between INP concentrations and dimethyl sulfide, equivalent black carbon and aerosol surface area

To investigate possible sources of the INPs we detected over the central Arctic Ocean, we have correlated the ice-nucleating activity of the aerosol with: i) dimethyl sulfide (DMS), a product of marine biological activity, particularly in the marginal ice zone (MIZ, the transitional zone between open sea and dense ice) [Leck and Persson, 1996]; ii) equivalent black carbon (eBC), based on aerosol absorption at 637 nm; and iii) aerosol surface area, derived from size distribution measurements. We present the time series for aerosol particle surface area, DMS and eBC concentrations, as well as the Pearson's r coefficient between ice-nucleating activity and each quantity in Figure 3a.

DMS is found in the marine atmosphere, originating from the metabolites of some marine algae [Leck and Persson, 1996; Lohmann and Leck, 2005]. Hence, the presence of DMS indicates that an air mass has origins in a location rich in biological activity, which may also be expected to correlate with marine biological INP sources. DMS is thought to be relatively short-lived in the atmosphere, with a lifetime on the order of 1-3 days [Kerminen and Leck, 2001; Khan *et al.*, 2016]. Therefore, it is a useful indicator for the interaction of air masses with the MIZ at the outer edge of the pack ice region, and possibly the open leads or melt ponds within the pack ice if they were producing DMS at that time.

The concentration of DMS during the cruise was highest in the outbound 24-hour MIZ station (2^{nd}-3^{rd} August), where the ship was close to open water, but was variable whilst in the pack ice (Figure 3), and remained relatively low in the inbound MIZ station (19^{th} September). The data in Fig. 2 clearly shows that there is no obvious correlation between DMS and INP activity ($r = 0.15$) suggesting that MIZ marine biogenic sources exerted little influence on the measured INP concentrations.

Equivalent BC (eBC) is a quantity derived from aerosol absorption and is the equivalent black carbon mass concentration needed to produce the observed absorption. Other aerosol types such as dust, brown carbon or other organic aerosol might also produce absorption, thus potentially contaminating the small signal we observed. However, absorption by BC is much stronger at 637 nm than other materials, hence the signal is most likely dominated by BC. BC is produced through a range of combustion processes, including biomass burning, wildfires and fossil fuel combustion, which are all remote from the central Arctic. Other potential contributors to the absorption signal, such as dust or brown carbon are also remote from the central Arctic. Rigorous procedures were in place to ensure that BC (and other aerosol) from the ship stack did not affect measurements (see methods for details). Therefore, eBC is used here as an indicator of long-range transport. The literature indicates that BC is a relatively ineffective ice nucleator under mixed-phase cloud conditions [Adams *et al.*, 2020; Chen *et al.*, 2018; Schill *et al.*, 2020; Vergara-Temprado *et al.*, 2018a], hence we would not necessarily interpret a positive correlation as an indication of ice nucleation by BC. However, combustion processes are thought to be a source of ice-nucleating aerosol, even if BC itself is not an effective INP [Barry *et al.*, 2021; Jahn *et al.*, 2020; Umo *et al.*, 2015]. Thus, a correlation between BC and INP concentrations would indicate that aerosol particles transported along with BC from outside the central Arctic Ocean nucleate ice. Wildfires around the Arctic are a potential source of BC, and we note that during the cruise there were persistent Siberian wildfires, as can be seen using the NASA Worldview satellite imagery tool (e.g. clear skies on the 14^{th} August reveal widespread fires; https://worldview.earthdata.nasa.gov/). There is also industry, shipping and mining along the Arctic coast of Russia, but the role of gas flaring is thought to be a particularly important source of BC [Stohl *et al.*, 2013].

The overall correlation between eBC and INP concentration ($r = 0.65$) is much stronger than for DMS. In fact, the eBC concentration appears to track the INP concentration in Figure 3 until the 27^{th} August, after which the INP concentrations stay relatively low whilst the eBC remains highly variable. The decoupling of eBC and INPs later in the campaign indicates that distant sources of BC are not always related to distant sources of highly active INPs.

The surface area concentration of the bulk aerosol follows a similar trend to the eBC concentrations, but with a slightly weaker correlation with the INP activity ($r = 0.52$). This indicates that the variability in INP concentrations at the North Pole is not simply driven by aerosol surface area, rather that some

specific component(s) of the aerosol population are ice-active and these particles are most likely associated with specific sources at latitudes further south than the MIZ.

3.4 Trajectory analysis of aerosol collected in the central Arctic

Backward trajectories from the sampling location near the surface are presented in Figure 5a. We only show points that are in in the boundary layer and for a maximum of 7 days. The figure shows a clear relationship between the origin of the aerosol – considered here as the boundary layer points along the trajectories - and the measured INP concentrations. The origin of the air with the most active INPs is around the Russian Arctic coast including the Barents, Kara and Laptev Seas. Out of the 30 filter runs, those filters with the highest INP activity (the top 20 % of filters; -9 °C $T_{[\mathrm{INP}]=0.1} > -13.5$ °C) sampled air masses originating over the Barents and Kara Seas. The next seven highest (23 %; -13.5 °C $T_{[\mathrm{INP}]=0.1} > -22$ °C) filters, in terms of INP activity, sampled air originating from over the Laptev and East Siberian Seas. The next six filters with lower INP activity (20 % of filters; -22 °C $T_{[\mathrm{INP}]=0.1} > -25$ °C) sampled air that originated off the eastern coast of Greenland from over both the pack ice and open ocean. The 11 filters with the lowest INP activity (bottom 37 % of filters; -25 °C $T_{[\mathrm{INP}]=0.1}$ -30 °C) all sampled air which mostly originated from the pack ice adjacent to North America (also see Figure SI3).

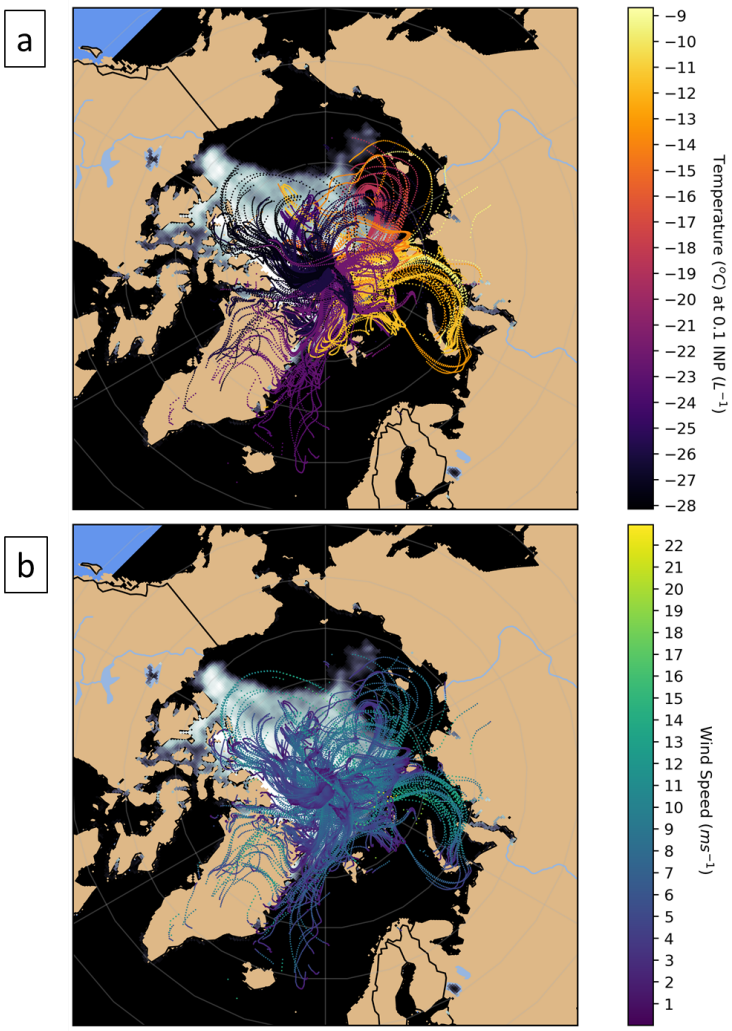

Figure 5. Backward trajectories over 7 days, starting at the ship location, for the INP samples taken throughout the campaign. Trajectories were launched every hour during the sampling period, and each point represents an hour in time along the back trajectory. The starting height for the trajectories was 32 m above mean sea level. Any points along the trajectories which were above the model boundary layer were removed, and any points along the trajectory that preceded precipitation events (>0.1 mm h-1) were removed. Hence, any potential sources of INP in the boundary layer are neglected if they occurred prior to a precipitation event (we assume precipitation removes INPs) a) The colour of the trajectories represents the temperature at which 0.1 INP L-1 was measured for that sampling period. b) The colour of the trajectories represents

the wind speed for each point along the trajectory. The sea ice extent is from the NASA National Snow and Ice Data Center [Maslanik and Stroeve, 1999].

It is striking that the trajectories with the lowest INP concentrations spent most of the preceding seven days over the pack ice and to some extent over the MIZ. These results indicate that, during this campaign, open leads, sea ice and the MIZ were weak sources of INPs, in conflict with previous suggestions [Bigg and Leck, 2001; Hartmann *et al.*, 2020a].

The highest ice-nucleating activities from sampled aerosol originating along the Russian coast were also correlated with high wind speeds along the trajectories, i.e., in the region of the aerosol origin (Figure 5b). This, together with the heat tests and INP size information, point to a wind-driven marine biological source of INPs associated with organic-rich film droplet sea spray aerosol. There were trajectories with high wind speeds over the North American continent, the pack ice and the coast of Greenland, but the ice-nucleating activity for these was not greatly enhanced. Hence, our results are consistent with a strong source of highly active INPs in the coastal marine waters of northern Russia which were aerosolised during windy conditions. Marine waters elsewhere in the world are thought to produce aerosol with relatively low ice nucleating activities [Vergara-Temprado *et al.*, 2017] (see Figure 4); however, our results suggest that the shallow seas off the Russian coast might be relatively strong sources of highly active INPs. Composition analysis of the aerosol during the peak ice nucleation activity on the 11^{th} August and the 19^{th} August is consistent with a marine source for many of these aerosol particles (samples were rich in Na, Cl and sulfate; see Table S3). Hence, the question is why the aerosol from near the coast of Russia is so much more active than aerosol derived from other marine locations such as the North Atlantic and the Southern Ocean [McCluskey *et al.*, 2018a; McCluskey *et al.*, 2018b].

A major difference to the North Atlantic and Southern Ocean is that the shallow seas off the coast of northern Russia are strongly influenced by riverine input from Russia that is rich in organic material, silt and nutrients [Ahmed *et al.*, 2020; Juhls *et al.*, 2019]. In fact, much of the dissolved organic matter in the Arctic Ocean is derived from river input [Juhls *et al.*, 2019] and the discharge of these rivers (and the amount of dissolved organic carbon flowing in the sea via rivers) is increasing [Ahmed *et al.*, 2020; Juhls *et al.*, 2020]. It has been shown that melting permafrost, which is known to enter river water [Juhls *et al.*, 2020], harbors copious quantities of warm temperature INPs [Creamean *et al.*, 2020]. Hence, it is possible that the highest INP concentrations we detected at the North Pole were derived from marine waters rich in terrestrially derived ancient biological INPs. Alternatively, ocean biology fertilised by the nutrient rich waters on the continental shelf may produce more INPs than are present in remote marine locations. A measurement campaign to quantify the INP content of the waters off the coast of Russia is clearly required.

Some of the back trajectories that had the highest INP concentrations passed over islands in the Barents and Kara Seas, including Svalbard, Franz Josef

Land, Novaya Zemlya and Severnaya Zemlya. Many of these locations have been identified as poorly defined dust sources [Bullard *et al.*, 2016] and dust from Svalbard has been shown to contain biological ice-nucleating materials [Tobo *et al.*, 2019]. However, in a further analysis of the back trajectory data (Figure 6), we find that there was little to no correlation with time spent over land, whereas the ice-nucleating activity increased with the time the air parcels spent over open ocean. This implies that the sources of INP were associated with the marine environment. Having said this, we cannot rule out relatively small island point sources being important sources of INPs.

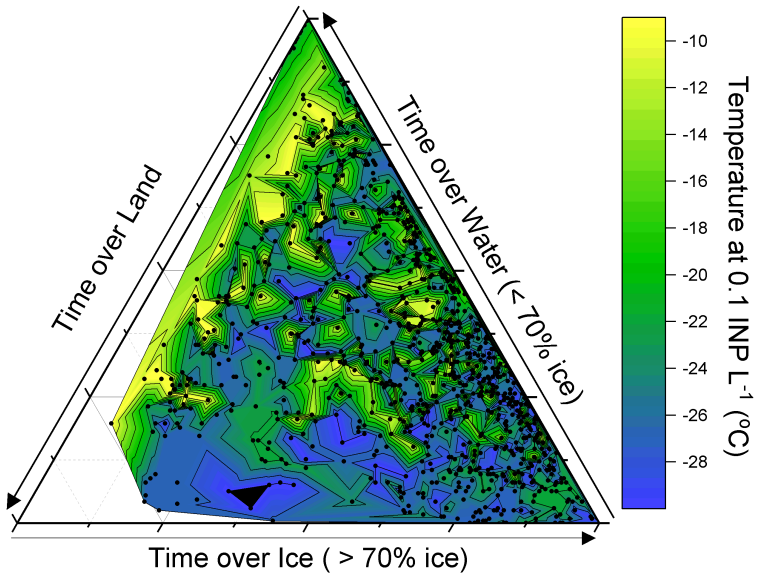

Figure 6. The time that air masses spent over water, ice and land derived using back trajectory analysis. The colour represents the temperature at which an INP concentration of 0.1 INP L-1 was reached. The black dots correspond to individual trajectories. The same filters as in Figure 5 were applied.

Overall, the evidence indicates that there is a strong source of biogenic INPs in the Barents, Kara and Laptev Seas off the Russian coast that can be sporadically transported to the central Arctic Ocean. There was high wind along the trajectories off the Russian coast which would be consistent with both the production of INPs in sea spray from these organic-rich seas or dust combined with terrestrial biogenic material from the various islands in this region. We also note that a recent study found that very active INP were produced in the Chukchi sea (near Alaska) under high wave conditions [Inoue *et al.*, 2021]. Furthermore,

we note that these potential sources are sensitive to a changing climate, with river discharge, permafrost melt and organic matter input into the ocean from the major Russian rivers increasing in a warmer world [Jahn *et al.*, 2020]. In addition, the removal of ice from the Arctic could also expose marine and terrestrial sources of ice-nucleating aerosol around the Arctic Ocean, where they can then be aerosolised by the action of wind [Schmale *et al.*, 2021].

3.5 Implications for ice production in boundary layer central Arctic mixed-phase clouds

In this section, we assess whether the measured INP concentrations are high enough to initiate a transition from liquid-dominated clouds to ice-dominated clouds. Model simulations indicate that on the order of one ice crystal per litre of air is required to remove the bulk of liquid water from an Arctic cloud, whereas lower concentrations still reduce the liquid water path [Stevens *et al.*, 2018; Vergara-Temprado *et al.*, 2018b]. Hence, we use our INP measurements, both at ship level in the surface mixed layer and those from the SHARK in the cloud mixed layer, to estimate the concentration of INPs that become active at the ambient temperature of the atmosphere ($[\mathrm{INP}]_{\mathrm{ambient}}$).

The quantity $[\mathrm{INP}]_{\mathrm{ambient}}$ combines the atmospheric temperature profiles from radiosonde measurements with the corresponding INP spectra as a rough indicator of primary ice crystal production. This is a crude analysis and a full cloud model would be required to represent ice crystal formation and sedimentation as well as INP recycling and latent heat release, to predict ice crystal concentration given an INP spectrum, but it does give an indication of what the measured INP spectra might mean for primary ice production in clouds. The highest $[\mathrm{INP}]_{\mathrm{ambient}}$ will be at the cold points, i.e. the top of the surface mixed layer and the top of the boundary layer (top of the cloud mixed layer), hence this is where we focus this analysis. We assume that ship level measurements are representative of the $[\mathrm{INP}]_{\mathrm{ambient}}$ at the top of the surface mixed layer and those from SHARK are representative of the top of the boundary layer, the assumption being that these respective layers are individually well-mixed.

The temperature minima within the main boundary layer were determined from radiosonde profiles, which were made every 6 hours throughout the entire cruise [Vüllers *et al.*, 2021]. The calculated $[\mathrm{INP}]_{\mathrm{ambient}}$ for the duration of the cruise are shown in Figure 7, where the minimum temperature at the top of the cloud mixed layer (main boundary layer) and the top of the surface mixed layer is indicated for each filter period. In many cases, the temperature of the atmosphere was higher than the highest INP measurement, hence we are only able to report upper limits to $[\mathrm{INP}]_{\mathrm{ambient}}$ in these cases.

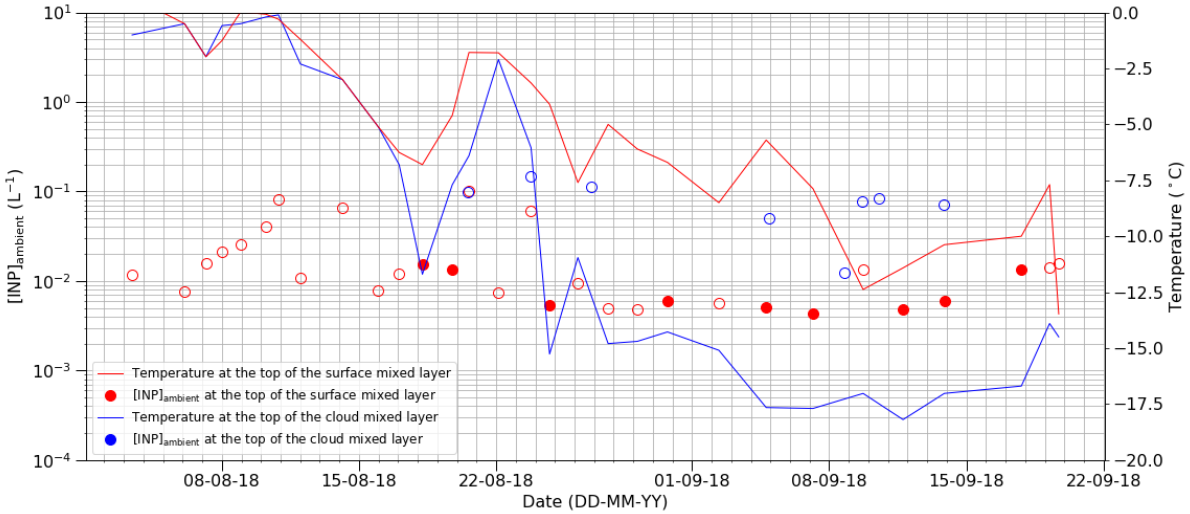

Figure 7. Time series showing [INP]ambient for both the measurements taken at ship height (within the surface mixed layer) and using the SHARK (within the cloud mixed layer). The temperature of the mixed layers is shown alongside these measurements. When an INP spectrum did not extend to the atmospheric temperature (i.e. where the highest temperature at which an INP concentration was reported was below the atmospheric temperature), the [INP] value associated with the highest temperature is provided as an upper limit to [INP]ambient (open symbols).

Generally, $[INP]_{ambient}$ was typically below about 0.1 INP L^{-1} at the top of both the surface mixed layer and the top of the cloud mixed layer. The periods of high INP concentration before the 23rd August coincide with periods of higher ambient inversion temperatures, whereas later in the campaign the opposite is the case, which results in a relatively invariant $[INP]_{ambient}$. Whether this is a coincidence, or if INP concentrations are correlated with ambient temperature, is unclear from this limited dataset. However, there may be a physical mechanism behind this apparent correlation. Transport of air to the North Pole from sources further afield will likely result in multiple cycles of cloud formation and dissipation in any one air mass, hence INP active at above the ambient temperature of those clouds will likely activate and be removed via precipitation (bearing in mind that the majority of INPs relevant for mixed-phase clouds only activate to ice in the presence of water droplets [Murray *et al.*, 2012]). While transport through the boundary layer likely removes INP active above the lowest temperatures experienced by an air parcel, further cooling subsequent to the measurement time will lead to primary ice production. Hence, the air masses sampled before the 23rd August with high INP concentrations have a great potential for primary ice production if or when these air masses become colder downwind of the sampling location. In the absence of local sources, the INP spectrum over the central Arctic Ocean must therefore be determined by a com-

bination of the characteristics of the upwind sources and the cloud temperature that these air parcels experience on transport.

The relatively low $[INP]_{ambient}$ values suggest that clouds not influenced by seeding from above should be mixed-phase, i.e. contain a substantial proportion of liquid water with some ice crystals. Observations of the phase of clouds during this campaign are discussed in [Vüllers *et al.*, 2021]. Overall, the fraction of single layer clouds (where seeding from above is unlikely) throughout the troposphere that were mixed-phase was relatively constant throughout the campaign: mixed phase frequency ~20-30 %, ice cloud frequency ~10-30 % and liquid clouds were generally infrequent; see Figure 15 in [Vüllers *et al.*, 2021]), despite the strong decrease in temperature during the campaign. Similarly, in the bottom few kilometres the frequency of occurrence of mixed-phase clouds were often between 25% and 50%, with ice clouds about half as frequent and liquid only clouds much less frequent throughout the campaign. Overall, the observations of cloud phase for single-layer clouds reported by [Vüllers *et al.*, 2021] are qualitatively consistent with our relatively invariant $[INP]_{ambient}$ measurements.

Clouds were multi-layered around 50 % of the time, with many situations identified where seeding of ice from higher, colder clouds into lower clouds might occur. Clouds were regularly observed up to around 8 or 9 km, where temperatures [Vüllers *et al.*, 2021] were low enough for homogeneous freezing [Herbert *et al.*, 2015], and frequently occurred in the mid-troposphere where heterogeneous nucleation on INPs was most likely important. These higher clouds were often in the free troposphere where our boundary layer INP measurements are not necessarily relevant. Indeed, aerosol and INPs in the free troposphere may have different sources to those in the boundary layer. In order to obtain a more complete picture of primary ice production in clouds in the central Arctic, INP measurements in the free troposphere in this region would be needed and should be a target of future campaigns.

4 Summary and conclusions

Arctic mixed-phase and supercooled clouds play a crucial role in Arctic climate, but the processes that dictate their characteristics are poorly understood. Here, we show that INP concentrations at 88 - 90°N are extremely variable, and throughout the MOCCHA campaign between the 1st of August 2018 and the 18th of September 2018 the temperature at which 0.1 INP L^{-1} was reached varied between −9 °C and −30 °C. The highest 20% of observed INP activity is related to air masses originating in the ice-free ocean environment off the Russian coast, while the lowest 37 % of observations related to air masses which originated and circled over the pack ice north of Canada for most of the 7-day back trajectory. Trajectories of air with intermediate INP activity also originated over the ice-free ocean. These results indicate a strong dependence of the measured INP concentration on the origin of the air with pack ice, open leads, and the MIZ apparently being weak sources of INP, whereas ice-free oceans, especially those near the Russian coast when wind speeds were high, were a significant source.

The heat sensitivity of the most active INPs indicates the INP to be proteinaceous, biogenic origin. This, together with the trajectory analysis, indicates that there are strong biogenic sources of INP in the shallow seas over the Russian continental shelf. The ice-nucleating activity of the aerosol at the North Pole derived from off the coast of Russia is much greater than that for sea spray aerosol in remote oceans (such as the Southern Ocean [McCluskey *et al.*, 2018a] or the North Atlantic [McCluskey *et al.*, 2018b]). This may indicate the marine waters off Russia are very rich in ice-nucleating material, perhaps related to the substantial riverine input, or alternatively the islands in this region may be sources of biogenic INPs. More work is needed to define what the key sources are along the Russian coast and to see if similar sources exist elsewhere around the Arctic and Antarctic.

By making measurements of INP spectra both above and within the surface mixed layer of decoupled boundary layers, we found that surface measurements were often not representative of the INPs in the cloud mixed layer. Hence, measurements at altitude, within the cloud mixed layer, are necessary in order to define primary ice production in Arctic mixed-phase clouds. In addition, our measurements allowed us to estimate the INP concentration active at the temperature of the top of the surface mixed layer and also at the top of the boundary layer. This revealed that, despite massive variability in INP spectra, the INP concentration at ambient temperature was typically less than 0.1 L^{-1}, which is consistent with remote sensing observations that indicate the persistence of mixed-phase clouds (in the absence of seeding of ice from above). We also recommend future studies focus on INP measurements throughout the free troposphere where primary ice production may lead to seeding of ice in lower level clouds.

Overall, it is striking that INP concentrations at the summertime North Pole vary from some of the lowest measured anywhere in the world, to as high as the highest INP concentrations in terrestrial locations rich in biological INPs such as in the UK [O'Sullivan *et al.*, 2018]. Since these INPs are transported from the seas off the Russian coast, they may be sensitive to changes in climate. In particular, reduced sea, land ice and permafrost may open up more sources for more of the year around the Arctic, which may increase the future strength (and may already have done so) of the sources of INPs that are important for mixed-phase clouds in the central Arctic. More work needs to be undertaken to understand how climate change may affect INP sources around the periphery of the Arctic and how this may influence Arctic clouds and feedback on Arctic climate.

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Competing interests

All other authors declare they have no competing interests.

Data Availability Statement

All data will be made available via the Research Data Leeds Repository and the Bolin Centre Database.

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