Variation of Ice Nucleating Particles in the European Arctic Over the Last Centuries

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Abstract The historical development of ice nucleating particle concentrations (N_{INP}) is still unknown. Here, we present for the first time N_{INP} from the past 500 years at two Arctic sites derived from ice core samples. The samples originate from the *EUROCORE* ice core (Summit, Central Greenland) and from the *Lomo09* ice core (Lomonosovfonna, Svalbard). No long-term trend is obvious in the measured samples, and the overall range of N_{INP} is comparable to present-day observations. We observe that the short-term variations in N_{INP} is larger than the long-term variability, but neither anthropogenic pollution nor volcanic eruptions seem to have influenced N_{INP} in the measured temperature range. Shape and onset temperature of several INP spectra suggest that INP of biogenic origin contributed to the Arctic INP population throughout the past.

1. Introduction

The Arctic is one of the regions most sensitive to climate change, and changes are proceeding at an unprecedented pace and intensity (Serreze & Barry, 2011). Surface air temperature, the most prominent variable to indicate Arctic change, increased almost twice as fast in the Arctic compared to the rest of the globe since the midtwentieth century (Overland et al., 2011; Serreze & Barry, 2011). This enhanced warming phenomenon is referred to as Arctic Amplification. Arctic peculiarities, such as extended snow and ice cover, low elevation of the atmospheric boundary layer, and abundance of low-level clouds, together with multiple feedback mechanisms, like the surface albedo, water vapor, or cloud feedback, are known to contribute to the enhanced climate sensitivity of the Arctic. However, the relative importance, strength, and interconnection of these peculiarities and feedback mechanisms are still disputed (Pithan & Mauritsen, 2014; Serreze & Barry, 2011).

Clouds with their specific microphysical properties and the cloud phase (liquid water droplets or ice crystals) are one of the key factors of Arctic Amplification, because they affect the energy budget of the Arctic boundary layer. They tend to warm the surface by reflecting long-wave radiation (Intrieri, 2002; Shupe & Intrieri, 2004) and consequently can enhance sea ice melt (Vavrus et al., 2011). This in turn can lead to increased evaporation and cloud formation, a feedback that might accelerate in the future (Liu & Key, 2014; Park et al., 2015). A reduced ice cover promotes biological activity in the marine (Arrigo et al., 2008) as well as in the terrestrial environment (Hinzman et al., 2005), which goes hand in hand with the alteration of aerosol particle sources, that in turn may affect clouds and their properties.

Clouds in the Arctic are in a mixed-phase state for about 50% of the time; that is, they consist of water droplets and ice particles at the same time (Intrieri, 2002; Pinto, 1998; Shupe et al., 2006, 2011; Turner, 2005). Despite the unstable nature of mixed-phased clouds, owing to the Bergeron-Findeisen effect, which describes the growth of ice crystals at the expense of liquid droplets, Arctic clouds are extraordinarily long-lived and extend over large areas (Morrison et al., 2011). This makes them important players for the Arctic radiative budget. Primary ice formation in mixed-phase clouds takes place through heterogeneous ice nucleation (IN); that is, ice nucleating particles (INPs) are needed to induce freezing of supercooled cloud droplets (Pruppacher & Klett, 2010), and hence, they affect precipitation, lifetime, and radiative properties of clouds (Loewe et al.,

2017; Ovchinnikov et al., 2014; Prenni et al., 2007; Solomon et al., 2015). Generally, two main sources for atmospheric INP are thought to exist: mineral dust, primarily material from deserts or soils, and biogenic macromolecules originating from bacteria, fungi, lichen, marine biota, and pollen. Mineral dust particles, while being abundant in number, are mostly ice active at temperatures (T) below −20 °C, whereas biogenic INP tend to nucleate ice at higher temperatures of up to −5 °C (Augustin-Bauditz et al., 2016; DeMott et al., 2003, 2016; Kanji et al., 2017; Murray et al., 2012; Si et al., 2018). However, very little is known about the abundance, nature, properties, and sources of INP in the Arctic and how they might change with a changing climate.

Studying the variations of INP concentrations and properties over the past centuries might hence help to better understand potential future changes. Currently, knowledge about historical changes in INP concentrations does not extend far beyond a few decades. Even present-day measurements of INP number concentrations (N_{INP}) are still scarce in the Arctic regions and they usually cover only short periods of time.

Ice cores are a type of climate archive that is widely used to reconstruct and understand Earth′s atmospheric history (Lorius et al., 1990; Petit et al., 1999; Steffensen et al., 2008). However, such investigations have focused on parameters like temperature, greenhouse gases and particulate matter concentrations, or moisture sources. To our knowledge, we use for the first time ice core material to derive INP concentrations on historical time scales. We will discuss possible seasonal and long-term trends, elucidate anthropogenic influences, and provide data for driving, constraining, and evaluating both climate and smaller-scale cloud resolving models.

2. Experimental

2.1. Drilling Sites and Sampling

We used ice core material from two Arctic sites: Lomonosovfonna (Svalbard) and Summit (Greenland). Wendl et al. (2015) and Blunier et al. (1993) already dated and characterized (see 2.1.1 and 2.1.2) these ice cores, respectively. For the present study, samples were freshly cut from the remaining parts of the original ice cores. During the transport of the samples between laboratories, storage at a temperature well below 0 °C was ensured. At the Leibniz Institute for Tropospheric Research (TROPOS) the samples were stored at −24 °C until analysis.

2.1.1. Lomonosovfonna Ice Core

This ice core, hereafter referred as *Lomo09*, was drilled on the Lomonosovfonna glacier (Svalbard; 1,202 m above sea level; 78°49′24″N, 17°25′59″E) in 2009. The core was dated with a combination of reference horizons (1963 tritium peak and major volcanic eruptions), annual layer counting, ^{210}Pb decay, and a simple glacier flow model. The absolute dating uncertainty for the core varies between ±1 year and ±10 years depending on depth and proximity to reference horizons. Concentrations of water soluble major ions were determined, which will be used as complementary information to the INP analysis.

#### 2.1.2. Central Greenland Ice Core

This ice core, hereafter referred as *EUROCORE*, was drilled at Summit (Central Greenland; 72.58°N, 37.64°W) in 1989, at the location of the Greenland Ice Core Program (GRIP). The core was dated with an accuracy of ±2 years by a combination of annual layer counting based on stable water isotopes, electrical conductivity, and chemical data, with identification of volcanic reference horizons. Complementary information useful in context of INP analysis is lacking.

2.2. Measurement of INP Concentrations

We performed INP measurements with the two droplet freezing arrays LINA (Leipzig Ice Nucleation Array) and INDA (Ice Nucleation Droplet Array), based on the measurement principles described in Budke and Koop (2015) and Conen et al. (2012), respectively. The LINA instrument holds 90 drops of 1-μl volume, while INDA uses 96 drops with a volume of 50 μl. The average T_{50} (temperature where 50% of the drops are frozen) for the measurement of ultrapure water with LINA is −24.8 °C, and for INDA T_{50} = −24.4 °C. Further information regarding the measurement techniques can be found in the supporting information (SI).

2.2.1. Derivation of the Atmospheric INP Concentration

Petters and Wright (2015) derived atmospheric INP concentrations from precipitation samples. We argue that their method is also applicable to ice core samples, since ice cores are accumulated from snow precipitation. While particles found in ice cores originate from both dry and wet deposition, it is generally assumed that particles are conserved once occluded in the ice. Wet deposition dominates in the Arctic due to the high snow accumulation (Fischer et al., 2007; Gao et al., 2007). Therefore, we adopt the procedure from Petters
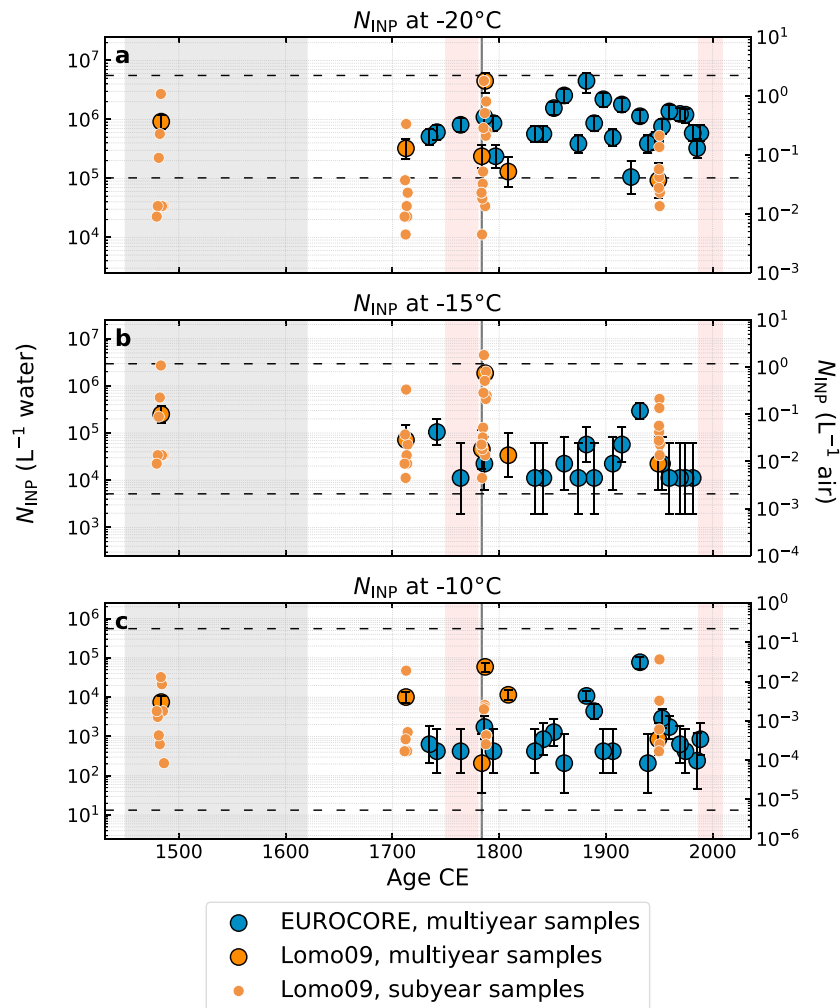
Figure 1. Time series of N_{INP} at $-20\,°C$ (a), $-15\,°C$ (b), and $-10\,°C$ (c) for multiyear and subyear samples at both sites. Note that N_{INP} at -20 and $-15\,°C$ have been obtained with the Leipzig Ice Nucleation Array device, whereas N_{INP} at $-10\,°C$ have been measured with the Ice Nucleation Droplet Array device. Error bars indicate the uncertainty rooting in the methodology itself due to the Poisson distribution of INP (see supporting information for further details). As a reference, black dashed horizontal lines are included which show the range of values observed by Petters and Wright (2015) for present North America and Europe. The shaded grayish area delineates a period of reduced Arctic sea ice extent found by Kinnard et al. (2011), and shaded reddish areas show intervals of particularly warm summers on Svalbard by D'Andrea et al. (2012). The black vertical line indicates the period of the 1783 CE Laki eruption as defined in Fiacco et al. (1994). INP = ice nucleating particle.

and Wright (2015) for estimating atmospheric INP concentrations from ice core water samples. However, it cannot be excluded that a small portion of the INPs measured from ice core samples originate from dry deposition. INP from ice core samples should therefore be seen as an upper maximum.

The basic idea behind the derivation is to relate the concentration of INP in the precipitation sample (number of INP per volume of precipitation) to the concentration of INP in the air (number of INP per volume of air) through the condensed water content (CWC), that is, the amount of precipitable water per volume of air.

$$N_{INP}(L^{-1}\text{water})/\text{CWC}(L_{water}/L_{air}) = N_{INP}(L^{-1}\text{air}) \qquad (1)$$

A key uncertainty in this conversion is the CWC itself, which can vary between 0.2 and 0.8 g/m³ (Petters & Wright, 2015). We calculate the atmospheric N_{INP} values with CWC = 0.4 g/m³, which leads to an uncertainty of a factor of 2. Model studies and field observations of Arctic clouds show that in the Arctic a CWC of 0.4 g/m³ is rarely exceeded (Lawson et al., 2001; Leaitch et al., 2016; Stevens et al., 2018). Therefore, the used value for the CWC can be seen as an upper bound and the resulting uncertainty might even be smaller
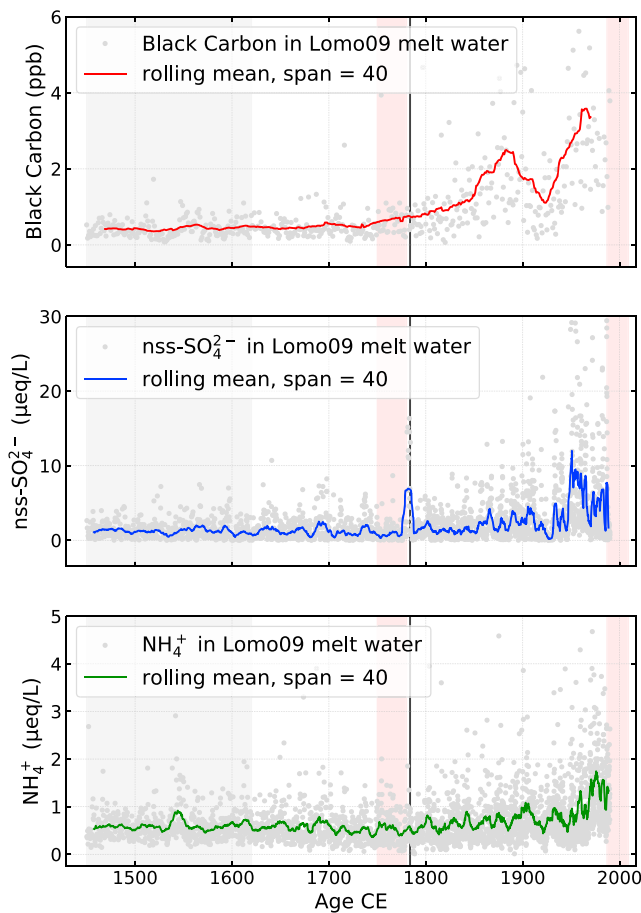
Figure 2. Time series of black carbon, non-sea-salt sulfate (nss-SO$_4^{2-}$), and ammonium (NH$_4^+$) derived from the melt water of *Lomo09*. Gray dots show the individual values, while the colored lines are the rolling means of 40 values. Original data from Osmont et al. (2018) for black carbon and Wendl et al. (2015) for nss-SO$_4^{2-}$ and NH$_4^+$. The shaded reddish areas show intervals of particularly warm summers on Svalbard by D'Andrea et al. (2012).

than a factor of 2. Further uncertainties are the presence of dissolved solutes and chemical aging of INP leading to estimated uncertainties of less than one order of magnitude. Petters and Wright (2015) also mention preconcentration of INP in the precipitated water due to high precipitation evaporation rates as factor to be considered at certain sampling locations (dry climates, e.g., Antarctica, and high cloud base). However, due to the usually low cloud bases (Morrison et al., 2011; Shupe et al., 2006, 2013) and high humidity below the clouds in the Arctic (Maturilli et al., 2013; Shupe et al., 2013), we consider this effect negligible for our study.

3. Results

Forty-two samples were investigated from the *Lomo09* ice core covering the period from 1480 to 1949 CE. Six of these samples span periods of 2 to 6 years (hereafter referred to as multiyear samples), while the remaining 36 samples span several months (hereafter referred to as subyear samples). The subyear samples are a subset of the multiyear samples, covering similar periods, but with higher temporal resolution (see SI Table S3).

Twenty-seven multiyear samples of the *EUROCORE* ice core were analyzed, each covering 2 to 5 years and representing the period from 1735 to 1989 CE (see SI Table S4). No subyear samples were available for this ice core.

Figure 1 presents a time series of N_{INP} at −20, −15, and −10 °C. As reference, the minimum and maximum $N_{INP}(T)$ observed by Petters & Wright (2015; black dashed horizontal lines) are included, indicating the range of N_{INP} obtained from precipitation samples collected mainly in present-day North America and Europe. Periods with different characteristics are highlighted in the figure: reduced Arctic sea ice extent (Kinnard et al., 2011), shaded grayish area, intervals of particularly warm summers on Svalbard (D'Andrea et al., 2012), shaded reddish areas, and the period of the 1783 CE Laki eruption as defined in Fiacco et al. (1994), vertical black line. The temperatures were chosen to be representative for the mixed-phase cloud temperature regime, while still providing a good temporal coverage with data points (the full freezing curves are given in the

SI). A Poisson distribution of INP characterizes our measurement methodology. Hence, the measurement uncertainty of each data point depends on the underlying number of frozen droplets (see SI and Figure 3). At both locations N_{INP} of the multiyear samples feature no clear trend and vary during the past 500 years mostly within the range reported by Petters and Wright (2015). The subyear samples display a similar, if not larger variation.

In Figure 2, time series of black carbon (BC; Osmont et al., 2018), non-sea-salt sulfate (nss-SO$_4^{2-}$), and ammonium (NH$_4^+$), both from Wendl et al. (2015), are shown for the *Lomo09* ice core. All three time series are influenced by anthropogenic emissions: BC originates primarily from fossil fuel and biomass burning; NH$_4^+$ from agriculture, livestock, and biomass burning; and nss-SO$_4^{2-}$ from fossil fuel combustion and metal smelting. Sudden peaks in nss-SO$_4^{2-}$ concentrations can also be attributed to volcanic eruptions, such as the eruption of the Laki fissure (Iceland) in 1783 CE. It can clearly be seen that all concentrations show increasing trends toward the present. BC increased from preindustrial times (before 1700 CE) to the last decades of the twentieth century (since 1950 CE) by a factor of 7.6, nss-SO$_4^{2-}$ by a factor of 4.5, and NH$_4^+$ by a factor of 1.99. However, in Figure 1, no comparable clear overall trend in N_{INP} can be observed at none of the freezing temperatures. At −20 °C samples from the *EUROCORE* ice core show medium-range N_{INP} values with comparatively little variation (between 0.1 and 1.0 L^{-1} air) from 1734 to 1840 CE, followed by a period with generally higher values, and also a larger variability (1840–1938 CE), followed again by a period of medium-range values. For the *Lomo09* ice core it is difficult to assign periods in a similar fashion, since the

samples cluster around six narrow time windows. However, it is worth mentioning, that the highest N_{INP} was observed subsequent to a warm period on Svalbard from 1750 to 1780 CE (D'Andrea et al., 2012).

In the following, we will discuss the time series and suggest hypothesis whenever possible.

4. Discussion

There are two key findings in the above presented result: First, our results indicate that N_{INP} varied within the same range as present-day observations and a long-term trend was not obvious. Second, the subyear samples show a larger variability of N_{INP}.

Human activity has been responsible for the change in specific atmospheric components since the industrial revolution around 1750 CE (Solomon, 2007). Figure 2 shows that anthropogenic emissions have clearly modified the aerosol characteristics in the Arctic; however, there seems to be no evident effect on the INP concentration. In fact, this observation is to be expected: Current knowledge of effects of anthropogenic emission on INP shows that apart from BC, which might act as INP through deposition freezing in cirrus cloud regimes (DeMott et al., 1999; Dymarska et al., 2006; Jensen & Toon, 1997) influences are negligible (Chen et al., 2018). Biomass burning can be another source of BC and INP may be co-emitted. However, the INP associated with biomass burning are generally ice active at low temperatures (McCluskey et al., 2014; Petters et al., 2009; Prenni et al., 2012). Also, Borys (1989) did not see a correlation between INP and Arctic haze aerosol, which consists mainly of accumulated anthropogenic pollution. This is in line with the findings presented herein: the increase in anthropogenic emissions transported into the Arctic does not result in an increase in N_{INP}. Volcanoes can have climatic impacts on global scales, and some samples used in this study were explicitly chosen to have temporal overlap with volcanic eruptions: for example, sample T139 covers the whole Laki eruption and the year after the eruption ended (an enlargement of this period can be found in the SI Figure S5). Volcanic ashes mainly act as INP at lower temperatures (< -23 °C), except for supermicron particles, which can initiate freezing already at around -13 °C (Durant et al., 2008; Gibbs et al., 2015; Hoyle et al., 2011). Nevertheless, neither the *EUROCORE* nor the *Lomo09* samples show significantly enhanced N_{INP} during times of an volcanic eruption. A reason may be that large ice active ash particles are not transported to the ice core sites. For both ice cores used in this study no data about the ash particle concentration is available, but the studies by Fiacco et al. (1994) and Kekonen et al. (2005) confirm that tephra originating from the 1783 CE Laki fissure eruption can be found in ice cores, which were drilled in close proximity (<30 km) to the *EUROCORE* and *Lomo09* sites, respectively. From Fiacco et al. (1994) an upper limit for the concentrations of supermicron tephra particles, which are known to be highly ice active, can be derived and compared to N_{INP}. Fiacco et al. (1994) found in ice core water up to 1 particle L^{-1}, but only roughly $4 * 10^{-3}$ L^{-1} could be identified as ash particles in the size range between 1 and 10 μm. N_{INP} at -15 °C ranges from roughly 10^3 to 10^7 L^{-1} in ice core water; that is, the ash particle concentration derived from Fiacco et al. (1994) is orders of magnitudes lower. Hence, it is not surprising that we do not detect higher N_{INP} connected to volcanic eruptions, even in a case of a major eruption in close proximity.

We observe that the short-term variability of N_{INP} is similar or even larger than the long-term variation displayed over the centuries in both ice cores. Short-term variations (hourly to daily) of a similar order of magnitude were already described by Bigg (1961), Bigg and Leck (2001), and Welti et al. (2018). The former two studies attribute this to wind direction and intermittent mixing of air from above cloud base toward the surface. Welti et al. (2018) explain the variability with the nature of the source and, for example, mixing during transport. They state that the frequency distribution of INP concentrations is lognormal as a consequence of successive random dilution events during their transport and that the shape of the distribution allows conclusions on the proximity of the source. The INP population experiences more random mixing with elongated transport. As a consequence, the frequency distribution broadens and follows more the shape of a log-normal distribution. A similar in-depth analysis is not possible for this study due to the comparatively low number of samples, but it was tested if N_{INP} follows a lognormal distribution at eight selected temperatures (see SI Table S1 for the *Lomo09* samples and Table S2 for *EUROCORE*. In both ice cores we see evidence that primarily at lower temperatures the data came from a lognormally distributed population (significance level $\alpha = 0.05$). Thus, long-range transport is a probable source for INP, which are ice active at lower temperatures. Accordingly, for INP that are ice active at higher temperatures, long-range transport is less likely to be the source. However, it cannot be excluded entirely. As a consequence, also an Arctic, possibly local, sources should be considered for INP ice active at high temperatures.
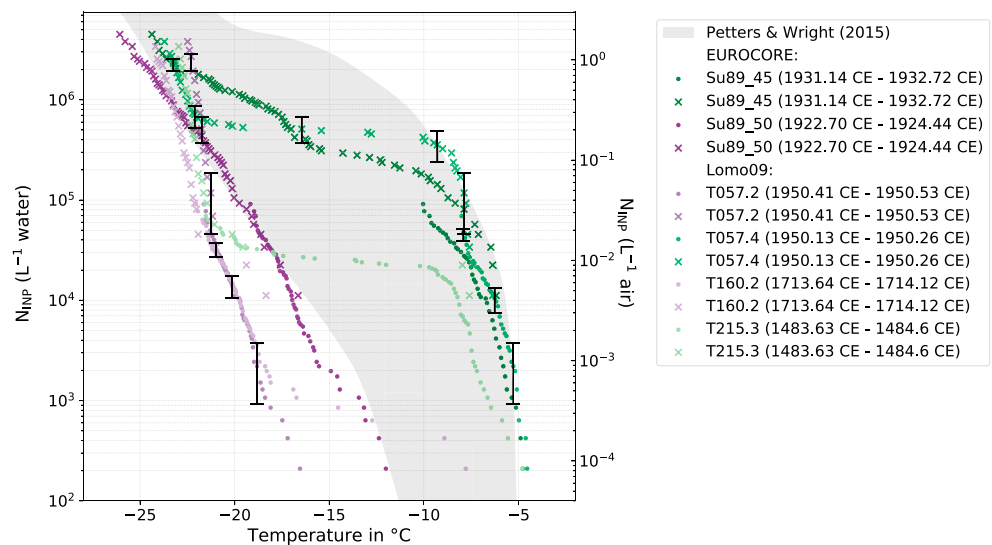
Figure 3. Exemplary selection of INP spectra from samples of both ice cores, showing indication for biogenic INP (greenish colors) and those which do not (purplish colors). Dots indicate INDA measurements and crosses indicate LINA measurements. Individual curves are labeled consistently with the sample list in the SI. Framed in black is an instance of a step-like increase as described in the discussion (section 4). For samples T057.2 and T057.4 the respective error bars of the INDA and LINA data are shown. For clarity only these exemplary error bars are shown. For information on their derivation refer to the SI. The gray shaded area denotes the envelope of all measurements of precipitation samples made by Petters and Wright (2015). INP = ice nucleating particle; INDA = Ice Nucleation Droplet Array; LINA = Leipzig Ice Nucleation Array; SI = supporting information.

Sources from within the Arctic, especially the emergence and vanishing of biological particle sources due to changes in the ice cover, are a possible reason for changes in N_{INP} (roughly 1 to 2 orders of magnitude) during seasonal transitions (Bigg, 1996; Bigg & Leck, 2001; Creamean et al., 2018; Wex et al., 2018). While the subyear samples of our study show a respective pattern, we do not find a statistically significant seasonal variation with high N_{INP} during a particular season. This might be connected to the fact that even the subyear samples often cover several months, which means that they often represent different seasons. Additionally, uncertainty in dating, particularly for the samples that accumulated longer ago, might play a role. Nevertheless, many samples in our study exhibit high N_{INP} already at relatively high temperatures (cf. SI Figures S3 and S4), which is indicative of biogenic INP (Christner et al., 2008; Lindow et al., 1982; O'Sullivan et al., 2018; Turner et al., 1991). The shape of the respective INP spectra also hints at biological INP since a step-like increase (Figure 3) is indicative for those (Beydoun et al., 2017; Welti et al., 2018). Therefore, it is likely that biological INP have contributed to the Arctic INP population throughout the past centuries.

Hicks and Isaksson (2006) show that birch (*Betula*) and juniper (*Juniperus*) pollen, both highly ice active, is present in ice cores from Lomonosovfonna, but the lack of temporal overlap between their and our samples hinders further comparisons. Results from the pollen analysis of the *EUROCORE* ice core show that the two samples (spanning 12.1 and 13.6 years) with the highest concentrations of *Betula alba* pollen occur in the same period where some of the highest and most variable N_{INP} (−20 °C) are observed (Brugger et al., 2019). A further distinction between the continental biological species of the Arctic aerosol (Fu et al., 2013; Hicks & Isaksson, 2006; Moffett et al., 2015; Santl-Temkiv et al., 2018), which can also be ice active (Pummer et al., 2015), and the INP of marine origin (DeMott et al., 2016; Leck & Bigg, 2005; Schnell, 1977; Schnell & Vali, 1975, 1976; Wilson et al., 2015), is not possible. The correlation plot in the SI (Figure S6) shows no clear correlation between N_{INP} at selected temperatures and concentrations of different chemical parameters for the *Lomo09* ice core, preventing further statements about the nature of the INP.

4.1. Atmospheric and Postdepositional Alteration of INP
INP may already be subject to alteration during their transport in the atmosphere, although not many studies on that topic exist. Amato et al. (2015) show that typical IN active bacteria retain their activity on time scales characteristic for the transport between the source and the cloud. Gute and Abbatt (2018) report no effect on IN activity due to UV or sunlight exposure; however, for deposition freezing mode they found that chemical in-cloud oxidation lowers the IN activity of pollen but not of mineral dust.

Postdepositional processes may alter the properties of INP, for instance, due to exposure to air and UV radiation, which is a concern especially for biological INP. Approximately 85% of photochemical reactions occur in the top 10 cm of the snowpack (King & Simpson, 2001). For Lomonosovfonna, with an average annual net accumulation of 0.58-m water equivalent and a density of 0.4 g/cm^3, a net accumulation of 12-cm snow per month is observed, so that the exposure time to photochemical reactions is less than a month. It is unclear if this is sufficient to change IN activity. To our knowledge, the only data available are numbers of viable bacteria, which did not correlate with the age of the ice. Ice deposited more than 12,000 years ago under cool, wet climate conditions on Sajama, Bolivia, contained more recoverable bacteria than modern ice, deposited at the same location during a warmer, dryer period (Christner et al., 2000). Even 5,000-year-old pollen grains are well preserved in ice and do not show any indication of deterioration, when visually inspected under the microscope (Brugger et al., 2018). Polen et al. (2016) postulate a shift of the freezing curves toward lower temperatures with increasing storage time and successive freeze/thaw cycles. However, the correlations in their presented data are not unambiguous and the observed temperature shift does not exceed 3 °C.

Under certain conditions like presence of liquid water and nutrients, bacteria and algae can grow on the glacier surface. In the Lomonosovfonna ice core, we identified a few millimeters thick layers containing colored particles, presumably snow algae. These layers were not collected for INP analysis.

Harrison et al. (2016), Kumar et al. (2018), and Peckhaus et al. (2016) investigated how feldspars, mainly K-feldspars, as representative for the most IN active mineral dust particles, deteriorate over a period of up to 16 months when kept in liquid water. In general, only a slight reduction in IN activity for feldspar particles was observed. A single exception was a sample of hyperactive albite in the study of Harrison et al. (2016), whose mean freezing temperature decreased by 16 °C. Kumar et al. (2018) additionally found that various solutes affect the IN activity of K-feldspar particles differently. However, their solute concentrations were 5 to 6 orders of magnitude higher compared to those found by Wendl et al. (2015) in the *Lomo09* ice core.

The Lomonosovfonna glacier experiences recurrent summer melting, which can alter the ice core records due to water percolation through the snowpack, leading to relocation of chemical compounds or even runoff in the warmest years. However, Pohjola et al. (2002) and Vega et al. (2016) concluded that most of the atmospheric signal was preserved at an annual or a biannual resolution, and at a decadal resolution melt impact was negligible on ionic species (Wendl et al., 2015). It is conceivable that melt events can result in the relocation of INP by several years.

Overall, literature suggests that biological and mineral INP deteriorate only slightly over time, if at all. Also, we do not observe a pronounced decrease in ice activity over time. This might be a further indication that INP are indeed well preserved in ice cores and that ice cores enable a reconstruction of INP concentrations in past climates.

5. Summary and Conclusion

To our knowledge, we present for the first time historic concentrations of INP (N_{INP}) over the past 500 years derived from ice core material at two Arctic sites. The range of INP concentrations was found to be similar to present-day observations (Petters & Wright, 2015) and without long-term trend. N_{INP} did not increase since the beginning of the industrialization, suggesting that anthropogenic pollution reaching the Arctic has not affected N_{INP}. High onset temperatures for IN and the general shape of the N_{INP} spectra indicate the presence of biogenic INP. For those, a local Arctic source that is more active during a particular time of the year is likely. However, with the available data it is not possible to differentiate between marine or terrestrial sources. Although the here presented record of Arctic INP concentrations does not exhibit a clear trend throughout the past 500 years, in view of Arctic Amplification, it has to be kept in mind that the present changes in the Arctic are unprecedented in speed and intensity. New sources of highly ice active biogenic INP may arise or existing ones could be amplified (Arrigo et al., 2008). This is a clear motivation for more studies on sources and transport of Arctic INP. To better understand the implications of potential future changes in INP concentrations, ideally, focused studies would

1. produce a continuous record of N_{INP} in ice cores that represent the last few centuries,
2. extend the historical record of N_{INP} back in time, preferably when significant global climatic changes happened (e.g., Last Glacial Maximum),

3. include joint chemical and biological analysis of substances and species relevant for INP, which are needed for their source apportionment,

4. cover ice cores from multiple sites around the Arctic, helping to investigate the spatial distribution of INP, and

5. link historic N_{INP} records with results from intense present-day field campaigns that study INP characteristics.

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