Ship-based measurements of ice nuclei concentrations over the Arctic, Atlantic, Pacific and Southern Ocean

André Welti[1,6], E. Keith Bigg[5], Paul J. DeMott[3], Xianda Gong[1], Markus Hartmann[1], Mike Harvey[8], Silvia Henning[1], Paul Herenz[1], Thomas C. J. Hill[3], Blake Hornblow[9], Caroline Leck[4], Mareike Löffler[1,7], Christina S. McCluskey[3,10], Anne Marie Rauker[3], Julia Schmale[2,11], Christian Tatzelt[1], Manuela van Pinxteren[1], and Frank Stratmann[1]

[1]Leibniz-Institute for Tropospheric Research (TROPOS), Leipzig, Germany
[2]Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, Villigen, Switzerland
[3]Department of Atmospheric Science, Colorado State University, Fort Collins, CO, USA
[4]Department of Meteorology, Stockholm University, Stockholm, Sweden
[5]Elanora Heights NSW, Australia
[6]Finnish Meteorological Institute, Helsinki, Finland
[7]Deutscher Wetterdienst, Centre for Agrometeorological Research, Braunschweig, Germany
[8]National Institute for Water and Atmospheric Research (NIWA), Wellington, New Zealand
[9]NIWA Blake Ambassador (Blakenz.org), Wellington, New Zealand
[10]National Center for Atmospheric Research, Boulder, CO, USA
[11]School of Architecture, Civil and Environmental Engineering, ole Polytechnique Frale de Lausanne, Switzerland

Correspondence: A. Welti (andre.welti@fmi.fi)

Abstract.

Ambient concentrations of ice-forming particles measured during ship expeditions are collected and summarized with the aim of establishing a spatial distribution and variability of ice nuclei in oceanic regions. The presented data from literature and previously unpublished data from over 23 months of ship-based measurements stretch from the Arctic to the Southern Ocean and include a circumnavigation of Antarctica. In comparison to continental observations, ship-based measurements of ambient ice nuclei show one to two order of magnitude lower mean concentrations. To quantify the geographical variability in oceanic areas, the concentration range of potential ice nuclei in different climate zones is analysed by meridionally dividing the expedition tracks into: tropical, temperate and polar climate zones. We find that concentrations of ice nuclei in these meridional zones follow temperature spectra with similar slope, but vary in absolute concentration. Typically, the frequency with which specific concentrations of ice nuclei are observed at a certain temperature follows a log-normal distribution. A consequence of the log-normal distribution is that the mean concentration is higher than the most frequently measured concentration. Finally, the potential contribution of ship exhaust to the measured ice nuclei concentration on board of research-vessels is analysed as function of temperature. We find a sharp onset of the influence at approximately -36 °C, but none at warmer temperatures that could bias ship-based measurements.

1 Introduction

A small fraction of atmospheric aerosol particles possesses properties that induce the nucleation of ice. Herein the term ice nuclei (IN) is used to describe these, without specificity about their nature, equivalent to the proposed term ice nucleating particles (Vali et al., 2015). The concentration of IN active at a certain temperature affects the extent of ice crystal formation in clouds. Thereby, IN have an effect on precipitation formation, cloud optical properties and cloud persistence (e.g. DeMott et al., 2010). Stratiform clouds are more affected by the IN concentration than convective clouds because of differences in cloud water content and dynamics (Zeng et al., 2009). Since the ratio between stratiform and convective cloud amount changes meridionally, with more stratiform clouds towards higher latitudes, the effect of IN concentration on cloud properties increases from the tropical to polar regions (Zeng et al., 2009). The most important ice nucleation mechanism for mixed-phase clouds is immersion freezing. The temperature range of immersion freezing, in which IN are first immersed in supercooled droplets before triggering ice formation, is limited at the lower end by the onset temperature of homogeneous ice nucleation (-38 °C) below which droplets freeze without the necessity for catalysing IN. At the high end, exceptionally efficient, biological IN are known to nucleate ice at temperatures as high as -1.3 °C (Schnell and Vali, 1972) whereas, for example, sub-micron desert dust IN typically show high activity only below -25 °C (Boose et al., 2016). After the onset of immersion freezing, the glaciation of mixed-phase clouds can be enhanced by secondary ice formation, a cascade process where supercooled droplets freeze and shatter on contact with ice crystals (Mason and Maybank, 1960) or by ejection of ice splinters during the formation of rime on graupel (Hallett and Mossop, 1974).

To describe the atmospheric IN population comprehensively, IN concentrations in the entire temperature range of immersion freezing (0 °C to -38 °C), their temporal fluctuation and geographical variation should be considered. The first summary and parametrization of the change in IN concentration as a function of temperature was given by Fletcher (1962), based on a collection of ambient measurements available at the time. Even before Fletchers parametrization, efforts had been made to quantify the concentration of atmospheric IN dependent upon air mass origin and weather conditions. For example, Findeisen and Schulz (1944) determined an IN temperature spectrum for Central Europe on the basis of 8 years of aircraft and ground based observations. The spatially most comprehensive measurements so far have been achieved by coordinated, ground-based measurements (e.g. Bigg and Stevenson, 1970), aircraft campaigns (e.g. DeMott et al., 2010) and ship-based observations in the Southern Ocean between Australia and Antarctica (Bigg, 1973). As 71% of earth's surface is covered by oceans, ship-based observations offer an effective way to cover large areas. The Bigg (1973) data set, averaging three years of observations (shown in Fig. 1), has been used as a reference on which modelled IN concentrations have been validated (e.g. Vergara-Temprado et al., 2017).

We present a summary of ship-based measurements from the literature and extend this data set with unpublished data dating back as far as 1996 and measurements from several recent ship expeditions. Definition of expedition acronyms and a description of expedition objectives can be found in Appendix A. The recent data-sets were obtained during expeditions in the Arctic (PS106), the Southern Ocean (ACE) and during Atlantic transects (ACE, PS95 and PS79). Previously unpublished -15 °C data

include observations from the Pacific (SHIPPO, ACAPEX), Southern Ocean (Tan1502) and central Arctic Ocean (AOE-96, AOE-2001).

2 Sampling and measurement

Ship-based measurements of IN concentration record the abundance of potential IN in the marine boundary layer, some meters above sea level. Much of available literature data are limited to IN concentrations at -15 °C or -20 °C, because of instrumental limitations and a focus on the influence of IN abundance on precipitation formation. During recent expeditions, emphasis was put on collecting data covering a broad temperature range (0 °C to -38 °C), because of a shift in focus to the cloud radiative effect and the realization that already low IN concentrations can have a large effect by triggering secondary ice formation (e.g. Sullivan et al., 2018). Additionally to the importance for cloud microphysics, observations over a broader temperature range allow the construction of IN temperature spectra, providing information on the abundance of potential IN sources from their slope (e.g. Welti et al., 2018).

Two measurement techniques are used for recent ship-based measurements. First, continuous flow diffusion chambers (CFDCs), e.g., CSU CFDC measuring during CAPRICORN (McCluskey et al., 2018a) or SPIN (SPectrometer for Ice Nuclei, DMT, see Garimella et al., 2016, for technical details) measuring during PS95 and PS106. CFDCs cover the low temperature range from approximately -38 °C to -24 °C. Secondly, drop freezing techniques, using aerosol collected on filters during expeditions. Drop freezing techniques typically cover the temperature range from -26 °C to 0 °C.

For the new data sets in the Arctic, Atlantic and Southern Ocean (marked as circles in Fig. 1), sampling was conducted on board the research vessels Polarstern (expeditions indicated with PS) and Academic Tryoshnikov (during ACE). For a description of the ACE and PS106 (PASCAL) expeditions we refer to Schmale et al. (2019) and Wendisch et al. (2018), respectively. On board both vessels we used the upper deck (monkey bridge), about 30 m above the ocean surface, to install sampling devices. SPIN sampled from within a container through a whole-air inlet system with subsequent drying of the aerosol. The SPIN measurement strategy was to sample half hourly intervals at a constant temperature (selected between -38 °C and -24 °C) and constant relative humidity (85%, 95% or 103% with respect to water). Each sampling condition was repeated three times within 24 h. This allowed measurements of IN concentrations at different temperatures and relative humidity with a high spatial coverage. Measurements above water saturation, representing the immersion freezing mode, are reported here. Filter samples were collected in 8 h intervals using a Low Volume Aerosol Sampler (DPA14, Digitel) with PM10 inlet during ACE and PS106. During PS79 and PS95, 24 h filter samples were collected ~~during PS79 and PS95~~ using a High Volume Aerosol Sampler (DHA-80, Digitel) with PM1 inlet. The latter used 150 mm quartz fiber filters, from each of which 103 sub-samples were extracted. IN concentration was examined in a drop freezing array setup, similar to the technique used by Conen et al. (2012) and previously described in Welti et al. (2018). With the Low Volume Sampler, 8 h samples were collected on 47 mm-diameter Nuclepore polycarbonate membranes (Whatman) with a pore size of 0.2 µm. Field blanks, which had undergone the same handling except for aerosol collection, were collected regularly to determine the filter background. Filters were stored in a -20 °C cold room on board and kept cool during transport home before storage at -20 °C until analysis. To determine IN concentrations, membrane

filters are immersed in 10 $m\ell$ ultra pure water to extract collected particles and 96 aliquots (each 50 $\mu\ell$) of the sample solution were subsequently examined in a drop freezing assay. IN concentrations are estimated from the number of frozen aliquots at a specific temperature according to Vali (1971). Data at -15 °C ~~data~~ from AOE, TAN1502, ACAPEX, SHIPPO were also obtained from filter samples. TAN1502, ACAPEX and SHIPPO filters were analysed using the Colorado State University Ice Spectrometer (IS) (Hill et al., 2016). Collection (open-faced, 0.2 µm filters) and processing protocols for the IS are as described in McCluskey et al. (2018a, b). During these expeditions IN concentrations have been measured over a broader temperature range, and results will be reported elsewhere. A discussion of the membrane filter technique used to determine IN concentrations during AOE can be found in Bigg (1990). Tab. A1 provides an overview on the location, on-board equipment, sampling time intervals and sampled air volume during expeditions from which data are included. An analysis showing that contamination from ship exhaust had no influence on ship-based IN concentration measurements during PS106 and ACE, is presented in Sec. 6 and Appx. C.

3 Worldwide coverage of maritime observations

Fig. 1 summarizes all available data at -15±1 °C from the literature and recent expeditions. This temperature is chosen to include observations from the literature (e.g. Bigg, 1973; Schnell, 1977; Borys, 1983; Rosinski et al., 1986, 1988, 1995), for some of which only -15 °C data are available. Literature data covers the Southern Ocean, North Atlantic off the coast of Nova Scotia, Greenland Sea, Pacific Ocean, Gulf of Mexico and the East China Sea. Note that although the spatial coverage of the literature data is broad, the number of data points reported is low and often only average values were given. Recent expeditions added data from the Atlantic, Pacific, Southern and Arctic Ocean. So far unexplored in terms of ship-based measurements of IN concentration remain e.g., the Indian Ocean, the Southeast Pacific or the Mediterranean Sea.

4 Geographic variability

Regional variations of IN concentrations on the continents coincide with strong local sources (e.g. Isono et al., 1959; DeMott et al., 2003) or IN generating events (e.g. McCluskey et al., 2014; Suski et al., 2018). In remote maritime environments the sources of the most common IN type by concentration can be long range transport of mineral dust (e.g. Bigg, 1973) or oceans themselves. Oceans have been identified as a source of IN (e.g. by Brier and Kline, 1959; Schnell and Vali, 1975; Ickes et al., 2020) able to produce IN active in a broad temperature range (DeMott et al., 2016). Based on numerical simulations, Burrows et al. (2013) suggested that in remote oceanic areas, the absence of mineral dust means that maritime IN determine the impact of heterogeneous ice formation on clouds. McCluskey et al. (2019) reported that the contribution of mineral dust or maritime aerosol changes depending on season and altitude, with strong variations from day to day. During AOE-91 and AOE-96, Bigg and Leck (2001) found evidence for a microbiological, marine source of Arctic IN, in a strong seasonal dependence of IN concentration and a correlation with the concentration of dimethyl sulfide (DMS), a metabolite in some marine algae. They observed IN concentrations to start decreasing very rapidly after ~~mid September~~ mid-September to virtually zero by late

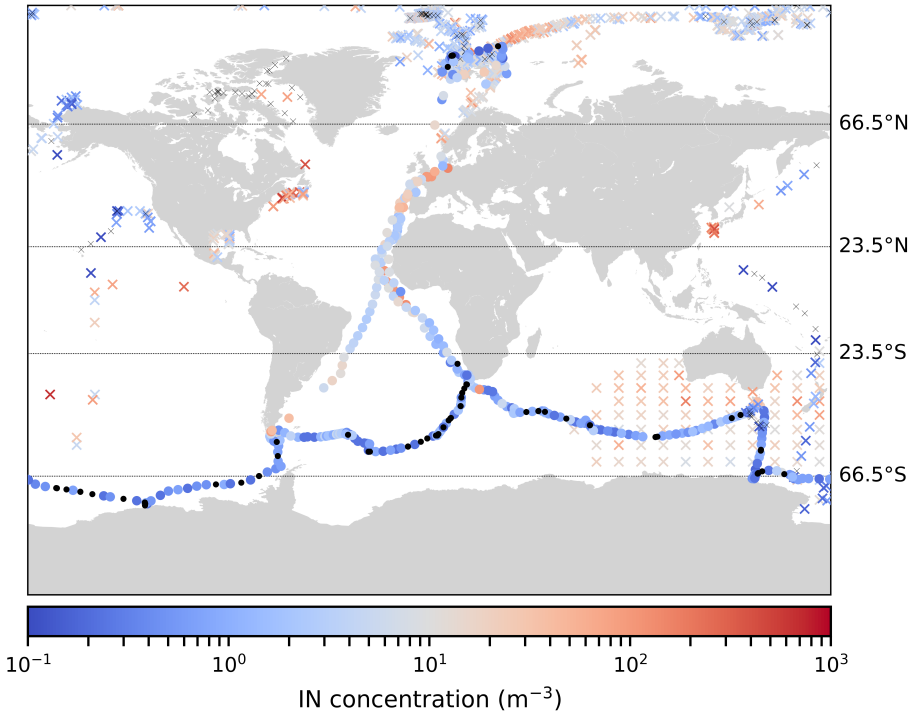

Figure 1. World map showing the locations where IN concentration data at -15 °C are available from ship-based measurements. Literature data from ~~Bigg (1973); Schnell (1977); Borys (1983); Rosinski et al. (1986, 1988, 1995); DeMott et al. (2016); McCluskey et al. (2018a); Creamean et al~~ Bigg (1973); Schnell (1977); Borys (1983); Rosinski et al. (1986, 1988, 1995); DeMott et al. (2016); McCluskey et al. (2018a); Si et al. (2018); Irish et a and unpublished data from AOE, Tan1502, ACAPEX and SHIPPO are marked as crosses. Data from ACE, PS79, PS95 and PS106 are marked as circles. The symbols are colour coded according to the colour bar below the map, indicating the measured IN concentrations. Locations where the observed IN concentration was below the detection limit (< ~~0.1~~sample volume^{-1}, see Tab. A1) are marked in black. Climate zones are divided by horizontal lines, their fraction of the earth surface area can be found in Tab. B1.

October.

Due to the different conditions (wind speed, temperature) and location (e.g. proximity to desert) of different parts of the world's seas and oceans, meridional variations in the IN temperature spectrum (Sec. 5) could be expected. Effects of atmospheric transport and local sources include, for example, the westward transport of large amounts of dust from the Sahara desert, higher biological productivity along coasts and in higher latitudes, and furious storm tracks or sea ice cover that promote or prevent wave-derived aerosol generation.

A comparison of continental and maritime IN concentrations observed at -15 °C, from the surface-based measurements in the study of Bigg and Stevenson (1970) and meridional averages of ship-based data from Fig. 1, is shown in Fig. 2. Averaging zones of Fig. 2(b) are indicated in Fig. 1 and defined in Tab. B1. Concentrations over the Ocean are lower than the reported

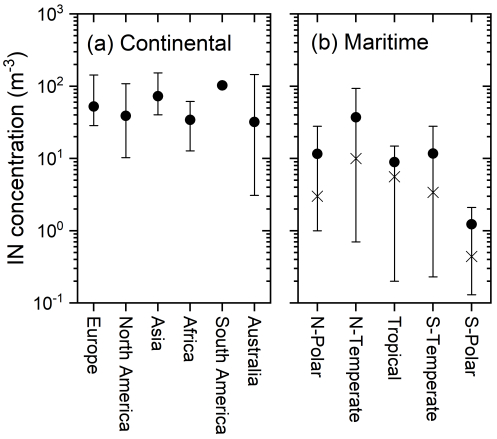

Figure 2. (a) Regionally averaged continental IN concentrations at -15 °C (surface-based measurements, Bigg and Stevenson, 1970); (b) Meridional averaged maritime IN concentrations at -15 °C (shown as dots). Vertical bars indicate the concentration range where 80% of observations lay, excluding the 10% highest and 10% lowest values. Most frequently measured (mode) concentration is indicated by crosses. Continental concentrations in (a) are higher, with a lower variability than maritime IN concentrations in (b).

average concentrations over continents and the variation is higher. The continental data set consists of measurements taken in the same year and season with one type of sampling device and analysis method, while ship-based measurements are a compendium of data obtained with different methods over decades, which could add to the variability in the ship-based data set. In Fig. 2(b), with the exception of clearly lower IN concentrations in the S-Polar zone, the average IN concentration further north (S-Temperate to N-Polar), shows low zonal dependence at -15 °C. In accordance, surprisingly similar concentrations on different continents have been found in the coordinated attempt to measure IN concentrations globally (simultaneous sample collection in 44 locations; Bigg and Stevenson, 1970). However, some contemporary data (e.g. Mason et al., 2016; DeMott et al., 2017) suggest that the values from Bigg and Stevenson (1970) may underestimate the present magnitude and range typifying the boundary layer over continental North America and Europe by a factor of 3 or more, while other data, e.g., in the Eastern Mediterranean region remain consistent with the prior assessment (Gong et al., 2019).

In general, the highest concentrations of an ice active aerosol are found at its source and concentrations decrease with increasing distance away from the source due to random dilution with IN-free air during transport (Anderson et al., 2003). An increase in dilution time can have the same effect as transport. Random dilution causes concentration to vary even at a fixed position relative to the source (Ott, 1990). The higher concentration and smaller variation range of continental IN concentrations could indicate vicinity to the sources (minimal dilution), while the IN population on the ocean is more diffuse, suggesting extended, random dilution during transport or low, fluctuating source strength.

5 Temperature spectra

The ship-based data are used to construct zonal temperature spectra of the IN concentration and to investigate the frequency with which concentrations are measured (shown in Fig. 3). The ambient temperature spectrum at any location is a superposition of IN contributions from several sources activating at specific temperatures. As different sources can contribute IN active at different temperatures, a change of nearby sources and source strength, can generate a change in the temperature spectrum shape (cf. Bigg (1961); Welti et al. (2018) for a discussion of temperature spectrum features). In the ship-based data, examples of non-monotonic temperature spectra with step-like features were observed close to continents, e.g. elevated IN concentration at -15 °C in the vicinity of harbors in Fig. 1. An increased slope at about -10 °C in the meridional averaged temperature spectra is found in the tropics (measurements 350 km - 680 km off the coast of West Africa, see Fig. 1), suggesting a local source that sometimes contributes IN in this temperature range. Excluding these occasional observations of steps in temperature spectra (occurring in $\leq 5\%$ of observations, see Fig. 3), we find a consistent exponential increase of the IN concentration with decreasing temperature, in all climate zones, with only a small variation in slope on a log-scale.

On the left side column of Fig. 3, the meridional separated temperature spectra of data collected during PS79, PS95, PS106, and ACE are shown. Data measured with the CSU CFDC during CAPRICORN are included in the S-Temperate zone. At -15 °C IN concentrations from all additional data sets (cf. Tab. A1) are also shown to compare the range in which ambient concentrations vary. The normalized frequency with which concentrations occur at each temperature in the temperature spectra is shown in the right column of Fig. 3. The symmetry of contours in Fig. 3 and the offset of mean concentrations from the centers of the ranges in Fig. 2(b) suggest that concentrations follow normal distributions when plotted on a log-scale, indicating an underlying log-normal distribution. It has previously been noted that the frequency with which specific IN concentrations are observed follows such a distribution (described in Isaac and Douglas, 1971; Welti et al., 2018). Two consequences of the log-normal frequency distribution can be seen in Fig. 3. While the complete data set (left column of Fig. 3) shows a spread over 3 to 4 orders of magnitude, frequently measured concentration ($\geq 20\%$) are confined to less than one order of magnitude fluctuation (reddish colours in right column of Fig. 3). Note that at temperatures where data is sparse, high occurrence frequency is biased towards the available data points, which may compromise its reliability. Within the range of the most frequent concentrations (yellow and reddish colours) the temperature dependent change in IN concentration, i.e., the slope of the IN concentration versus temperature, is similar for all five meridional zones and the absolute concentration changes by less than two orders of magnitude between zones. The skewed (log-normal) frequency distribution causes the mode (most frequently measured IN concentration) to be less than the arithmetic mean. Therefore, individual concentration measurements are often below average concentrations.

The temperature above which it becomes most probable to measure no IN in a sample of approximately 10 m^3 (highest frequency on zero-line in the right column of Fig. 3) is at -14 °C in N- and S-Polar and the S-Temperate zone. By contrast, the temperature threshold in the Tropical and N-Temperate climate zone is found between -10 °C to -9 °C.

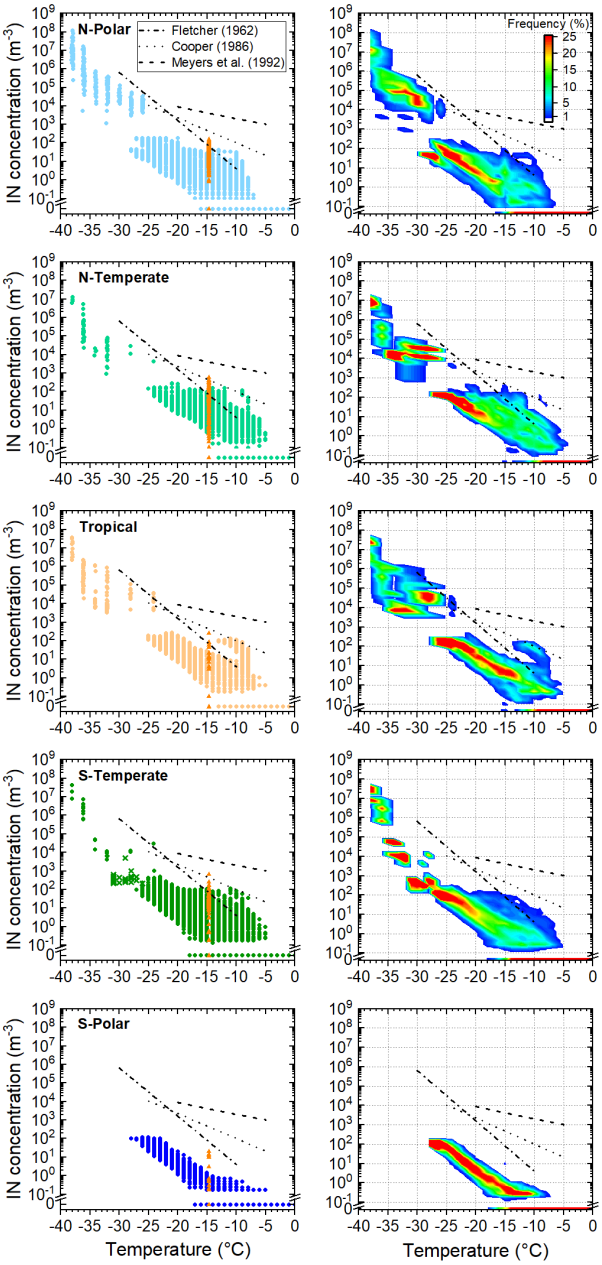

Figure 3. Top-down, maritime data are stratified into N-Polar, N-Temperate, Tropical, S-Temperate, S-Polar (zones are indicated in Fig. 1). **Left column** shows meridional temperature spectra of observed IN concentrations. Data from recent ACE and Polarstern expeditions are shown as circles. In the S-Temperate zone, CSU CFDC data from CAPRICORN are shown as crosses. Literature data at -15 °C (the same shown as crosses in Fig. 1) is overlaid as orange triangles for comparison. **Right column**, frequency of occurrence of measured concentrations at each temperature (1 °C temperature bins) is shown. While the total variation in IN concentration at a certain temperature is large, there is a range of approximately one order of magnitude containing most frequently ($\geq 20\%$) measured concentrations (reddish colors). The frequency with which no IN are detected is shown on the zero-line. Only small, meridional differences in absolute IN concentration and slope of the temperature dependent change can be identified. Three often used parametrization from Fletcher (1962); Cooper (1986); Meyers et al. (1992) are given for comparison.

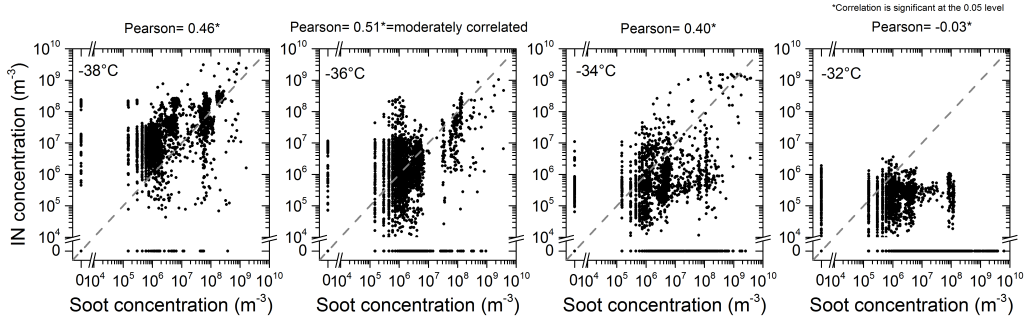

Figure 4. IN vs. soot concentration (10s averages) measured during PS106. Dashed 1:1 lines indicate equal soot and IN concentration. Temperature at which the IN concentration is measured is indicated in the upper left corner of each sub-figure and Pearson correlation of soot and IN concentration is given on top. Moderate correlation is found at -36 °C.

6 Contamination from ship exhaust

To avoid contamination during ship-based measurements, the aerosol inlet and filter sampling devices are placed in front of the ship's chimney towards the bow of the ship. In situations with wind speed from the rear exceeding the speed of the ship, i.e. relative wind direction from the chimney towards the sampling location, high number concentrations of ship exhaust aerosol particles are encountered. Moderate contamination by turbulent mixing is also observed at relative wind speed close to zero. The potential contribution of ship exhaust particles is determined by comparing IN concentration measured with SPIN to soot particle concentration measured with a single particle soot photometer (SP2, DMT). Fig. 4 shows the concentration of soot versus IN concentration, both binned to common 10 s intervals. Moderate correlation is found at -36 °C, whereas soot concentration is higher than the IN concentrations at higher temperatures and lower at lower temperatures. Comparing the temperature spectra in Fig. 3, where a distinct increase in IN concentration at -36 °C can be seen, confirms that ship exhaust potentially distorts the ambient temperature spectrum at and below -36 °C, but not above. Increased concentrations at -38 °C could be caused by the onset of homogeneous freezing. This gives confidence that the effect of contamination from ship exhaust on the immersion freezing experiments with filter samples is negligible. Fig. C1 in Appendix C confirms no effect of sampling air coming from the direction of the chimney of the ship on filter measurements at T $\geq$ -20 °C. It can be argued that at temperatures above -36 °C, the effect of soot if any should be a suppression in activity of other particles by covering their ice active sites (Mossop and Thorndike, 1966). Note that the present analysis is valid for the RV Polarstern exhaust. Other type of fuel or ship engine might yield a different result (Thomson et al., 2018). However, Schnell (1977) also tested a possible contamination from the exhaust of the USNS Hayes by purposely exposing membrane filters to the exhaust plumes from galley and engines and found no IN from either source active at -15 °C. Also McCluskey et al. (2018a) found no statistical significant difference due to varying black carbon levels from exhaust contamination at temperatures between -16 °C and -26 °C on board the RV Investigator during the CAPRICORN campaign.

7 Discussion

The maritime IN concentration data presented here can be used to predict the temperature spectra in a range of oceanic regions. This can help the future development of measurement techniques and sampling strategies. As an example, to avoid artefacts from the sensitivity range of measurement techniques, sample volume and probed temperature range need to be matched. In the current data set, the narrowing of variation below -20 °C in the filter measurements from the temperate climate zone (Fig. 3, right column), is an artefact caused by the upper limit of detectable concentration and can be avoided by sampling smaller volumes and by using dilutions of filter suspensions. In general, the sample volume and sampling time over which concentration is averaged to one data point, affects the absolute value and the variability between measurements. Smaller sample volumes result in larger scattering of the data within experimental detection limits, including measurements of practically zero alternating with high concentrations at the upper detection limit. The smaller the sample volume the higher the limit of detection and vice versa, i.e. larger, sampled air volumes allow the detection of low concentrations, while smaller sample volumes are more suitable for accessing high concentrations.

To develop parametrizations, concentrations averaged to scales required for the application should be used (Gultepe et al., 2001). Grid-box scales in numerical models are much larger than the practical sampling volume of ambient measurements. As averaging any distribution of concentrations by means of probing larger air volumes or averaging collected samples over time converges to the arithmetic mean concentration, the mean IN concentration (shown in Fig. 2(b) for -15 °C in five climatic zones and in Fig. 5(a) averaged over each hemisphere) is the correct input value for large scale numerical models, even if it is not the most frequently measured (mode) concentration. This also underscores the importance to use sufficiently large data-sets for parametrization development to avoid bias.

The frequency distributions in the right column of Fig. 3 illustrate that within the sample volumes where IN concentrations can be measured, low values are more frequent than high values. Averaging concentrations from a skewed (log-normal) frequency distribution results in mean concentrations higher than the most frequently measured concentration. Fig. 5 depicts the temperature spectra of a) average and b) most frequently measured IN concentration in the north and southern hemisphere. The discrepancy between averaged and most frequently measured concentrations is most pronounced for low concentrations. Within the sensitivity limits of measurements presented here, most frequently no IN are detected above -15 °C in 10 m^3 of maritime air.

Between -25 °C and -30 °C, a jump in the N-hemisphere temperature spectra can be seen in Fig. 5. It could be speculated that the jump coincides with the temperature where mineral dust particles start to exert a strong influence (e.g. Boose et al., 2016) and the N-S difference could be due to more dust emitting land masses in the north. Another reason for the jump could be that it is an artefact from the combination of filter measurements reaching their upper detection limit with SPIN measurements reaching their lower detection limit in this range of IN concentration. Further measurements and the development of measurement techniques that can bridge this gap are needed.

In Fig. 1 a clear change towards lower IN concentrations measured recently in the Southern Ocean and the literature values from Bigg (1973) can be seen. Bigg reported averaged concentrations measured in 5x5 degree sectors. Noting the log-normal

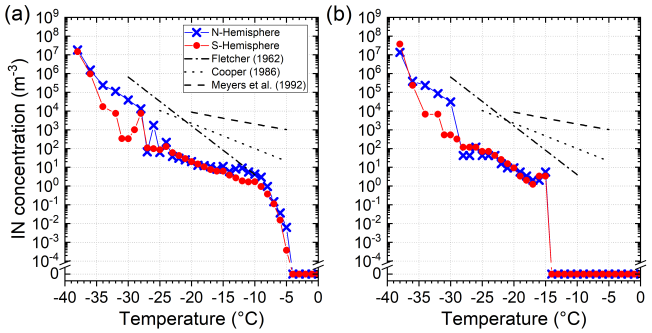

Figure 5. (a) comparison of the *average* IN concentration between the northern and southern hemisphere oceanic regions; (b) comparison of the *most frequent* observed IN concentration in the northern and southern hemisphere oceanic regions. Parametrization from Fletcher (1962); Cooper (1986); Meyers et al. (1992) are given for comparison. The average IN concentrations over both hemispheres shown in (a) can be approximated by: $IN(\mathrm{m}^{-3}) = (T(^{\circ}\mathrm{C}) + a) \cdot (-b \cdot \exp(c/(T(^{\circ}\mathrm{C}) + d)))$, with $a = 5\,^{\circ}\mathrm{C}, b = 10^{-5}\,\mathrm{m}^{-3\,\circ}\mathrm{C}^{-1}, c = 500\,^{\circ}\mathrm{C}, d = 60\,^{\circ}\mathrm{C}$, for $-36\,^{\circ}\mathrm{C} < T < -5\,^{\circ}\mathrm{C}$.

frequency distribution of observed IN concentrations, the offset between the average and most frequently occurring concentration at -15 °C (see Fig. 5) could be one factor helping to explain the difference. A decline in IN abundance in the Southern Ocean over the last decades (suggested by Bigg, 1990) could be another reason.

8 Conclusions

Ship-based observations are an efficient strategy to obtain measurements with a wide spatial coverage representing IN characteristics over the (remote) maritime environment, distant from continental sources. The present study provides a summary of IN concentrations observed during ship expeditions in the last 50 years, in different oceanic regions. This overview can be used to validate parametrizations of IN concentrations in numerical climate models (see data availability below) and to plan future expeditions.

In the data presented here, the lowest IN concentrations are observed in polar regions and highest in the temperate climate zones. The overall geographical variation in maritime IN concentration is surprisingly small (below two order of magnitudes). A zonal difference was found in the temperature above which most frequently no IN are detected (-9 °C in N-Temperate, -10 °C in Tropical, but much colder at -14 °C in S-Temperate, N-, and S-Polar climate). The largest difference between northern, and southern hemisphere is found at temperatures below -28 °C, where IN concentration is higher in the northern hemisphere. This coincides with the temperature range where desert dust particles become especially active ice nuclei.

A comparison of ship emitted soot and IN concentration in this and other studies shows that contamination from ship exhaust during expeditions does not bias IN concentration measurements at temperatures above -36 °C.

Recent, maritime ice nucleation measurements show lower IN concentrations compared to reported mean values from the

1970s and 80s. Apart from the possibility of an actual decrease in the maritime IN concentration, the log-normal frequency distribution of measured IN concentrations could account for some of the difference in this direction. Individual observations from a variable following a log-normal frequency distribution are most frequently lower than the arithmetic mean. This can also be one reason for the difference between recent observations and e.g. the Fletcher (1962) parametrization for IN concentration which is based on older data that has been reported as mean values.

To establish an unbiased climatology of IN concentration in the maritime environment, measurements pursuing to extend the spatial coverage and to capture the temporal variability of IN are needed, e.g. to capture the dependence on marine productivity, season, meteorology, special aerosol emissions and to identify IN sources. To achieve these goals a continued effort to include ice nucleation measurements as important observations on board of scientific expeditions is needed.

Data availability. Data sets are available from the authors upon request and can be obtained from the following repositories: Literature data, PS79, PS95, AOE-91, AOE-96, AOE-2001 can be accessed in the BACCHUS ice nucleation database (https://www.bacchus-env.eu/in/). ACE data set (available from CT, doi in preparation), ACAPEX (https://www.arm.gov/research/campaigns/amf2015acapexicenuclei), SHIPPO data set (available from PJD), CAPRICORN (https://hdl.handle.net/10217/192127), PS106 data set (https://doi.org/10.1594/PANGAEA.919194), Tan1502 data set (available from PJD), INARCO data is available at https://doi.org/10.1594/PANGAEA.903695.

Author contributions. AW analysed the data and prepared the manuscript with contributions from all co-authors. Expedition preparation, sample collection and experimental work was conducted by all co-authors. FS, SH, MvP, JS, CL, PJD, TCJH acquired funding.

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

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Table A1. List of ship expeditions on which IN concentrations were measured. References indicate publications that reported IN concentrations.

[1] Schnell (1977), [2] Schacher (1976), [3] Bigg (1973), [4] Borys (1983), [5] Rosinski et al. (1986), [6] Rosinski et al. (1988), [7] Rosinski et al. (1995), [8] Bigg (1996), [9] Bigg and Leck (2001), [10] McCluskey et al. (2018a), [11] Si et al. (2018), [12] Irish et al. (2019), [13] Creamean et al. (2019), [*] new IN data set

Ref.	Expedition	Year	Location (lat)	Sampling Equipment	Interval	Volume
[1, 2]	Marine fog cruise	Jul.-Aug. 1975	40-46°N	Filter	2-13 h	1.4-9.1 m^3
[3]	USARP	1969-72	20-78°S	Filter	12 h	0.3-3 m^3
[4]	YMER-80	Aug.-Sep. 1980	79-83°N	Filter	24 h	1 m^3
[5]	Marine Sinks of Atmospheric CO_2	Feb.-May 1984	7°N-57°S	Filter	2 h	0.6 m^3
[6]	On board of RV H-02	Jul.-Aug. 1986	19-29°N	Filter	2 h	0.6 m^3
[7]	On board of RV Xiangyanghong 14	Apr.-May 1992	29-32°N	Filter	10 min	0.1 m^3
[*]	ACE	Nov. 2016 - Mar. 2017	53°N-78°S	LowVol Filter	8 h	10 m^3
[*]	PS 106	May-Jul. 2017	53-80°N	LowVol Filter	8 h	10 m^3
				SPIN	30 min	30 ℓ
[*]	PS 95	Oct.-Dec. 2015	53°N-34°S	HiVol Filter	24 h	3 m^3
				SPIN	30 min	30 ℓ
[*]	PS 79	Apr.-May 2012	54°S - 53°N	HiVol Filter	24 h	3 m^3
[8]	AOE-91	Aug.-Oct. 1991	75-90°N	Filter	1-24 h	0.3-3 m^3
[9, *]	AOE-96	Jul.-Sep. 1996	75-90°N	Filter	1-24 h	0.3-3 m^3
[*]	AOE-2001	Jun.-Aug. 2001	75-90°N	Filter	1-24 h	0.3-3 m^3
[*]	TAN1502	Jan.-Mar. 2015	42-75°S	Filter	24 h	8-20 m^3
[*]	ACAPEX	Jan.-Mar. 2015	27-38°N	Filter	24 h	4-5 m^3
[*]	SHIPPO	Jul. 2012, 2014	63°N-43°S	Filter	4-47 h	3-24 m^3
[10]	CAPRICORN	Mar.-Apr. 2016	43-53°S	Filter	21-63 h	19-55 m^3
				CSU CFDC	3-33 min	(167x) 4-50 ℓ
[11]	NETCARE	Jul. 2014	55°N	Impactor	6 h	11.2 m^3
[12]	NETCARE	Jul.-Aug. 2014	67-81°N	Impactor	20 min	0.2 m^3
[13]	INARCO	Aug.-Sep. 2017	54-74°N	Impactor	12-24 h	20.6-41.2 m^3

Appendix A: Overview of expeditions contributing data

Tab. A1 gives an overview on all expeditions that contributed data sets, when and where they took place and how IN concentration has been obtained. In the following, the expeditions from which IN concentration data are reported for the first time (indicated by [*] in Tab. A1) are briefly described. References to more detailed information on expeditions and expedition reports are included.

The Antarctic Circumnavigation Expedition - ACE from November 2016 to March 2017 was the first expedition carried out by the Swiss Polar Institute. On board the Russian research vessel Akademik Tryoshnikov the voyage started with the transit from Bremerhaven, DE to Cape Town, ZA from where on the Antarctic continent was circumnavigated in 3 legs. Leg 1 from Cape Town to Hobart, NZ; leg 2 Hobart to Punta Arenas, CL; leg 3 Punta Arenas to Cape Town. The final leg 4 was the return from Cape Town to Bremerhaven. ACE included atmospheric, glaciological, oceanographic, biological and biochemical observations to better understand Antarctica's ecosystem and the interplay of sea and atmosphere in the Southern Ocean (Walton and Thomas, 2018).

The PS106 campaign to the Arctic (PS106/PASCAL in the framework of AC^3, Wendisch et al., 2018) on board of the research vessel Polarstern was from May 23 to July 20, 2017. For the first leg of the campaign, Polarstern took off from Bremerhaven, DE, towards the Arctic sea to establish an ice camp and drift with the ice shelf north of Svalbard. During the second leg Polarstern crused the sea north-east of Svalbard. The expedition completed in Tromsø, NO. Oceanographic, atmospheric and biological observations were conducted during both legs (Macke and Flores, 2018).

The PS95 campaign on board of research vessel Polarstern was an Atlantic transit ~~cruse~~ cruise (October 29 - December 3, 2015) from Bremerhaven, DE to Cape Town, ZA. Observations included biological oceanography, atmospheric aerosol and remote sensing (Knust and Lochte, 2016).

The PS79 campaign (ANT-XXVIII/5) on board of research vessel Polarstern left Punta Arenas, CL on the April 11, 2012 to cross the Atlantic and reach Bremerhaven, DE on May 16, 2012. The transect was used for biological, atmospheric and marine observations and to measure sea-atmosphere fluxes (Bumke, 2012).

AOE-96 was the Arctic Ocean Expedition departing from Gothenburg, SE on a Swedish research vessel, the icebreaker Oden. The expedition lasted from July to September 1996. During the first leg observations were taken in the Barents Sea and the pack ice of Nansen and Amundsen basins, including an ice camp. On the second leg the expedition reached the North Pole before sailing back to Gothenburg. Observations included chemical and physical atmospheric measurements, meteorology, biochemical measurements and seawater chemistry (Leck et al., 2001).

AOE-2001 was an interdisciplinary Arctic Ocean Expedition on the Swedish research vessel Oden. Departing on June 29 from Gothenburg, SE the cruise reached pack ice north of Svalbard and continued to the North Pole. A 3-week ice camp was established on the way back and the expedition concluded on August 26. The observational program included marine biology, atmospheric chemistry, aerosol chemistry and physics, meteorology, oceanography and seismology (Leck et al., 2004).

TAN1502 refers to the NZ-Australia Antarctic Marine Ecosystems Voyage of the NIWA RV Tangaroa vessel, which occurred from 28 January to 15 March, 2015, sailing from Wellington, NZ to the Ross Sea and back. The main objective of this voyage was to gather baseline ecosystem information at the establishment of the worlds largest Marine Protected Area located in the Ross Sea region of Antarctica in 2017 and legislated to be in place for 35 years. The voyage included underway oceanographic and atmospheric observations.

ACAPEX was the U.S. Department of Energy's Atmospheric Radiation Measurement (ARM) Cloud Aerosol Precipitation Experiment. Samples were collected on the NOAA RV Ronald H. Brown vessel, from 14 January 2015 to 10 March 2015, while sailing between Honolulu, Hawaii and San Francisco, CA (DeMott and Hill, 2016).

Table B1. Latitudinal subdivision of datasets

Classification	Dividing criteria, latitude	Fraction of earth surface area
N-Polar	66.5°N - 90°N	4.5%
N-Temperate	23.5°N - 66.5°N	25.5%
Tropical	23.5°N - 23.5°S	40%
S-Temperate	23.5°S - 66.5°S	25.5%
S-Polar	66.5°S - 90°S	4.5%
N-hemisphere	equator - 90°N	50%
S-hemisphere	equator - 90°S	50%

SHIPPO, the SHIpborne Pole to Pole Observations included two campains, SHIPPO-2012 and SHIPPO-2014, both of which occurred on the Korean Polar Research Institute's ARAON vessel. SHIPPO-2012 was conducted in July 2012, sailing from Incheon, KR to Nome, AK, USA (DeMott et al., 2016, see SI for data). SHIPPO-2014 sailed from Incheon to Christchurch, NZ.

Appendix B: Latitudinal subdivision of ship-based observations

Along the cruise tracks, sample locations are subdivided into five general climate zones (Tab. B1) to investigate the average IN concentration at -15 °C shown in Fig. 2 and zonal temperature spectra shown in Fig. 3. Differences between IN concentrations over the northern and southern hemisphere in comparison to three often used parametrizations is shown in Fig. 5.

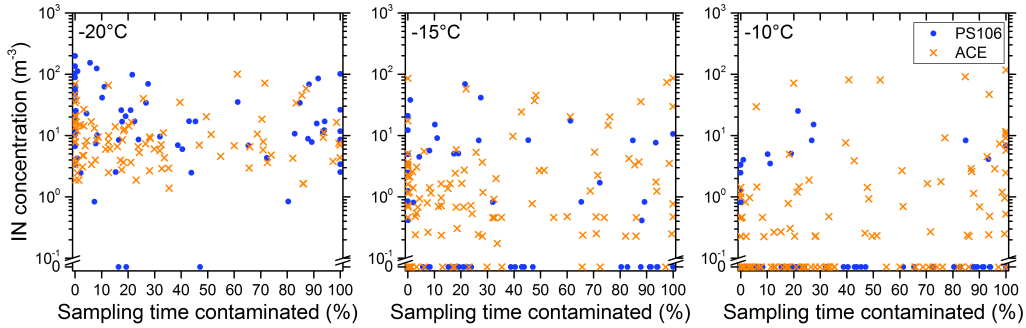

Figure C1. IN concentration vs. sampling time in ship exhaust. Measured IN concentration at -20C, -15C and -10C show no correlation to the time air was sampled from direction of the ships stack. Ships: RV Polarstern (PS106), Academic Tryoshnikov (ACE).

Appendix C: Influence of ship exhaust on IN concentration at T≤≥-20 °C

Duration of sampling time in wind direction of the ship exhaust emission is compared to the concentration of IN collected on 8 hour filter samples. Fig. C1 shows the IN concentration in dependence of duration of sampling from the ship exhaust, at -20 °C, -15 °C and -10 °C. The data show no correlation, indicating that ship exhaust does not affect the measured IN concentration at these temperatures. This is in accordance to the data in Fig. 4 showing that ship emissions only contribute to the IN temperature spectrum at temperatures below -36 °C.

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