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Seasonal Variability of Ice Nucleating Particles (INP) in Southern Sweden

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Examiner: Mattias Hallquist, Department of Chemistry and Molecular Biology

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Abstract

Cloud ice crystals are formed by ice-nucleating particles (INPs). The micro-physical properties of clouds, precipitation formation and the life cycle of clouds are strongly influenced by the presence or absence of ice. Therefore knowledge of atmospheric INP concentrations is crucial to improve weather forecasting and climate projections. But global INP concentration vary depending on geographic region, time of the year, their source and also what type of INP being studied. So in-situ measurements which are typically constrained in time and cannot capture more long-term trends such as e.g., a seasonal cycle. Here in this study we investigate daily samples on polycarbonate filter at the Hyltemossa station (Southern Sweden) from March to July 2021 and analyzed their ice nucleating ability. The project involves finding out typical INP concentrations, their variations over time and relationship with other meteorological and aerosol parameters. The main findings we get is INP concentration vary between 0.01 to 1.93 particles L^{-1} but did not show any trend throughout the whole time and no strong obvious correlation with any of the meteorological parameters. An analysis with frequency distribution of INPs with a freezing temperature of -17 °C showed a significant result which indicates sampled air is well mixed and that the INP active at that temperature originate from long-range transport rather than more local sources. We also compared our measurements with parametrizations based on aerosol particle concentration and ambient air temperature. The latter shows reasonably good agreement, while the former over-predicts the INP concentration.

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Introduction

Cloud properties around the Earth's atmosphere are important because they regulate the global distribution of precipitation and influence Earth's radiative budget. One of the most important factors that influences the micro physical properties of clouds is the amount of liquid water and ice present in clouds [8]. Cloud droplets in the atmosphere are formed by condensation of water vapor on aerosol particles and the freezing of cloud droplets can be catalyzed so called Ice Nucleating Particles (INPs). However, forecasting and predicting weather based on cloud processes remains highly uncertain because having poor knowledge about the critical parameters that determined atmospheric INP concentration [9]. One type of clouds called mixed phase clouds (MPCs) where there is a co-existence of super cooled liquid and ice crystals. Here, formation of ice crystals by INP take place at a temperature range between 0 to -38°C [10]. Generally the concentration of active INPs will be increasing with the decreasing temperature. Since there is a large variation of atmospheric condition, so parameters effect on INPs are hard to define but could be useful in predicting processes of primary ice formation [9].

This study aim to investigate the seasonal variability of INP concentration in the filter samples collected in Hyltemossa station for the time period March 2021 to July 2021. Hyltemossa is a research station situated in the south of Sweden and it is an ACTRIS aerosol in-situ station. A Droplet Freezing Assay (DFAs) will be used to analyze the samples collected. The DFA will be able to measure the concentration of INPs above -38℃. To be able to get further insight in how the concentration of INPs are influenced and controlled, additional data from collocated meteorological and aerosol particle measurements will be used. Additionally, the measured INP concentrations will be compared with the predicted INP concentrations obtained from different parametrizations.

2

Background

2.1 What are Aerosols

The term aerosol refers to a suspension of solid or liquid particles in a gas [11]. Aerosol particles are tiny (typically below a few µm in diameter), but still can have a large impact on climate and human health. Aerosol particles tell us about the history of the air mass they are sampled from based on their structure and composition. We can also get the information about the process that the air passed through the last couple of days or weeks. Aerosol particles are typically poly-disperse and have a wide range of sizes; can start from a few nanometers (nm) to several hundred micrometers (µm) in diameter [11]. There are many different anthropogenic and natural sources of aerosol particles. Depending on their source, aerosol particles exhibit a wide variety in their chemical markup. Mineral dust from deserts, pollen from plants, and sea salt from the oceans, are examples for natural aerosol particles, whereas Black Carbon and sulphates often originate from fossil fuel combustion, hence representing anthropogenic aerosol particles. However, it should be noted that also natural phenomena such as wildfires can emit these particle types into the atmosphere [11].

Table 2.1: General sizes of different types of atmospheric aerosol particles.

Aerosol	**Size range**
Dust	1 - 100 µm [12]
Pollen	15 - 50 µm [13]
Bacteria	0.5 - 5 µm [14]
Black carbon	$<$ 2.5 µm [15]
Sulphate	$<$ 200 nm - 6 µm [16]
Sea salt	0.05 - 10 µm [17]
Algae	0.3 - 15 µm [18]

Aerosol particles in the atmosphere vary greatly in sizes, as shown in table 2.1. A variety of factors determine the lifetime of aerosol particles in the atmosphere, including their size and density. Smaller particles are short lived. So coagulation or growth by condensation is limited by smaller particles, while gravitational settling is

rapid for larger particles [19]. When these particles are exposed in the atmosphere, some are capable of changing their shapes, sizes and chemical compositions [20]. Apart from natural and anthropogenic particles, aerosol particles are also divided into primary and secondary particles depending on their mode of formation. Primary particles typically has a diameter of a few micrometers and are generated directly from the surface through dispersion processes [19]. Stratospheric secondary aerosol particles typically have a lifetime of one year compared to particles near the surface that last several days. These particles are mainly removed by wet deposition through precipitation [19]. Some example of primary aerosol particles are sea spray, mineral dust, microbial-/plant materials, dust dispersion by wind etc. Secondary aerosol particles generate from combustion, nucleation of gases or precursor gases during so-called gas-to-particle conversion. In most cases, precursor gases are released from the combustion of fossil fuels, but they may also come from residual emissions of volatile organic compounds (VOCs) [21]. Across urban environments, the aerosol particle density in the atmosphere is nearly ten times higher than that of pristine rural air, and over the ocean, it reaches less than ten times higher than that of pristine rural air [19].

2.2 Aerosols and Climate

Atmospheric aerosol particles have a significant influence on Earth's climate. Climate change, though, is not an anomaly in historical terms. The earth has already experienced numerous warmer and colder periods from the very beginning. Previously climate change has been results of natural climate fluctuations. But nowadays the global climate is changing rapidly, particularly in the Arctic, due to increasing anthropogenic greenhouse gas emissions and will be even more pronounced in future [22].

According to IPCC report 2014 [23], global mean surface temperature is increasing. Over the last couple of decades, global warming has accelerated, particularly in the Arctic and it is expected to be increase several folds until the end of the century [24] [22]. This wide range of predicted temperatures is due to the fact that many processes and feedback mechanisms influence the climate system (positively and negatively). So, it is important to better understand how the climate system interacts with both positive and negative feedback in order to predict future climate change with higher certainty.

A major uncertainty associated with future climate predictions is the contribution of aerosols to climate forcing [23]. In future climate predictions, aerosols are a major contributor to climate forcing. There are direct aerosol effect and indirect aerosol effect, both have a cooling effect on climate Fig 2.1. First of all, scattering the sunlight back to space and absorbing incoming solar radiation by aerosol particles which is direct aerosol effect [25]. Due to the ability to absorb and scatter sunlight, aerosols influence our planet's climate by trapping heat or cooling it down [26]. The so-called indirect aerosol effect refers to all effects that result from the interaction of aerosol particles with clouds. As aerosol particles may act as cloud condensation

nuclei (CCN) and form cloud droplets, they can affect the radiative properties of clouds. So any kind of change in aerosol number, it's concentration and composition can alter the chemical composition, lifetime as well as cloud frequency.

Aerosol particles can also act as ice nucleating particle (INP) which initiate the freezing of cloud droplets. The relative amounts of liquid and ice in a cloud then in turn determines how reflective it is for short- and long-wave radiation. Aerosol particles have impact on precipitation formation, cloud lifetime and also affects surface albedo.

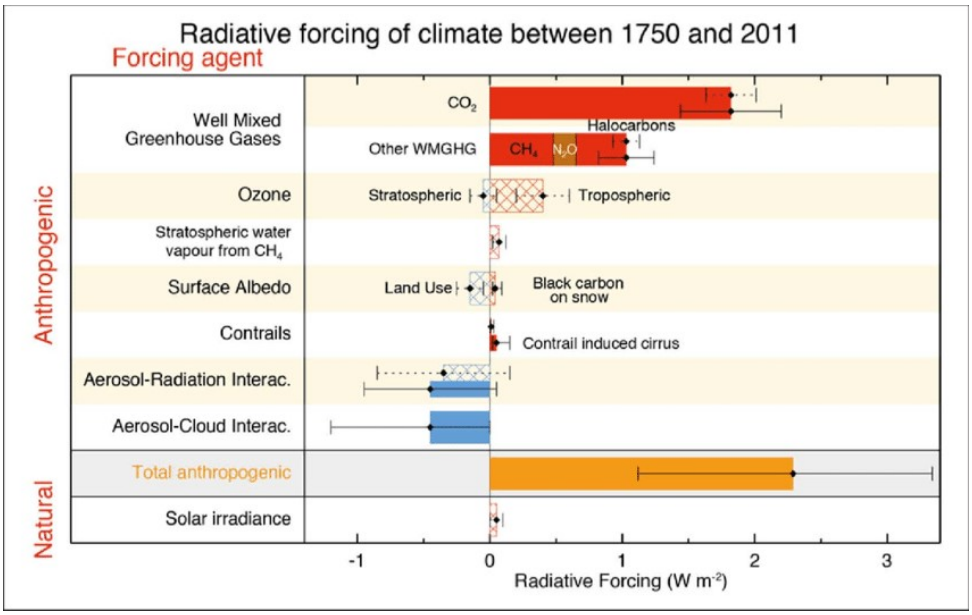


Figure 2.1: Radiative forcing contribution to global temperature from different forcing agents, including uncertainty bars. (IPCC 4th assessment report 2007 [1])

In figure 2.1 we can see there are different forcing agents that are affecting the earth temperature either by cooling or warming. In terms of cooling potential of the anthropogenic aerosols, it may be equivalent to the warming potential of greenhouse gases. Though the uncertainty bars for aerosols effect on radiative forcing are relatively large.

So there are still big gaps in understanding the mechanisms and effects behind the aerosol and climatic forcing as well as aerosol composition and how aerosols are dispersed regionally and globally. Overall, we see there is a net cooling effect from anthropogenic particles. There is a possibility that part of the mean global warming caused by greenhouse gases has been masked by this cooling. By reducing anthropogenic emissions and imposing stricter air pollution regulations, this hidden warming will be revealed [27]. We need more research in this field for quantifications of the climate effect arising from aerosol emissions.

2.3 What are INPs?

It is the ice nucleating particles or the INPs that are able to catalyze the formation of ice crystals above the freezing temperature of pure water droplets (-38°C) whereas without them cloud droplets would remain in a supercooled state [2]. This freezing process above -38°C that is aided by INPs is called heterogeneous nucleation, whereas the freezing process that occurs without a catalyst below -38°C is called homogeneous nucleation. With decreasing temperature, the free energy barrier to nucleate ice is reduced, leading to a greater proportion of aerosol particles that can act as INPs.

There are different types of aerosol particles which act as INP in clouds. The ice nucleating efficiency of different INPs are compared by Murray et al. (2012). They plotted the ice active site density (n_s) against temperature (Fig 2.2).

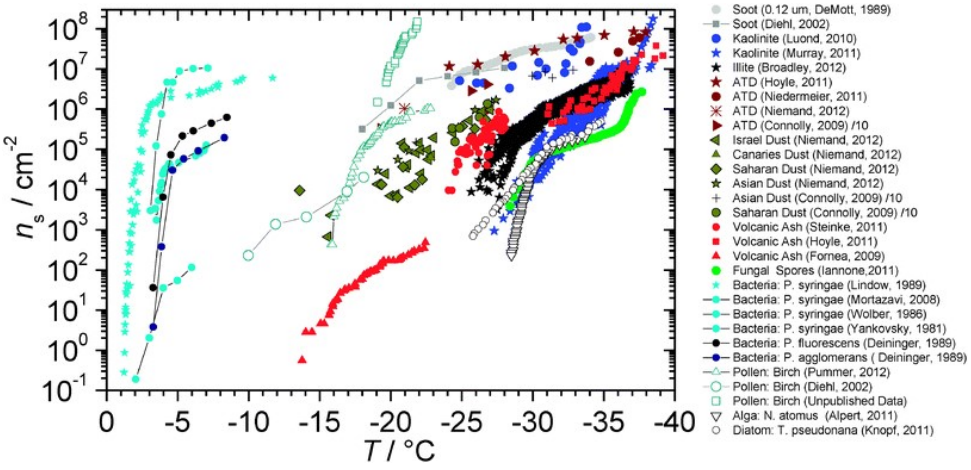


Figure 2.2: The cumulative ice active site density (n_s) values from literature data versus temperature. (Murray et al. 2012) [2]

From figure 2.2 we can see that mineral particles (e.g. ash, mineral dust, etc.) dominate the temperature range below -15°C, and biological particles (e.g.bacteria, pollen, etc.) dominate the high temperatures of -15°C and above.

2.3.1 Heterogeneous ice nucleation: A general description

In thermodynamic system, the nucleation is a process which occurs when an initial metastable phase is replaced by a stable phase [28]. The nucleation process can take place in two different ways. The first, homogeneous nucleation (formation of ice particles from superscooled vapor without the aid of any substrates) and the second heterogenous nucleation (nucleation facilitated by the presence of any nuclei or ion as a third phase).

In case of heterogenous nucleation, primary ice formation in the atmosphere at temperatures (T) > -38°C occurs because it is INPs that act as a substrate for the formation of ice crystals much faster. An extremely small fraction, approximately

one in a million atmospheric aerosol particles can trigger heterogeneous ice formation at -20°C [29]. Having a solid surface lead to lowered critical Gibbs free energies in supercooled liquid droplets and start heterogenous nucleation. The reason for this is when there is solid surface preset in supercooled water droplet, then less water molecules will be needed to the cluster to reach a critical size and formation of ice.

2.3.2 Modes of heterogeneous ice nucleation

In the atmosphere, heterogeneous ice nucleation can be happened by four mechanisms according to Vali (1985) [30] (Figure 2.4): deposition, contact, immersion and condensation.

Deposition nucleation, where liquid water phase is absent. Supersaturated water vapor directly nucleate ice in a supersaturated regime on a solid substrate. Despite the fact that this mechanism is not significant for the formation of ice in mixed phase clouds, it has been proven relevant for the formation of cirrus clouds [6].

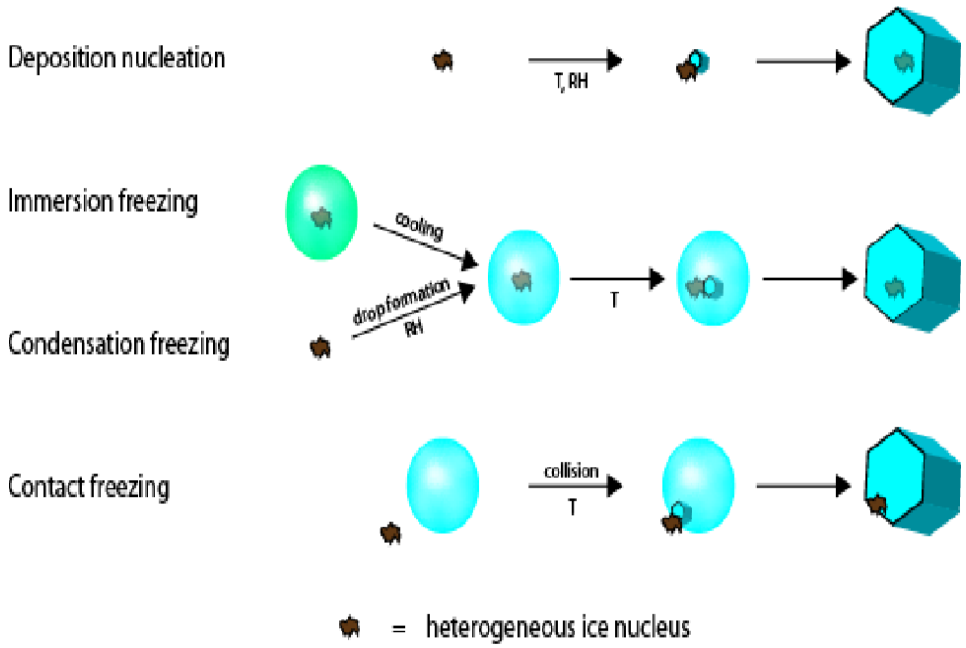


Figure 2.3: Four different mode of heterogeneous ice formation in the atmosphere. (Figure from Chou C. 2011 [3])

Contact freezing is caused by two ways, either by hitting the INP to a supercooled liquid droplet from outside or by moving of an INP from inside toward the droplet surface edges.

Nucleation of supercooled water droplets is triggered when an INP is immersed in a supercooled aqueous solution, which is immersion freezing. This method is the

most important freezing process in mixed-phase clouds [31].

Condensation freezing is sometimes referred as deliquescence freezing or sometimes it is considered similar to the immersion freezing. Water vapor condenses on solid particle in presence of hygroscopic materials and then freeze. [2] [6].

2.3.3 Sources of INP

INPs can come from variety of sources, like biological sources, combustion, volcanic eruptions, geological sources, agricultural practices, transportation etc. In the following an overview about the most important biological and non-biological sources of INPs are presented.

2.3.3.1 Biological Sources

Biological INP sources are among the most important one for INPs that can nucleate ice at high temperatures (above -15℃). Laboratory studies shows that, INPs derived from inorganic materials are generally exhibit significant activity below -15℃), whereas those derived from biological sources can be quite effective above -15℃). Both active and passive processes contribute to the aerosolization of biological particles. It can both be natural and anthropogenic process that produce biological aerosols. They are generally organic particles. The natural sources can be plant debris, spores from bacteria, fungi, pollen from forest, debris from skin or hair, insect eggs etc. The anthropogenic particle sources are produced by industrial processes or farming practices [2]. The distinction between natural and anthropogenic aerosol can be quite challenging when it comes to INP sources.

2.3.3.2 Non-Biological Sources

Geological aerosols are the sources for non-biological INP. They can be both natural or man-made. They come from different sources like soil, deserts, volcanoes, oceans etc. from where dust, sea salt etc. particles can originate. Generally amount of INPs are higher in continental regions than coastal [32]. A study conducted by Kanitz et al. (2011) [32] showed that, there is a large difference in the heterogeneous freezing behavior of clouds containing ice in different locations in the world. They conducted the experiment in four different areas: Leipzig (densely populated area in the mid-east of Germany), Stellenbosch (South Africa), coastal Punta Arenas and Cape Verde. They found that in Leipzig, at around -20°C almost all clouds exhibit some fraction of ice. But in coastal areas (Punta Arenas and Cape Verde) it needs almost -27°C to -33°C for 50% freezing fraction. So of course the location of INP sources is crucially influenced by geography.

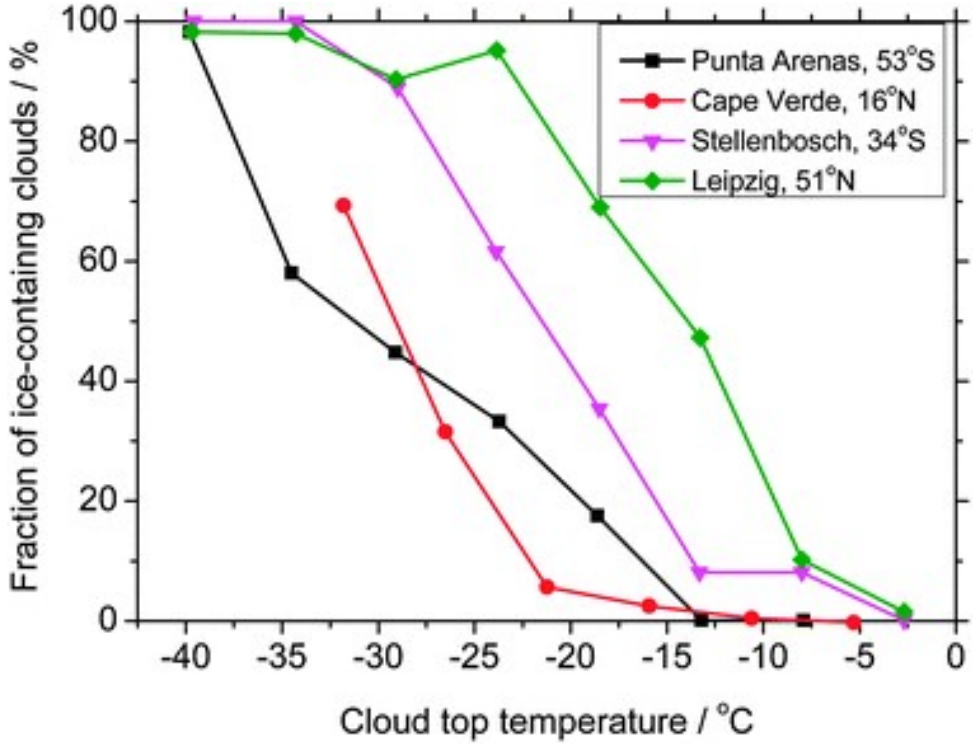


Figure 2.4: The fraction of ice (f_{ice}) containing clouds (stratus clouds) in different locations at an altitude of 2-8 km with a thickness of less than 1 km (Murray et al. 2012) [2]

Combustion and mineral dusts are also non-biological sources of aerosols. They have a large emission rate and efficiently have high ice nucleating ability above -15℃ [8]. But still geographical sources (where INP sources has originated) play the most important role for nucleating ice. Biomass burning, soot from combustion processes, aircraft and car emissions, black carbon are just some examples of human-generated aerosols. However, natural sources such as lightning and wildfires can also contribute to the production of aerosols.

Nowadays human activity has resulted in dramatic increases in carbonaceous combustion aerosol emissions [33].Fossil fuel combustion is the main source of carbonaceous combustion and they have a significant impact on cloud properties.

2.3.4 What makes an effective ice nucleating particle?

There are a number of criteria for determining if an IN is effective. The main ones are the IN's insolubility, size, requirement of its chemical bond and its crystallography to template ice (Pruppacher and Klett, 1996) [28]. In spite of this, only a very small percentage of atmospheric aerosols are capable of triggering heterogeneous ice nucleation, including inorganic, organic, and biological particles (the ice nucleation efficiency of soot particles has been proved to be very little or even null) [34]. Particle size and crystalline properties determine the freezing temperature of each INP.

In addition, the free energy barrier for nucleation decreases with decreasing temperature, resulting in an increase in the number of particles able to act as INPs. [10]

2.4 Cloud Formation and Mixed phase cloud

In terms of regulating the Earth energy budget, clouds play an important role [23]. But still one of the main source of uncertainty in climate projections is clouds and how they respond to global warming [23]. This is because, in climate models, clouds and their associated processes are represented inappropriately, either because they are not understood or because they are incompletely represented.

Cloud formation occurs when the water vapor goes up in the troposphere and condense there into small water droplets on aerosol particles [35]. When cloud is formed, the aerosol particles play important role for cloud properties as because aerosol particles that act as so called Cloud Condensation Nuclei (CCN) [36].

One type of cloud called the mixed-phase clouds (MPCs) contains ice crystals and super cooled liquid droplets. This type of cloud can be found at all latitudes and therefore their processes are important for prediction of Earth's weather and climate. As because super-cooled liquid water has a lower saturation vapor pressure than ice, MPCs are thermodynamically unstable. [37] There is a gap of knowledge how MPCs influence on the climate gives an uncertainty in today's climate model. [38]

2.4.1 Aerosol–cloud interactions

Aerosol concentration, composition, and size have a significant impact on the radiative properties and lifetime of clouds. As because the troposphere lacks favourable conditions for nucleating droplets directly from vapour, this sensitivity arises. So to form liquid clouds droplets or to form ice particles in the clouds, aerosol particles must serve as cloud condensation nuclei (CCN) or ice nuclei (IN), respectively. Droplets form in clouds more frequently when the number of aerosols increases. As a result, clouds with a higher number of droplets scatter light more effectively, which increases albedo and increases the lifetime of clouds [2].

Aerosol concentration increases albedo in the atmosphere, which in turn increases the amount of solar radiation reflected back to space. At present, it is difficult to quantify further changes to clouds caused by ice nucleation. Cloud physical processes like radiative transfer, precipitation, cloud electrification etc are greatly influenced by atmospheric ice particles. Also, they contribute considerably to the chemistry of the troposphere [39]. A major influence on the troposphere's composition is to be found in the active chemical reactivity caused by ice particles. They also scavenge semi volatile gases and acidic trace gas species and help to promote reactive heterogeneous chemistry.

At temperatures between 0°C and about -37°C, mixed phase clouds are present. These clouds are usually found in the low and middle troposphere, where they have

a significant influence on the climate. Due to the presence of more microphysical processes as well as changes in the liquid phase, MPCs are more susceptible to anthropogenic aerosols than warm clouds. A study conducted by Kanitz et al. 2011 [32] and further complied by Murray et al. 2012 [2] shows that the due to different geological location, a fraction of cloud can contain different fraction of ice.

2.4.2 Impact of ice nucleation in mixed phase clouds on climate

There has been a dramatic change in aerosol particles and their composition because of industrialization. We are living in an era when the number of aerosol particles in the atmosphere is greatly increased over the continental regions as a result of human activity [40]. Nowadays, since anthropogenic sources are close to continental surfaces, continental aerosol loads are orders of magnitude greater than those over oceans [31]. An illustration of the dramatic changes in aerosol loading in the atmosphere since pre-industrial times is shown in figure 2.5 .

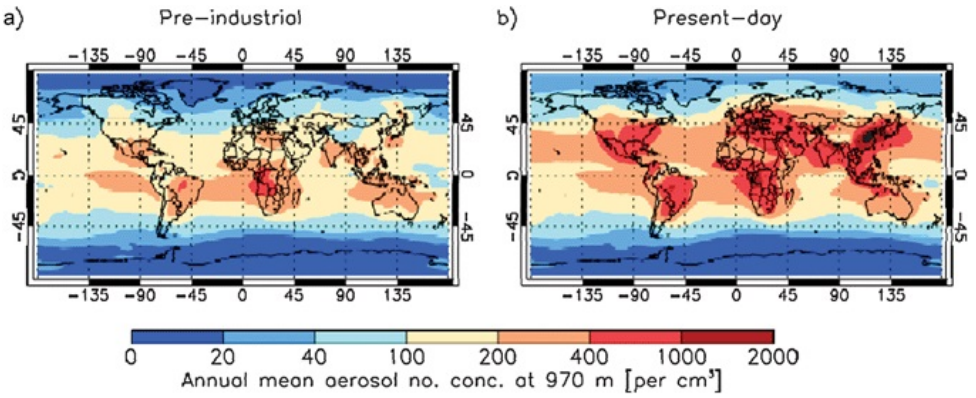


Figure 2.5: Mean annual aerosol particle concentration in the pre-industrial and present day situation (figure from Murray et al. 2012) [2] [4].

There are different direct and indirect effects on climate when INP present in mixed phase clouds compared to clean natural condition. Because cloud droplets grow to larger sizes when there are relatively few cloud condensation nuclei (CCN). The clouds make ice free warm rain process where the droplets are quite large enough because it coalesce with more water droplets during sedimentation. Consequently, liquid water is removed from the cloud, which results in a higher and colder cloud top since latent heat is removed from the cloud. But when IN are present, it makes rapid cloud glaciation [41]. Lohmann and Diehl [42] conducted a study where they showed that when there are IN present, it reduce the lifetime of clouds. Taking into account the fact that ice nucleation increases precipitation, that reduce the lifetime of clouds, leading to substantial warming.

3

Methods

3.1 Sampling of atmospheric aerosol particles

The filter sampling of aerosol particles took place at the Hyltemossa field station (56°06 N, 13°25 E) (figure 3.1). It is located in the south part of Sweden and is a managed spruce forest which is a nature reserve of tree plantation. It is a combined Atmosphere and Ecosystem station. The Hyltemossa research station is part of and managed by ACTRIS. ACTRIS is the European research infrastructure of Aerosol, Clouds and Trace Gases. The INP- and aerosol measurements belong to the ACTRIS activities. Research station cottage's roof is equipped with a filter sampler insert model and manufactured with a PM_{10} inlet. The air is continuously sampled every hour by an automated filter. One cubic meter of air is sampled each hour(flow rate $1m^3h^{-1}$).

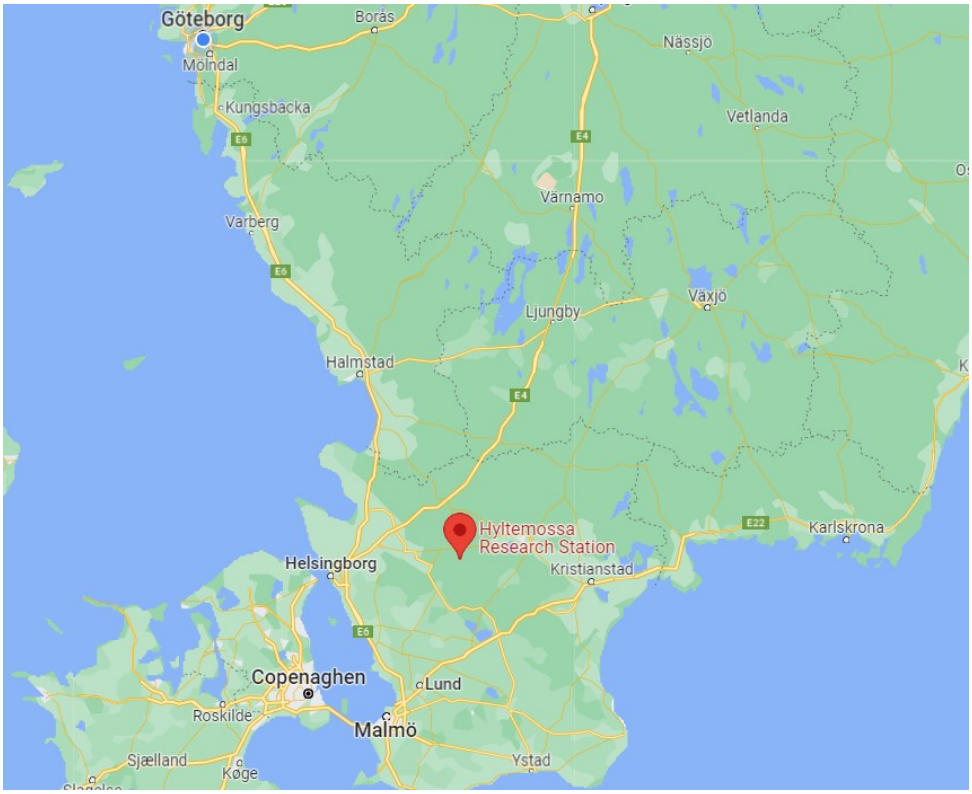


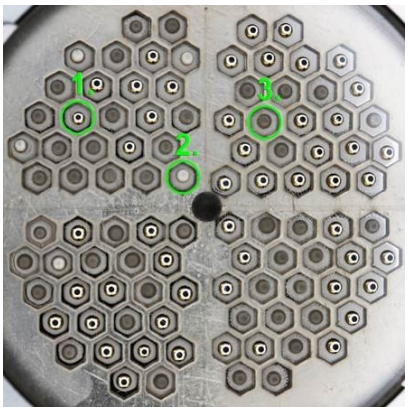
Figure 3.1: Hyltemossa Research Station. The sampling of the aerosol particles took place here (Google Maps, 2022) [5].

Samples of aerosol particles from Hyltemossa research station were taken to Lund University and from there it was taken to Gothenburg University (Atmospheric Science Division) Laboratory. In both places they were stored in a freezer to keep it safe from potential biological growth and chemical reactions. The filters are made from polycarbonate (Whatman Nuclepore Track-Etch Membrane) and have a pore diameter of 0.4 µm. Each filter collected aerosol samples for 24 hours. The filter samples that were collected from 01-03-2021 to 15-07-2021 were analyzed with a droplet freezing assay, which is described in the following section 3.1.1.

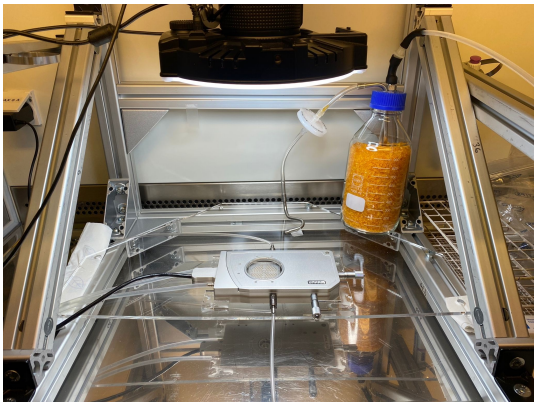
3.1.1 Droplet Freezing Assays

The droplet freezing assay (DFA) is a long been used tool to investigate ice nucleating abilities of different types of aerosol particles. An important pathway for the formation of ice crystals in mixed-phase clouds is immersion freezing, which is specifically studied with such an instrument called Droplet Freezing Assay (DFA) ((Murray et al. 2012). The basic principle of a DFA is that a liquid sample is divided into several aliquots which are then cooled in a controlled manner. During the cooling it is observed how many aliqouts are frozen at a given temperature, which later can be converted into an INP concentration at that respective temperature (see section 4.3). In our study, experimental work was conducted using a freezing apparatus based on cold-stage setups described previously and was named the Lund University

Cold-Stage (LUCS) [43]. LUCS consist of a environmental chamber with a Peltier element inside. The Peltier element is used to cool the sample aliquots. Hydrophobic glass slides are placed on top of the Peltier element. A camera is placed above the environmental chamber and takes pictures in defined intervals during the cooling process. These images are then evaluated by a custom image processing alorithm in order to determine the amount of frozen aliquots at a specific temperature. So, all the INP measurements were performed by this DFA methods in the laboratory. 100 droplets each of 1 µL was placed on the cold stage. A continuous flow of 0.3-0.4L/min of dry nitrogen was assured to minimize the fogginess and condensation during the cooling ramps. It also helps to avoid mixing of ambient air from outside to the sample droplets. The cold-stage have three parameters that can be used to determine the cooling program. The first ramp had to set at 23°C. During the placement of the drops, the temperature is held at 23℃, then it is rapidly cooled down to -5℃, held at that temperature so that also the drops reach that temperature, and then the actual measurement starts with the 2℃/min cool rate during which images are taken every 5 seconds. Through analysis of the taken photographs, we analyze the freezing pattern of the collected samples. After crystallization, frozen droplets do not reflect a circular light source Fig 3.2, (a) as the liquid droplets do, and the freezing events are recorded by the reflection of circular light.



(a) Cold Stage



(b) All the setup of Droplet Freezing Assay

Figure 3.2: Figure a) shows the cold-stage where 100 droplets are placed. Green circle 1 in unfrozen droplet and reflect the light in circular pattern, 2 is the droplet that just undergoing freezing and 3 is the totally frozen droplet . b) The whole setup of Droplet Freezing Assay. Here upside present a camera which takes the pictures after each 5 seconds.

3.1.2 Sample preparation

Samples can potentially be prepared in many ways, ranging from preparing particle suspensions, laboratory generated filter samples to environmental samples. However, as the analysis is extremely sensitive it is crucial that the samples are not contaminated to obtain sensible results. This means that all sample preparation were

conducted under highly controlled conditions and all materials were ultra-clean. All work was done in a LAF-cabinet. Clean gloves were used when handling materials and changed frequently. Sterile sample vials and pipette tips was used as well.

The standard procedure that was followed for preparing the filter samples is as followed:

First the filter cassette was opened in a LAF-cabinet and the filter was cut in half with clean tools. A sterile cryogenic vial was rinsed with ultra-pure water for 3 times and the half filter was transferred to the vial. Then 2 mL of ultrapure water was added to the vial. It is important to make sure that the filter is not folded too much and that the sampled surface is exposed to the water. The sample was then shaken for 3 minutes at high speed on a laboratory vortex shaker to disperse the sampled particles in the water. A blank sample was also prepared in the exact same way. A total of 100 droplets (each of 1 µL) were placed on the cold-stage figure 3.3. Before placing the droplets, the whole surface was cleaned with ethanol. Then four mono-layered super hydrophobic siliconized glass slides cleaned with ultra-pure water was placed on the cold-stage holder. A silicone grid with 100 cavities was placed over the slides and pressed gently down so that it seals around the edges of the hexagonal cells. By using an autopipette one sample drop (1 µL) was placed in the center of each cell. And finally, a glass lid was placed over the grid to encapsulate the droplets in the chamber. It helps to avoid interaction between the droplets with the outer environment and to minimize contamination and avoid evaporation.

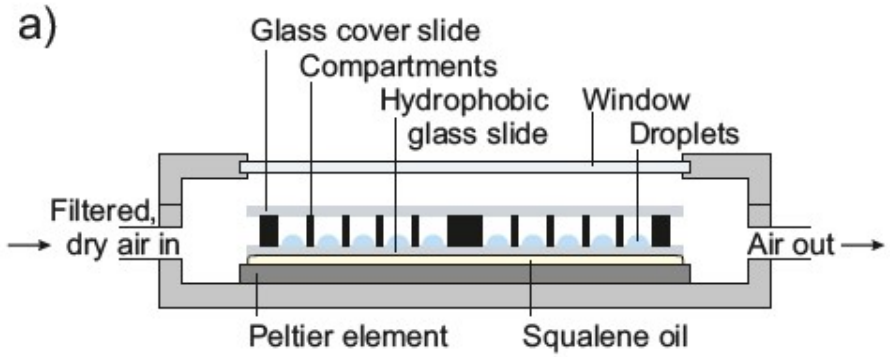


Figure 3.3: Schematic diagram of the side view of LINA cold stage device with droplet array. LUCS is little bit different from LINA. i) LUCS doesn't use the squalene oil (the hydrophobic glass slides are directly on the Peltier element) ii) instead of filtered dry air, LUCS uses nitrogen gas (N_2) This figure is courtesy of Sarah Grawe. [6]

4

Data analysis

4.1 Back Trajectories:

Back trajectories are used in order to see from where the air masses come from. Hyltemossa station is situated on the southern part of Sweden. Back trajectory is calculated to get the result of 10 days back to see the path in which it has travelled. So it help to see if the air masses originate from which part of globe (may be from the Atlantic ocean or if it is continental air-masses). Back trajectories were calculated with the HYSPLIT model (Stein et al. 2015 [7] and Rolph et al. 2017 [44]) and is presented here in figure 4.1 for the whole period of time. Here GDAS (Global Data Assimilation System) (1° grid) meteorological data source was used as input for HYSPLIT. During every 6 hours, trajectory arrivals at Hyltemossa were obtained from 150 m above ground level (AGL). So air mass can travel here from far away or from nearby areas. Generally in a sense, marine air masses contain fewer aerosol particles than the ground air. But of course depend on geographical locations eg. marine air masses from the North Atlantic and Arctic Ocean have lesser aerosol particles in comparison to air masses from Northern Europe. Not only that, INPs from ground air mass are different (as oceanic air mass bring less biological INPs) than the oceanic air INPs. It gives an idea and visualisation the relationship between different meteorological data and INP concentration. Air masses that arrived at the research station is analysed through back trajectory.

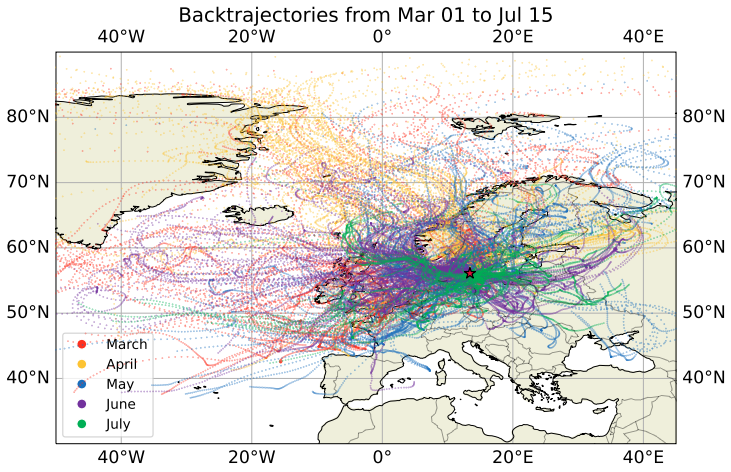


Figure 4.1: Back trajectories (5 days back) for March 1 to July 15, 2021 from the HYSPLIT model (Stein et al. 2015) [7].

Fig. 4.1 shows the back trajectories and the different colors indicate different months (red indicates March, yellow indicates April, blue indicates May, purple indicates June, green indicates July) and the red star shows where Hyltemossa is located.

4.2 Meteorological Data Analysis

The filter samples we analyze were sampled at the Hyltemossa station which is operated by two Europe wide research infrastructures: ACTRIS (The Aerosol, Clouds and Trace Gases Research Infrastructure) and ICOS (Integrated Carbon Observation System). ACTRIS is a research infrastructure build around aerosol particle measurements, while ICOS deals with greenhouse gases and carbon. Different kinds of meteorological data and vegetation data can be collected at 4 continuous plots around the main measurement tower. Several instruments at Hyltemossa Field Station can measure additional aerosol- and meteorological parameters. So a lot of meteorological data like relative humidity, air temperature, short and long wave radiation, black carbon, wind speed etc was analysed to compare with the INP data. By comparing them we can get a clearer picture of different relationships between these ambient variables. Different kinds of meteorological data analysis are shown in Appendix A.

4.3 Determination of INP

From the Droplet Freezing Assay we get thousands of pictures. Pictures were analysed by running them through Matlab codes and then through the Python. This analysis yields the number of frozen droplets in that specific temperature as a primary result. With N_{frozen}, the frozen fraction of ice (f_{ice}) can be calculated by:

Here,

$$f_{\text{ice}} = \frac{N_{\text{frozen}}(T)}{N_{\text{Total}}}, \tag{4.1}$$

$N_{\text{frozen}}(T)$ is the number of frozen droplets at temperature T and N_{total} is the total number of droplets placed on silicon grid. Then we calculate the INP concentration with the equation given by Vali (1971) [30]:

$$N_{\text{INP}}(T) = \frac{-ln(1 - f_{\text{ice}}(T))}{V}, \tag{4.2}$$

This formula (4.2) is used to calculate the concentration of INPs as a function of the frozen fraction (f_{ice}) measured at each temperature.

$$V = \frac{V_{\text{air}} \cdot (filterfraction) \cdot V_{\text{drop}}}{V_{\text{wash}}}, \tag{4.3}$$

In the above equation (4.3) V_{drop} is 1µL and V_{wash} is 2 mL. The figure 4.2 shows an example of $N_{\text{INP}}(T)$ derived for one sample with equation 4.2. INP concentration with it's freezing temperature. Here showing one single filter which represents one day sample (Here April 16, 2021). The INP concentration ranged from 1.7 m^{-3} to around 770 m^{-3} in the temperature range of -6°C. to -20°C.

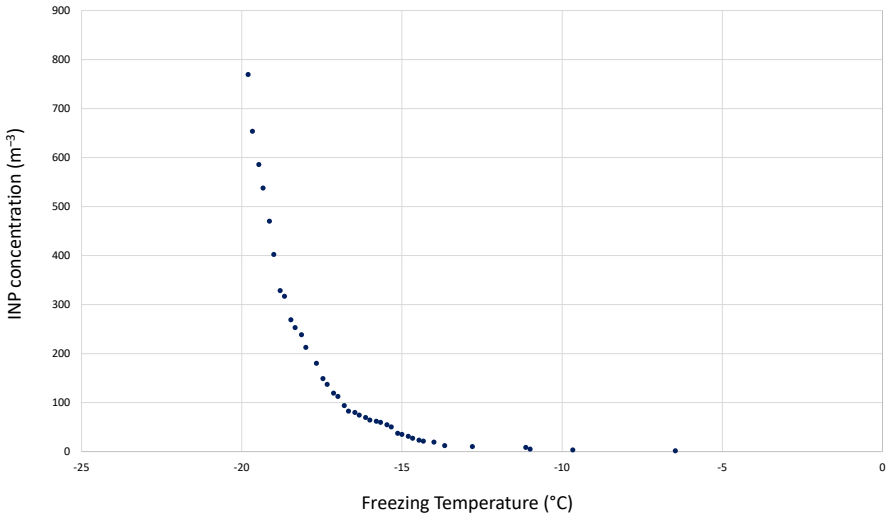


Figure 4.2: The INP concentrations from one day sample (16 April) plotted as a function of temperature.

5

Results

5.1 Freezing Spectra

INP spectra are generated from each daily INP sample. Based on this pattern, we can determine whether a sample is going to freeze at either high or low temperatures. Sometimes it is possible for a sample to start freezing at temperatures as high as -6.5°C , whether in some cases samples need very low temperature to start the freezing. Sometimes all drops during the measurement of one sample freeze before reaching -18 °C and in some cases, the samples freeze throughout the whole temperature range.

Samples were collected from March to July, 2021. Figure 5.1 shows $f_{\text{ice}}(T)$. Each curve corresponds to one of the sampled filters analyzed. The curves in Fig. 5.1 are color-coded according to the month the samples were collected (March = red; April = yellow; May = blue; June = purple; July = green). For comparison, the freezing spectra of pure MilliQ water is shown in black color (left side). Here we can see that there is a clear difference in ice nucleation temperatures for the exposed filters than the pure water standard. MillQ water freezes at much lower temperature than the filter samples. The activation curve for MilliQ water range in around -20 °C to -37 °C where freezing is observed between -5 to -26 °C for the filters. The data in Figure 5.1 is used to determine the temperature-dependent INP concentration for each filter sample.

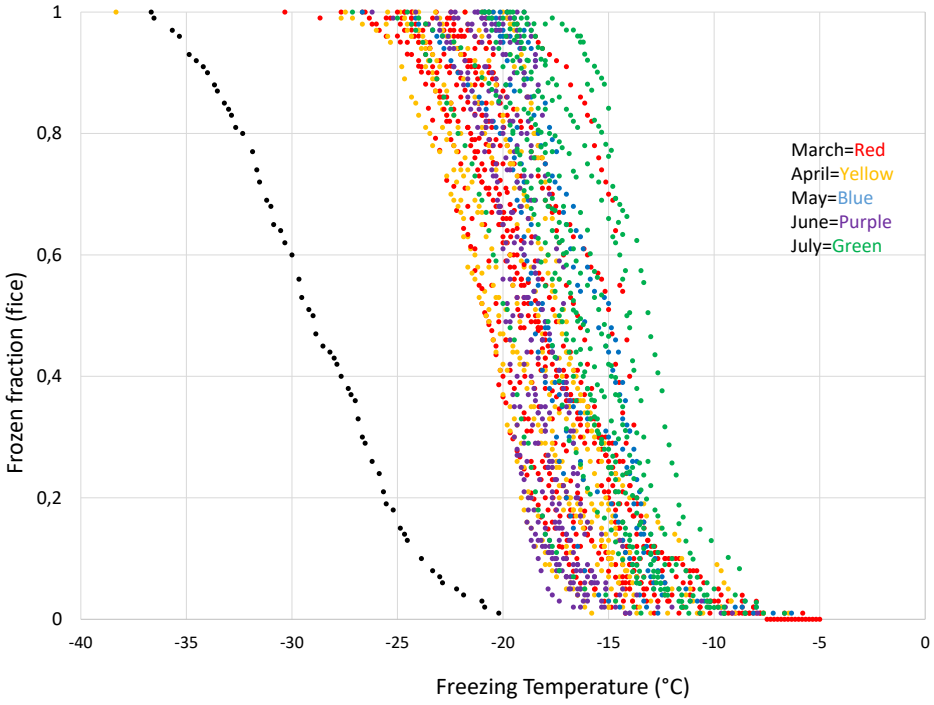


Figure 5.1: Fraction frozen of all samples collected between (March and July 2021) and pure MilliQ water as a function of temperature. Different color represent different months (March in Red, April in Yellow, May in Blue, June in Purple, July in Green).

5.2 Meteorological parameters and correlations with INP time series

We compare the INP time series with other data sets like meteorological and aerosol data to determine factors that influence INP abundance and might explain its daily and seasonal variability. Figure 5.2 shows a time series of the daily air temperature, aerosol particle concentration (diameter $\geq 0 \cdot 5\,\mu m$) measured by an optical particle counter (OPC) and INP concentration at five selected temperatures (-15°C, -17°C, -20°C, -25°C and -30°C). The air temperature and aerosol particle concentration measurements were performed at higher than daily frequency, but were down-sampled and averaged to match the frequency of the INP measurements.

The development of INP over the entire time period is shown in bottom figure 5.2 (a). There are five temperature points (-15°C, -17°C, -20°C, -25°C and -30°C) where INP concentration are split as because there is a substantial data set. Most of the freezing events occur in between these temperature range. The result doesn't indicate an overall trend like decreasing/increasing trend for INP concentration from March to July. It is rather scattered throughout the whole time.

The INP time series is compared with other meteorological data (figure 5.2 (b)

aerosol particle concentration (diameter $\geq 0{\cdot}5\,\mu m$) and figure 5.2 (c) air temperature. The lowest air temperature is -1.2 °C (March 8) and the highest temperature is 24.84 °C (June 18). The average ambient temperature is approximately 10 °C. The air temperature shows an overall upwards trend as expected as the time period was from early spring to summer during the investigated time (March to July). Average temperature in March is 1.4 °C, in April 4.9 °C, in May 6.3 °C, in June 18 °C and in July 18 °C. The strongest increase in air temperatures is observed during May to June, where it rises from 6.3 °C in May to 17.8 °C in June.

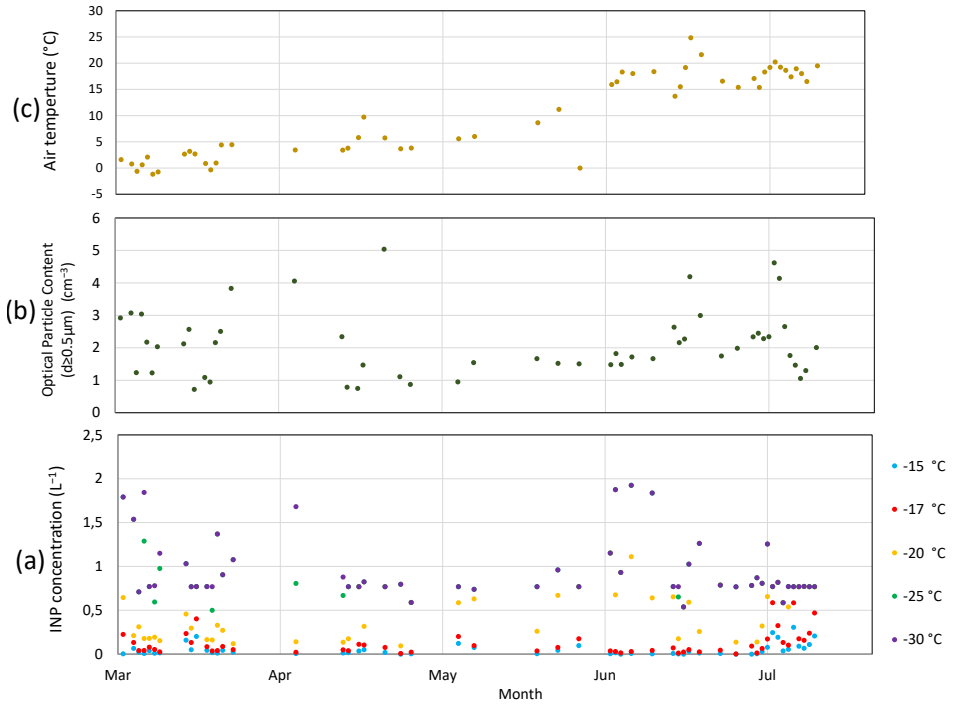


Figure 5.2: The time series of INP concentration (L^{-1}) (a) with the time series of the aerosol particle concentration (diameter $\geq 0{\cdot}5\,\mu m$) measured by (b) an Optical Particle counter (diameter $\geq 0{\cdot}5\,\mu m$) and (c) air temperature (°C).

In contrast to the air temperature time series, the time series of the aerosol particle concentration (diameter $\geq 0 \cdot 5\,\mu m$) does not exhibit an overall trend. Instead it shows short episodes of elevated concentrations 4th April, 21th April, 18th June, 4th and 5th July. The mean aerosol particle concentration (diameter $> 0.5\mu m$) over the whole investigation period is 2.11 cm^{-3}. But during the episodes with elevated concentrations, this value is increased up to a factor of five. Similarly to the aerosol particle concentration times series, also the INP times series does not show an overall trend at any of the five temperatures in whole five months.

5.3 Comparisons to INP parameterization

Based on observations of heterogeneous ice nucleation exposed to constant humidity and temperature conditions for a short time, several parameterizations have been proposed [45]. A comparison is made between the measured INP concentrations and predicted through the parameters presented by DeMott et al. 2010 [45]. In Fig. 5.3, the measured INP concentrations from experiment are plotted versus the INP concentrations predicted by a parameterization from (DeMott et al. 2010). Here different color dots represent INP concentration at different temperature and the dotted black line represents the 1:1 fit. Using the number concentration of aerosol particles with a diameter (diameter $\geq 0 \cdot 5\,\mu m$) and activation temperature to determine INP concentration, DeMott et al. (2010) [45] developed temperature-dependent parameterizations. They developed the following formula:

$$n_{\mathrm{INP}}(T_{\mathrm{k}}) = a(273.16 - T_{\mathrm{k}})^{b}(n_{\mathrm{a}\geq 0.5\mu\mathrm{m}})^{(c(273.16-T_{\mathrm{k}})+d)} \tag{5.1}$$

where a=0.0000594, b=3.33, c=0.0264, d=0.0033, T_K is cloud temperature in Kelvin (K), $n_{a\geq 0.5\mu m}$ is the number concentration (std cm^{-3}) of aerosol particles with diameters larger than 0.5 μm. In this figure all the dots are above the 1:1 fit. The predicted N_INP(T) is always higher than the measured one. And that discrepancy is especially large if measured N_INP(T) < 0.1 std L^{-1}.

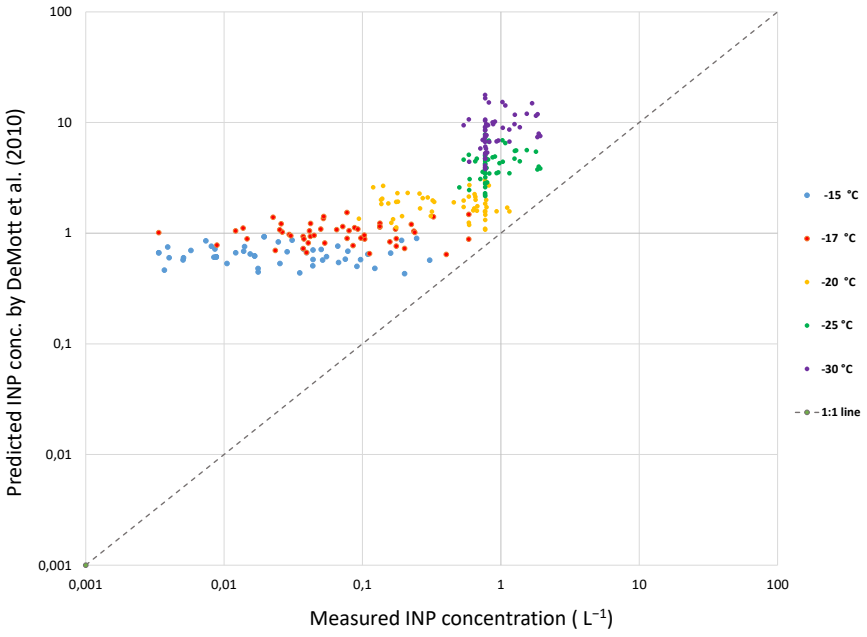


Figure 5.3: INP measured by Droplet Freezing Assay compared with INP predicted from the DeMott et al. (2010) parameterization based on particle number concentrations for diameters larger than 0.5 μm.

5.4 Frequency distribution

It is important to consider the source of INP and the degree of modification during transportation to measure the INP variability and concentration. Frequency distribution is used to measure the effect of transport. Figure 5.4 shows the relative frequency distributions of INP concentrations at (a)-15°C, (b)-17°C, (c)-20°C, (d)-25°C and (e)-30°C). In x axis there is log(N_INP) L^{-1} and y axis shows the normalised frequency in blue bars. After that, we can understand that the dilution effect is of importance by fitting a log normal distribution with the shape of INP frequency distributions. The orange line shows the fitted normal distribution (least square fit) to the frequency distribution of log (N_INP). The mean μ and the standard deviation σ of the fitted normal distribution are given in the legend of each panel. The fit for -15°C and -17°C is quite good than the other temperatures. Here we observe almost unimodal and regular bell shape curve.

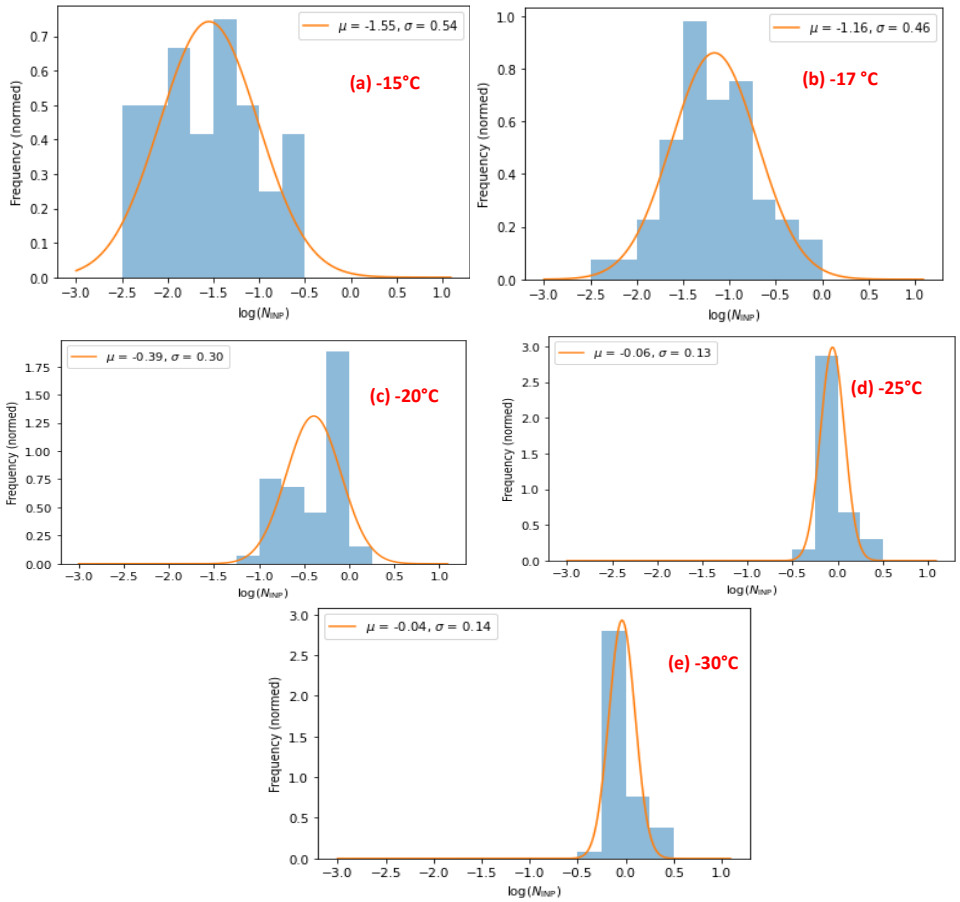


Figure 5.4: Frequency distribution plots in blue bars and fitted Gaussian distribution in red line of log (N_INP) at temperature (a)-15°C, (b)-17°C, (c)-20°C, (d)-25°C and (e)-30°C

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5.5 Back Trajectory

To investigate the sources of INP and how the air mass changed during a sampling period, backward trajectory has been done through NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model. Every 6 hours, trajectory arrivals at Hyltemossa were obtained from 150 m above ground level (AGL). After analysing the filter samples we got the most and least ice-active sample for each month (shown in table 5.1) and could compare the back trajectories associated with these samples.

Table 5.1: The days that have highest and low concentration on INP in the investigated time scale (March to July 2021)

Month	Highest Concentration	Lowest Concentration
March	16/3/2021	6/3/2021
April	16/4/2021	4/4/2021
May	5/5/2021	20/5/2021
June	30/6/2021	27/6/2021
July	4/7/2021	1/7/2021

Some of the highest and lowest concentrations days are shown in figure 5.5 and 5.6. Here the star mark is the Hyltemossa station. For example in 16 March and 5 May, almost 100% of the air masses sampled spent the majority of their time in North atlantic and greenland coast. The back trajectories for the highest concentration days indicate that most of the time air masses passes through the ground level. Here air masses approached the sampling site spending most of the time in the boundary layer ca <500 m. But for the lowest ice active samples spent most of the time above the boundary layer.

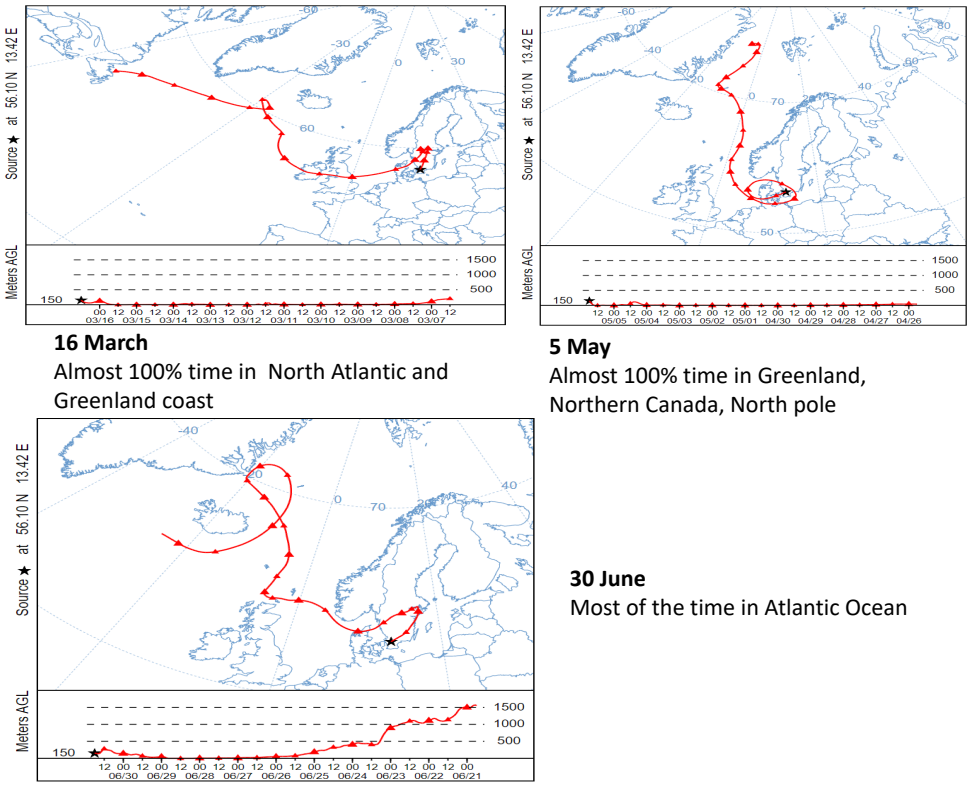

Figure 5.5: Some of the highest concentration days with the areas that air mass travels back 10 days.

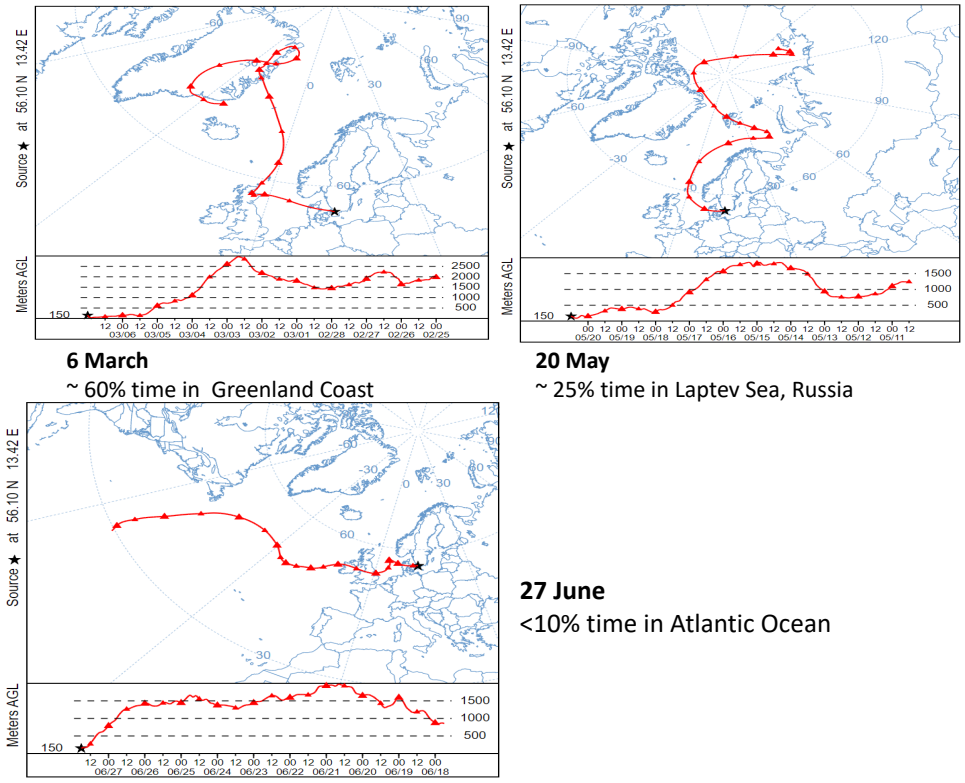

Figure 5.6: Some of the lowest concentration days with the areas that air mass travels back 10 days.

5.6 Parameterization

Figure 5.7 shows the temperature-dependent concentrations of INP over the the time period March - July 2021. The selected INP parameterizations are also depicted in this figure. Here different shaded regions represent DeMott et al. 2010 [45] (green shaded area) where the maximum and minimum aerosol particle concentration ($d \geq 0.5\mu m$) that was observed during the investigation period was used to show the full span of predicted $N\_INP(T)$. The parameterization by Schneider et al.(2021) [10] is shown in purple shaded area where minimum and maximum air temperature is used. DeMott parameter is in equation 5.1. The Schneider parameterization is as follows:

$$c_{\mathrm{INP}} = 0.1exp(a1 - T_{\mathrm{amb}} + a2)exp(b1T + b2) \tag{5.2}$$

Here, $a1 = 0.074 \pm 0.006k - 1, a2 = -18 \pm 2, b1 = -0.504 \pm 0.005k - 1, b2 = 127 \pm 1$; with the activation temperature T and ambient air temperature T_{amb} in kelvin. Here Schneider et al.(2021) parameterization fits well with our result. However, it overestimates the INP concentrations measured in some samples at colder temperatures as well. Conversely, the DeMott et al.(2010) [45] parameterization clearly over estimated at the higher temperature and as a result it does not incorporate the behavior of ice nucleation observed in droplet-freezing experiments. The temperature when

the ice nuclei is activated is compared by Fletcher parameterization (developed by Fletcher 1962 [46] presented in dark black dotted line). Several ice nucleation studies use the parametrization as a reference.

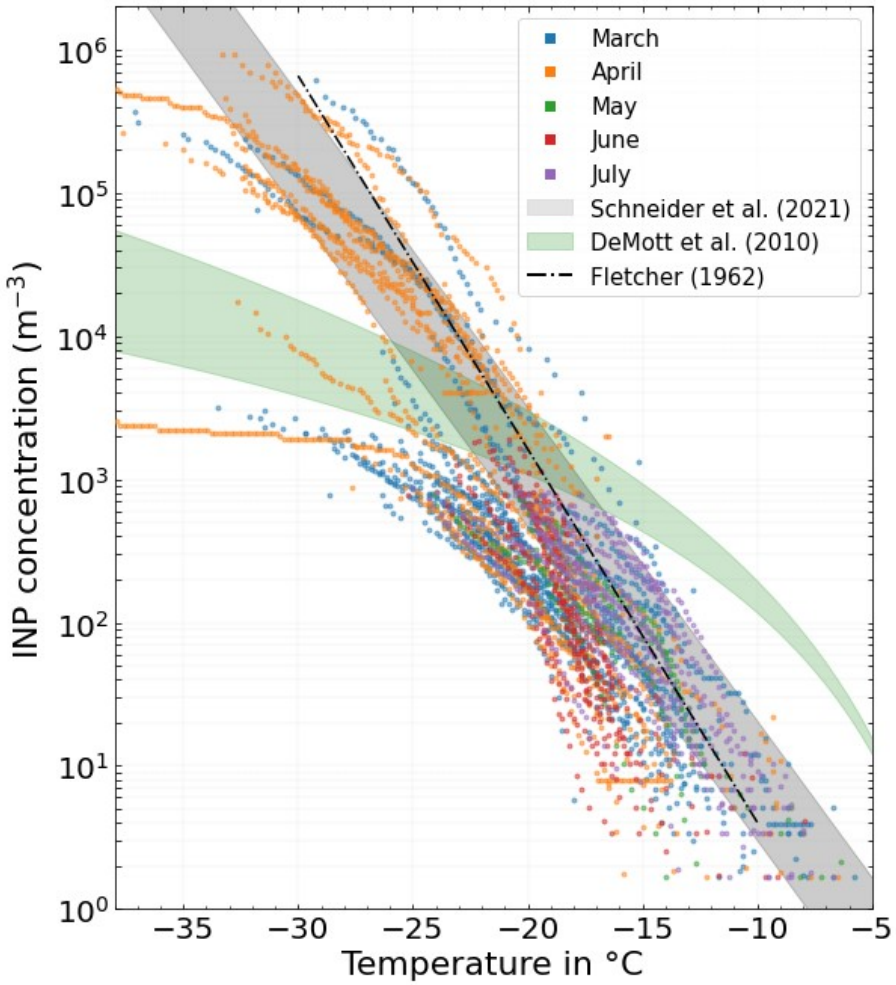

Figure 5.7: INP temperature spectra from all the filters measured from March- July 2021.

6

Discussion

From the frozen fraction spectra figure 5.1 we can see that, the activation curve for MilliQ water range is around -20 °C to -37 °C whereas freezing is observed between -5 to -26 °C for the filters. So we can say that there must be some number of Ice Nucleating Particles in the filter sample spectra. Because they freeze in higher temperature than the MilliQ water spectra and we use this spectra to calculate the INP concentration which is in the time series figure 5.2.

In figure 5.2 we compare aerosol particle concentration ($d \geq 0.5\mu m$) and air temperature with INP concentration. From visual inspection between these we don't see any trends or correlation that immediately emerged. We can see the air temperature shows an overall upwards trend as expected as the time period was from early spring to summer during the investigated period (March to July). But interestingly we don't see any strong dependence that jumps out from the aerosol particle concentration ($\geq 0.5\mu m$) related to temperature. A reason for that could be that the INP population at Hyltemossa contains a high fraction of INP smaller than 0.5µm. With the available data this hypothesis cannot be confirmed or denied, but such cases are reported in the literature. For example Mason et al.(2016) [47] conducted a study where he found that,in Alpine region larger particles did not seem to influence INPs. Towards colder measurement temperatures around -25°C a considerable fraction of INPs were <0.5 µm. Similarly we don't see any trend with INP concentration as well that clearly related to either any of them. So we can say that, our result is consistent with the results from Fountain and Ohtake (1985) [48]. They analysed different meteorological parameters including air temperature, precipitation, fog, and wind direction but did not find any correlations with INP concentrations at any freezing temperatures. As compared to Radke et al. (1976) [49], who measured INP concentrations in Utqiagvik, Alaska, during March, he stated that local weather condition affect the INP concentration.

In figure 5.3 we see that the parametrization by DeMott et al. (2010) [45] always overestimates the INP concentration. Reasons could include different investigation regions as he combined data from nine field studies from a variety of locations. DeMott mostly sampled over the continental USA, Canada, the Amazon Basin and over the Pacific Ocean. And these regions are not necessarily comparable to Southern Sweden. The parameterization was empirically based on temperature below 15°C. It

should be also mentioned that the data sets used by DeMott are from different times of the year (they took 14 year time period data), whereas our data set only covers the time from March to July. A final difference to DeMott is the applied method. DeMott used data originates from CFDC (continuous flow diffusion chamber) measurements which is a method that is not very sensitive for low INP concentrations roughly 1 per L and lower, while the present study used DFAs. We speculate that the lack of consideration of field measurements in our kind of environment which is totally different from DeMott, might be the main reason why their parameterization is not able to match with ours.

Ott (1990) [50] showed with the example of pollutants in the environment that a concentration resulting from successive random dilutions tends to be log normally distributed. Welti et al. (2020) [51] transferred that concept to INPs: For INP concentratin at a specific T, a log normal distribution is to be expected if the sampled air mass was transported over longer distances and hence experienced several random mixing events along the way. This means that deviations from a log normal distribution can be attributed to either additional sources for INP active at the selected T, that are somewhat closer to the measurement location or the mixing of clean air masses without any INPs. In figure 5.4 the coldest temperature (i.e. T < -20°C) only reveal the counting limit of the method as because we freeze everything in that time. This can be seen in the figures as the tall bar at approx. $\log(N_{inp}) = 0$, which corresponds to the highest measurable INP concentrations with the setup used in this study. At -17°C the frequency distribution matches well a log-normal distribution. This indicates towards long range transport and well mixed air masses. For the warmer temperature (-15°C) we might get deviation from log-normal distribution, especially at the left and right tail of the distribution. The more frequent occurrence of low INP concentrations (deviation at the left tail) can be seen as hint that more clean air masses without INP were transported to the sampling site. On the other hand, the deviation at the right tail of the distribution, shows the more frequent occurrence of high INP concentrations. This could be related to local sources that inject INP into the atmosphere.

Comparing the back trajectories associated with the most ice-active and least ice-active of each month showed that high ice activity typically occurs when the sampled air masses spent most of the time close to the ground. Low ice activity was observed when the sampled air masses travelled primarily in several kilometers height. This suggests that highly ice active INP are coming from more regional sources rather than long-range transport.

In figure 5.7 Fletcher 1962 [46] parametrization is used as a reference here. Another parameterization by Schneider et al. (2021) capture overall trend. Temperatures between -12°C and -25°C, for which Schneider's parameterization was developed, give Schneider's parameterization better performance. But it also overestimates for the INP concentrations measured at colder temperatures. On the other hand, DeMott et al. (2010) [45] capture only few data points and clearly over estimated at the higher temperature. One reason can be the experiment was conducted in very

different environments than ours and thus bias can be expected. For Schneider et al. (2021), they measured in the similar environment as ours. Their research area was Boreal forest of Hyytiäla, Finland and in our case it is mixed coniferous forest dominated by Norway spruce in Southern Sweden.

7

Conclusion

From our above discussion we can make some conclusions. There were no obvious trends, annual cycles, or well-defined peak concentrations in the time series. Quantitative INP predictions face new challenges due to the observational data we have presented, as seasonal trends must be taken into account in order to be accurate.

If we say about the measurements from the inter-comparison with different parameterization, it might not giving any representative outcome as we have limited data. The Schneider et al.(2021) [10] parameterization is giving better fit but DeMott et al.(2010) [45] every time overestimates the INP concentration at higher temperature as because his kind of methodology is different from ours and it cannot capture the ice nucleation behaviour by our Droplet Freezing Assay method.

The relative frequency of observed INP concentrations matches well a log-normal distribution at -17°C which suggest that INPs being long-ranged and well mixed within sampled air masses. For the warmer temperature (-15°C) we might get deviation from Log-normal distribution, especially at the left and right tail of the distribution.

In addition to that, INP distribution vary depending on types and sources. As a result continuous measurements do not accurately reflect either of their short-term variability or the long term seasonal cycle. For getting a strong seasonal variability of INP concentration, one need to take a larger time scale which is absent in our study. So a comprehensive causal link between INP concentration with seasonal variability remains unclear.

Last but not the least, it is my hope that lessons can be learned from my small but unique measurements that will assist in future INP monitoring efforts.

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Abbreviations

BC Black Carbon

CCN Cloud Condensation Nuclei

CR Cooling Rate

DOY Day Of Year

ICOS Integrated Carbon Observation System

IN Ice Nucleation

INP Ice Nucleating Particle

IPCC Intergovernmental Panel on Climate Change

OC Organic Carbon

PM Particulate Matter

RH Relative Humidity

VOCs Volatile Organic Compounds

A

Appendix A

Different meteorological parameter analysis. Time scale March-July 2021. Data obtained from www.icos-sweden.se/hyltemossa.

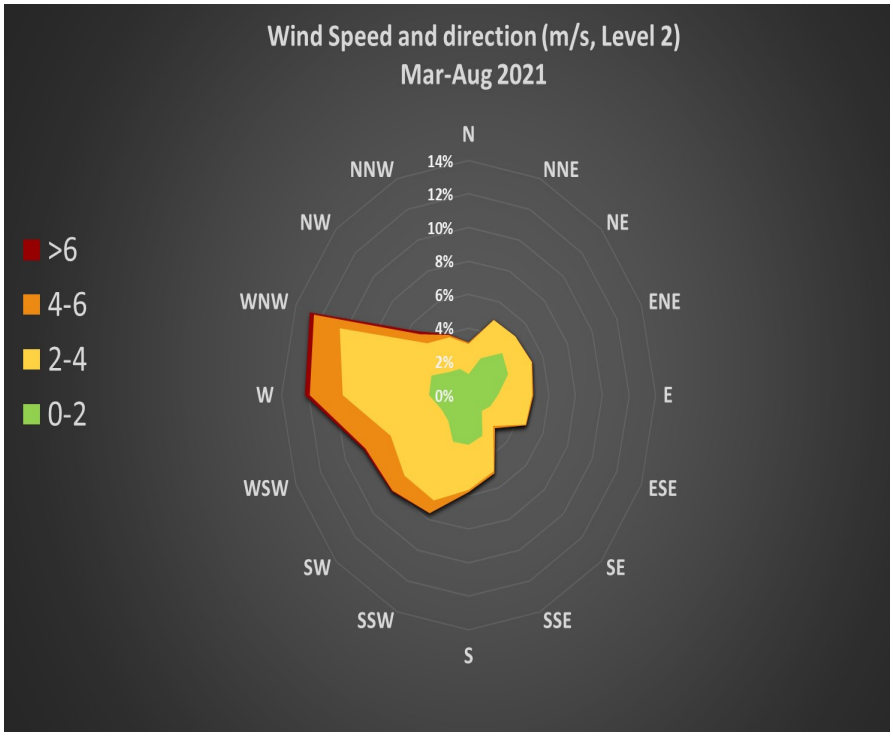

Figure A.1: Wind direction from March to July 2021.

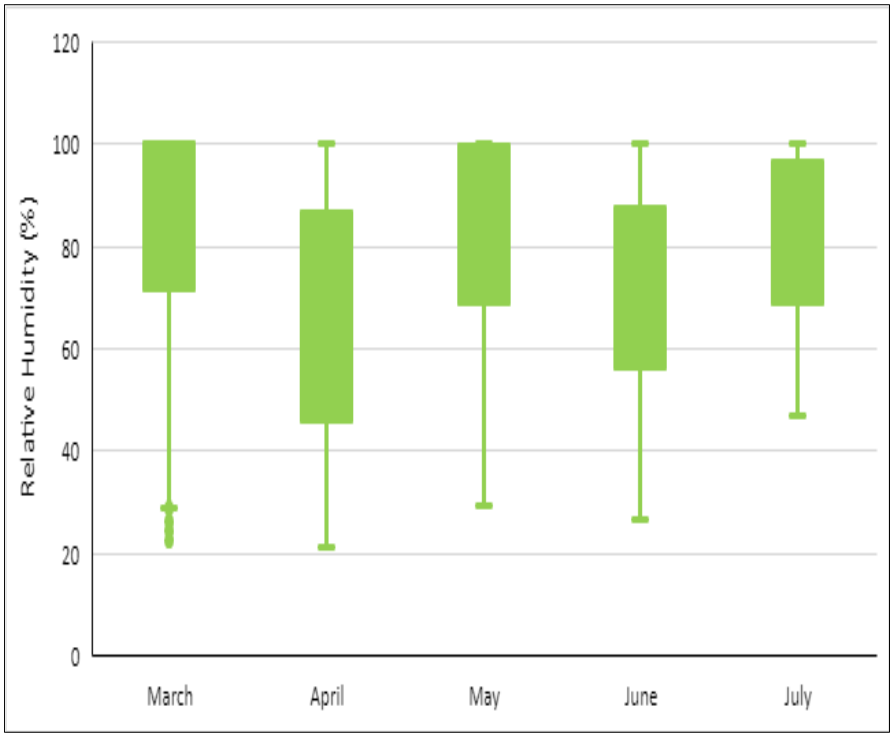

Figure A.2: Relative Humidiity (%) from March to July 2021.

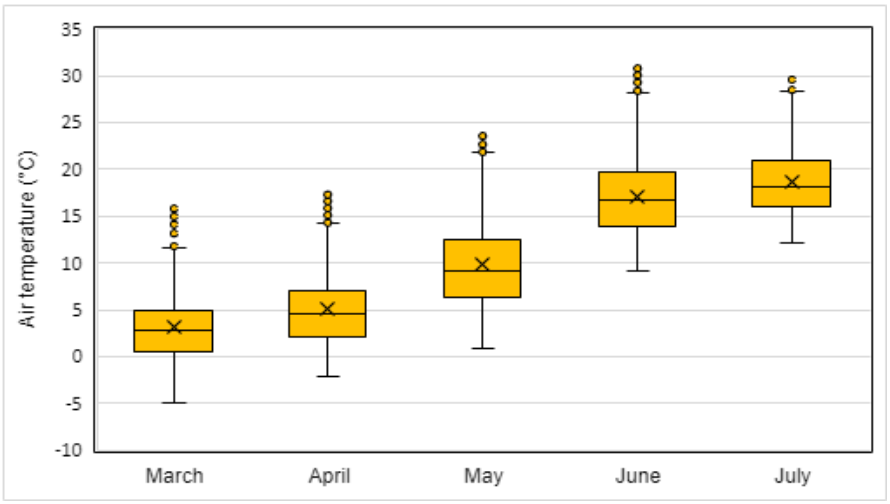

Figure A.3: Air temperature (°C) from March to July 2021.