TÉCNICO LISBOA

Seasonality of fluorescent particles in the Arctic

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Preface

The work presented in this thesis was performed at the Extreme Environments Research Laboratory (EERL) of the École Polytechnique Fédérale de Lausanne (EPFL) in Sion, Switzerland, between February 2021 and October 2021, under the supervision of Prof. Dr. Julia Schmale and Co-supervision of Prof. Dr. Arsénio Fialho from the home university, Instituto Superior Técnico, University of Lisbon, Portugal. Initially, this project aimed the investigation of the biogenic sources, seasonality, and spatial variability of Ice Nucleating Particles in the Southern Ocean, during the ACE-SPACE expedition. This work involved going to the United Kingdom and to Germany, as well as Switzerland. The pandemic situation made this project nearly impossible to happen, or at least very uncertain. Because of this, the scope of the project was changed to a most recent campaign (MOSAiC), this time in the Arctic, with a different focus (fluorescent particles).

Declaration

I declare that the work contained in this document is original and of my authorship and that it fulfills all the requirements of the Code of Conduct and Good Practices of the Universidade de Lisboa.

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Firstly, I would like to thank my supervisors, Professor Doctor Julia Schmale and Professor Doctor Arsénio Fialho, for all the patience, guidance, wisdom, knowledge, support and kindness given throughout this past year.

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Abstract

One of the most sensitive regions to climate change is the Arctic and these alterations are coming faster than ever and with a great intensity (Serreze & Barry, 2011). The surface air temperature, which is one of the most important variables to indicate these changes, increased two times faster in the Arctic comparing to the rest of the globe, since the mid of the 20th century. This phenomenon is called Arctic Amplification. Atmospheric particles of biological origin, also referred to as bioaerosols or primary biological aerosol particles (PBAP), have importance in environmental systems. For example, bioaerosols play a role on the cloud formation. Recently, there has been an increase in the frequency of scientific publications using instruments based on ultraviolet laser/light-induced fluorescence (UV-LIF), such as the WIBS (wideband integrated bioaerosol sensor) or UV-APS (ultraviolet aerodynamic particle sizer), for bioaerosol detection both outdoors and in the built environment. The WIBS was used in the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) to measure fluorescent and total particle concentrations and particle size and shape. This data was analyzed and some of the final conclusions were that total particle concentrations are usually two orders of magnitude higher than the fluorescent particle concentrations. Moreover, both total and fluorescent particles have their highest median values of concentration in the December-January-February (DJF) and March-April-May (MAM) seasons, and the concentration peaks are reached in January and February. August and October are months with low values of concentrations. Regarding particle sizes, the conclusion to be taken is that fluorescent particles are generally the same size as non-fluorescent particles. Particle fractions were calculated, reaching the conclusion that AB type particles are clearly dominant in polluted periods. Finally, wind was seen as a possible mechanism to lift snow and sea particles, making them possible local sources of fluorescent bioaerosols.

Keywords:
Arctic Amplification
Bioaerosols
Fluorescent Particles
MOSAiC
WIBS

Resumo

Uma das regiões mais sensíveis às alterações climáticas é o Árctico e estas alterações estão a chegar mais rapidamente do que nunca e com uma grande intensidade. A temperatura do ar à superfície, que é uma das variáveis mais importantes para indicar estas alterações, aumentou duas vezes mais depressa no Árctico em comparação com o resto do globo, desde meados do século XX. Este fenómeno chama-se Amplificação do Árctico. As partículas atmosféricas de origem biológica, também referidas como bioaerossóis ou partículas de aerossóis biológicos primários (PBAP), têm importância nos sistemas ambientais. Por exemplo, os bioaerossóis desempenham um papel na formação de nuvens. Recentemente, tem havido um aumento na frequência de publicações científicas que utilizam instrumentos baseados em laser ultravioleta/ fluorescência induzida pela luz (UV-LIF), tais como o WIBS (wideband integrated bioaerosol sensor) ou UV-APS (ultraviolet aerodynamic particle sizer), para a detecção de bioaerossóis, tanto no exterior como no ambiente construído. A WIBS foi utilizada no Observatório Multidisciplinar para o Estudo do Clima Árctico (MOSAiC) para medir concentrações de partículas fluorescentes e totais e tamanho e forma das partículas. Estes dados foram analisados e algumas das conclusões finais foram que as concentrações totais de partículas são normalmente duas ordens de magnitude mais elevadas do que as concentrações de partículas fluorescentes. Além disso, tanto as partículas totais como as fluorescentes têm os seus valores medianos de concentração mais elevados nas estações Dezembro-Janeiro-Fevereiro (DJF) e Março-Abril-Maio (MAM), e os picos de concentração são atingidos em Janeiro e Fevereiro. Agosto e Outubro são meses com baixos valores de concentração. Relativamente às dimensões das partículas, a conclusão a tirar é que as partículas fluorescentes são geralmente maiores do que as partículas não fluorescentes. As fracções de partículas foram calculadas, chegando à conclusão de que partículas do tipo AB são claramente dominantes em períodos poluídos. Finalmente, o vento foi visto como um possível mecanismo para levantar partículas de neve e do mar, tornando-as possíveis fontes locais de bioaerossóis fluorescentes.

Palavras-chave:
Amplificação Árctica
Bioaerossóis
Partículas Fluorescentes
MOSAiC
WIBS

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Abbreviations

AA	Arctic Amplification
BC	Black Carbon
CCN	Cloud Condensation Nuclei
CMIP5	Coupled Model Intercomparison Project Phase 5
CPC	Condensation Particle Counter
DJF	December – January – February W0
IN	Ice-forming Nuclei
INP	Ice nucleating particles
JJA	June – July – August
MAM	March – April – May
MOSAiC	Multidisciplinary drifting Observatory for the Study of Arctic Climate
MPC	Mixed-phase Cloud
NADH	Nicotinamide Adenine Dinucleotide
OPC	Optical particle counter
PBAP	Primary biological aerosol particles
PSL	Standard monodisperse polystyrene latex
SON	September – October – November
SSA	Sea-spray aerosol
UV-APS	Ultraviolet aerodynamic particle sizer
UV-LIF	Ultraviolet laser/light-induced fluorescence
WIBS	Wideband integrated bioaerosol sensor
WIBS-NEO	Wideband Integrated Bioaerosol Sensor-New Electronics Option

1. Introduction

1.1. Aerosols and their importance. Why in the Arctic?

Several decades ago, the importance of aerosols started to be recognized as scientist came to realize they have a role in the oxidative capacity of the atmosphere, as well as cloud condensation (CCN) and ice-forming nuclei (IN). They can have some global and local impacts like climate change, toxicity and health hazards. Investigating the sources of this aerosols and how they can vary with time and space, has been an increasing need in these last few decades even though, particularly bioaerosols and their physical and chemical atmospheric processes, are still poorly understood.

Airborne particles or aerosols are important because they can directly (by absorbing or scattering radiation) or indirectly (related with their ability to form or act as CCN and IN, and hence lead to the formation of clouds, thus indirectly influencing the Earth's radiation budget) impact the Earth's climate. The properties of CCN and IN also impact rain formation. Precipitation regulates the wash-out of aerosols from the atmosphere. Therefore, aerosol particles can potentially affect the water cycle, and even agriculture and human health due to their chemical properties (Ariya et al., 2009).

One of the most sensitive regions to climate change is the Arctic and these alterations are coming faster than ever and with a great intensity (Serreze & Barry, 2011). The surface air temperature, which is one of the most important variables to indicate these changes, increased two times faster in the Arctic comparing to the rest of the globe, since the mid of the 20th century (Overland et al., 2011; Serreze & Barry, 2011). This phenomenon is called Arctic Amplification. Arctic has some peculiarities like the extended snow and ice cover, low elevation of the atmospheric boundary layer and abundance of low-level clouds that together with multiple feedback mechanisms (surface albedo, water vapor or cloud feedback) contribute to the increasing climate sensitivity of the Arctic. The relevance, strength and interconnection of these peculiarities and feedback mechanisms are yet unclear (Pithan & Mauritsen, 2014; Serreze & Barry, 2011).

Clouds are one of the key factors of Arctic Amplification since they affect the energy budget of the Arctic boundary layer. By reflecting long-wave radiation, they tend to warm the surface and therefore lead to sea ice melting (Vavrus et al., 2011), which will enhance the evaporation and cloud formation. This feedback will probably accelerate in the future**.** As the ice cover reduces, biological activity increases in the marine and terrestrial environment. This is related with the alteration of aerosol particle sources, that may affect clouds and their properties (Hartmann et al., 2019).

In order to comprehend more about the Arctic cloud feedback, some studies were made using the output of the Coupled Model Intercomparison Project Phase 5 (CMIP5). Pithan and

Mauritsen (Pithan & Mauritsen, 2014) analyzed these outputs and came to the conclusion that there was an overall small positive Arctic cloud feedback, with individual models disagreeing on the sign. On the other hand, another study another study based on the same exact models concluded that the feedback was clearly negative (Schmale et al., 2021; Zelinka et al., 2013).
The results of these studies indicate that our current knowledge on the AA (Arctic Amplification) is incomplete and limited by the absence of robust model representations of regional Arctic feedback processes, along with other things. Due to the simplification of some model descriptions, potentially key processes ca be missing, making it impossible apprehend the full range and the relative importance of different drivers of AA. Considering Arctic aerosols and their role in cloud formation, this matter is particularly true. When trying to simulate low-level Arctic mixed-phase clouds, comprising the transition between cloudy and clear states, large-scale models demonstrate difficulties (Morrison et al., 2012; Schmale et al., 2021). Part of this difficulty may stem from the insufficient representation of aerosol particles, or more specifically, the subset of the aerosol population acting as ice nucleating particles (INP) and cloud condensation nuclei (CCN), which under certain conditions are a limiting factor in Arctic cloud formation (Mauritsen et al., 2011; Schmale et al., 2021). One of the challenges for these models is to represent the multiple different aerosol processes of significance in the Arctic. For instance, summer in the Arctic is dominated by local secondary aerosol sources, while wintertime there is preeminence of wind-driven and transported aerosols (Schmale et al., 2021). In addition, aerosols contribute to AA by altering cloud properties and they also interact directly with radiation through scattering and absorption. As an example of the last phenomenon, black carbon and mineral dust particles may reduce the surface albedo, accelerating the ice and snow melting in spring and summertime (Schmale et al., 2021).

While in the past most of the attention on aerosol climatic effects went to long-range transported anthropogenic pollution (Arctic Haze) (Quinn et al., 2007; Schmale et al., 2021), nowadays, an emphasis on the inner-Arctic is starting to appear, especially when it comes to natural aerosol sources.

1.2. Fluorescent aerosols and their importance

Atmospheric particles of biological origin, also referred to as bioaerosols or primary biological aerosol particles (PBAP), have importance in environmental systems. Recently, there has been an increase in the frequency of scientific publications using instruments based on ultraviolet laser/light-induced fluorescence (UV-LIF), such as the WIBS (wideband integrated bioaerosol sensor) or UV-APS (ultraviolet aerodynamic particle sizer), for bioaerosol detection both outdoors and in the built environment. Some problems, despite all the efforts, have occurred when it comes to characterize the particles and understand which are from biological origin and which are not. These gaps in the current knowledge are caused by the detection ability of LIF

instrumentation (Moallemi et al., 2021; Savage et al., 2017). The wavelength detection ranges are chosen to match regions of fluorescence for biological compounds that are found ubiquitously in PBAP, such as tryptophan and Nicotinamide Adenine Dinucleotide (NADH) (Savage et al., 2017).

Changing the particle fluorescence threshold was shown to have a significant impact on fluorescence fraction and particle type classification. However, raising the fluorescence threshold has no impact in reducing the relative fraction of biological material considered fluorescent but can significantly decrease the interference from mineral dust and other non-biological aerosols. Some examples of highly fluorescent interfering particles are brown carbon, soot and cotton fibers (Savage et al., 2017).

1.2.1. Contributors to fluorescent signals: biological particles and interference from dust and Black Carbon

Bioaerosols are airborne particles or large molecules that range between 1 - 10 nm and 100 μm diameter and originate from multiple sources in nature. They are considered a subgroup of biogenic organic aerosols. They can be found alive, dead, dormant or like products released from living organism. Some examples of these particles are bacteria, viruses, fungi, metabolites, pollen, cell debris and biofilms, and these might be emitted by biogenic sources such as oceans, vegetation, soils, lakes, and living organisms (Ariya et al., 2009). A consistent classification of species is not possible since these aerosols can be derived from primary or secondary biogenic sources. It is yet to be known if they have a major contribution in the overall organic aerosol budget. In the likes of other aerosol particles, the ice nucleation ability varies depending on the type of particle and the importance they have changes with the tropospheric concentrations (Ariya et al., 2009).

Although it has been known that there are various types of bioaerosols in indoor air, the troposphere and the stratosphere, it was only until recently that it was found that about 25% of the particles suspended in air (m/m) are primary organic aerosol of biological origin, with some regions registering biological particles concentration of up to 74% (Ariya et al., 2009), (Matthias-Maser et al., 2000). The concentration of these particles varies with the temperature, radiation, relative humidity, rainfall, and wind speed and direction, as well as other seasonal factors such as fungal spores or airborne bacteria concentration (gram-positive and gram-negative), in the form of single spores or clusters. These are mostly common during spring and fall, as the majority of microorganisms are spread through the fall of leaves. There is evidence of these airborne particles being covered with a mucus-like material, strengthening the theory that suggests that residue of microbial films allow for transportation and survival means in the air. Other environmental conditions may influence the concentration of each organism, such as pH (acidic

favors the formation of fungi, neutral facilitates the presence of a great diversity of microorganisms).

Biological particle signatures have already been detected in some ice residues sampled from clouds, but it is still uncertain what is their impact concerning regional and global scale cloud formation. There are many evidence supporting the effectiveness of some aerosols and their IN capacity. For instance, it has been shown through some studies, using sum-frequency-generation spectroscopy and molecular dynamics simulations, that the active sites of the ice nucleation protein (inaZ) that can be found on the outer cell membrane of *Pseudomonas syringae* bacteria report a unique hydrophilic–hydrophobic pattern that supports the organization of the close water molecules and leading this way to ice nucleation (Pandey et al., 2016).

While the majority of bioaerosol studies focused on continental sources and on the cells or fragments of biological particles, there is new evidence suggesting that oceans are a significant source of biogenic INPs and that the exudates from marine microorganisms could nucleate ice under cirrus clouds and mixed-phase clouds (MPCs) conditions, with the ice-active material having sizes on the order of tens of nanometers, potentially being macromolecules (Ladino et al., 2016; Wilson et al., 2015).

Even though the bioaerosol emission rates are orders of magnitude smaller than dust particles (Després et al., 2012), a small fraction of some types may freeze at much higher temperatures than dust. This can increase their impact in the formation of ice in clouds through subsequent secondary ice processes (Field et al., 2016).

Throughout the years, some observational studies concerning ambient INPs, have been made in coastal and oceanic regions. When comparing number concentrations and surface site densities of aerosols obtained in some marine environments to those generated in the laboratory (where sea spray aerosols were the only INP source), the results implied that marine INPs are less efficient than terrestrial INPs.

Sea Spray Aerosols (SSA) properties have been vastly investigated in natural environments and in laboratory. The conclusion of this studies was that the characteristics of these primary marine aerosols depends on the organic matter content in the bulk seawater and of the wave breaking dynamics. In the laboratory SSA, the inference was that the characteristics were strongly dependent on the generation technique (Collins et al., 2014; Stokes et al., 2013).

While SSA properties are dependent on the production technique (laboratory) and on the different environments, some chemical and physical characteristics can be applied generally: The size distribution of SSA is studied to be log-normally distributed and centered at approximately 100-200 nm. The chemical composition is dependent on particle size, so supermicron SSA is dominated by sea salt particles, whereas submicron SSA contains more organic molecules (mostly exudates from phytoplankton and their breakdown products), gels (Orellana et al., 2011) and viruses, with the highest organic fraction found at the smallest sizes (Facchini et al., 2008; O'Dowd et al., 2004).

Furthermore, it was found that up to 17% of laboratory-generated supermicron SSA particles had chemical signatures indicative of microbes and their constituents (Prather et al.,

2013) and many of these constituents (including marine microbes and organic exudates) have been considered as potential INPs in SSA (Burrows et al., 2013). However, few studies have yet directly examined the previous constituents and the way they add to the marine INPs.

There is some evidence that phytoplankton might have a potential role in changing SSA chemical composition in a way that increases INP production (C. S. McCluskey et al., 2018).

Spatial correlations suggest that regions of oceanic upwelling, likely regions of phytoplankton blooms, are linked to an enrichment of INPs. Proof for marine source of biological INPs has been given by aircraft measurements made over the Arctic (D. C. Rogers et al., 2001) and filter collections on a ship in the high Arctic (Bigg, 1996).

Further ahead, demonstrations of the oceanic biological activity potential to engender INPs were made when seawater samples collected succeeding a phytoplankton bloom, had a bigger concentration of INPs than the seawater collected in other places (Christina S. McCluskey et al., 2017). Higher ice nucleation activity in phytoplankton-rich seawater compared to seawater containing low phytoplankton concentrations, ultimately leads to the deduction that biogenic INPs are emitted from biologically active ocean waters (C. S. McCluskey et al., 2018).

Marine INPs could be major contributors to the INP population in remote marine environments, according to some of the existing modeling studies that state that there is a linear relationship between organic matter and ice nucleation activity (Burrows et al., 2013; Items et al., 2015). Nevertheless, other studies have also illustrated that the climate-relevant properties of clouds in global model simulations are truly sensitive to modifications in the ice nucleation efficiency of marine aerosols. This means that, in order to properly evaluate these relationships, atmospheric aerosol measurements must be made, and it is also essential that these measurements take place in several ocean environments and in all seasons.

Mineral dust has been recognized as a major contributor to atmospheric ice nucleation at temperatures relevant for mixed phase and cirrus clouds (Heintzenberg et al., 1996). During large Saharan dust outbreaks, some results of models' analysis suggest that dust aerosol concentrations can reach 107 m^{-3} over Europe (Bangert et al., 2012), but it also appears that dust dominates the normal background ice nucleating particle (INP) and ice residual composition in the absence of these large dust events (Hande et al., 2015)**.** This knowledge leads to the notion that dust can have an important indirect effect on clouds (Sassen, 2002; Sassen et al., 2003) on seasonal timescales. Other important ice-nucleating aerosols are soot and biological particles (Hande et al., 2015), however their contribution to ice nucleation is on average lower than that of dust (Hoose et al., 2010). Case studies of the impact of dust events on INP concentrations in Europe have been performed (Chou et al., 2011; Mamouri & Ansmann, 2015); however, climatological estimates of dust number concentrations and the resulting INP concentrations, as well as an understanding of their seasonal variability, remain elusive.

Between all the studies involving dust as a source of INP, some of the conclusions are that the maximum median dust concentrations are around 3 × 105 m^{-3} during spring, with about an order of magnitude lower number concentrations in summer. There is a significant amount of variability in dust concentrations. The resulting potential immersion INPs reach maximum median

concentrations of 9.5×104 m^{-3} during spring. During the summer months concentrations are lower and occur at a higher altitude compared to all other months (Hande et al., 2015).

Conventionally, black carbon (BC) or soot is considered as the main light absorber in atmospheric aerosols over the spectrum ranging from ultraviolet to infrared (Wu et al., 2020). Soot is a black material found in smoke from wood and coal fires and it has been seen as the main representative air pollutant throughout history (Andreae & Gelencsér, 2006). It's made of carbon particles with the morphological and chemical properties typical of particles from combustion: Aggregates of spherules made of graphene layers, consisting almost purely of carbon, with minor amounts of bound heteroelements, especially hydrogen and oxygen. This does not include the organic substances (oils, etc.) frequently present in or on combustion particles. Recently, the scientists attention has been shifting from the role of black carbon as a pollutant to its importance as a driver of global warming (Andreae & Gelencsér, 2006). Some model calculations show that its climate impact may rival that of methane, and that nowadays, global warming due to black carbon may be as much as 0.3–0.4ºC, while others estimate even a lower climate effect from this substance (Andreae & Gelencsér, 2006). As a consequence of this, the benefits of reducing BC to mitigate global warming have been cause cause of substancial controversy in the scientific community (Andreae & Gelencsér, 2006). Since the climate effects of BC aerosol strongly rely on its physical and chemical properties, as well as on its residence time and distribution in the atmosphere, there's a need of a clear understanding of these properties and accurate techniques for the determination of BC in the atmosphere (Andreae & Gelencsér, 2006).

1.2.2. Indicators of INP's

An ice nucleating particle (INP) is a particle which acts as the nucleus for the formation of an ice crystal in the atmosphere.

The formation, dynamics, precipitation rates and lifetimes of mixed-phase clouds, which comprise both supercooled liquid droplets and ice crystals, are difficult to understand because of some uncertainties in representation of aerosol-cloud interactions. Heterogeneous ice nucleation is an essential process in mixed phased clouds, where supercooled droplets are nucleated into ice crystals by INPs. Even though INPs in the atmosphere are rare (1 INP for every 1 x 10^5 aerosol particles (Rogers et al., 1998)), they still have the capacity to rapidly transform a liquid-dominated cloud into an ice-dominated cloud via the Wegener-Bergeron–Findeisen process (Korolev, 2007), hence changing the cloud's precipitation rates, lifetime and radiative properties. The representation of ice in MPCs is dependent on several parameters such as precipitation microphysics, ice growth and cloud droplet size distributions (McCluskey et al., 2017), as well as the abundance of INPs. To better understand the special and temporal distributions of INPs, there

is a lot of research to be done to comprehend their abundance and characteristics associated with the various types of aerosols.

The formation of ice particles in tropospheric clouds plays a dominant role in the alteration of the microphysical and chemical properties of the clouds, influencing their radiative transfer, precipitation and cloud electrification characteristics. Ice nucleation in the troposphere can occur either homogeneously or heterogeneously (Cantrell & Heymsfield, 2005). Homogenous freezing occurs with temperatures lower than -38 ºC and with a relative humidity above 140%. If these conditions aren't met, then the heterogenous nucleation takes place, with the aid of the aerosol particles (INPs), while also scavenging semi volatile gas-phase and acidic trace gas species. This phenomenon is important because the amount of ice condensate is concurrent to the water vapor budget in the upper region of the troposphere, where gas-phase water is particularly relevant due to its highly active greenhouse gas effect (Huang et al., 2021).

Clouds in the Arctic are in a mixed-phase state (water droplets and ice particles at the same time) for about 50% of the time (Hartmann et al., 2019) and despite their unstable nature, due to the Bergeron-Findeisen-effect (describes the growth of ice crystals at the cost of liquid droplets), they are extremely durable and extend over large areas (Morrison et al., 2012). Hence, their importance in the Arctic radiative budget. The primary ice-formation in MPCs takes place through heterogeneous IN or, in other words, ice nucleating particles (INP - particle which acts as the nucleus for the formation of an ice crystal in the atmosphere) are necessary to stimulate freezing of supercooled cloud droplets, affecting this way the precipitation, lifetime and radiative properties of the clouds. According to the literature, two main sources for atmospheric INP are thought to exist: Dust (primarily material from deserts and soils) and Bioaerosols (biogenic macromolecules originating from bacteria, fungi, lichen, marine biota, and pollen). Mineral dust particles are mostly active at temperatures below -20ºC and they are abundant in number; biogenic INPs have a tendency to nucleate at higher temperatures higher than -5ºC. Even if we don't know a lot about the abundance, nature, properties, and sources of INP in the Arctic and how they might change with the alterations in the climate, studying the variations of INP concentrations and properties over the past centuries might hence help to better understand potential future changes. With this in mind, some expeditions were made and for the first time, ice core material was used to derive INP concentrations on historical time scales. This work gave the scientific community the possibility of discussing 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 (Hartmann et al., 2019).

1.3. Where were fluorescent particles observed so far?

Fluorescent particles have been observed all over the world since both pollution and biogenic airborne particles are found everywhere. From Colorado, USA (Robinson et al., 2013), to the South Western Germany (Toprak & Schnaiter, 2013), passing through the poles (Moallemi et al., 2021), several studies on fluorescent particles have been made in these past years. Although they can be found worldwide, the focus of this study is the Arctic. Arctic has an increasing number of studies and campaigns, being a place where the interactions and their influence on the climate can be studied.

1.4. The contribution of this study

This study aims at the clarification of the seasonality of fluorescent particles, discussing topics like the particle concentrations and sizes throughout the year, helping this way the understanding of when they are more concentrated and therefore can have a major effect on climate and when they are not. Moreover, the sizes discussion can help figuring out what type of fluorescent particles can we be finding on each season.

Other topics like particle fractions and potential sources will be discussed and hence, opening a discussion about the influence of pollution or the inverse, in the concentration of this fluorescent particles and where do they possibly come from.

There have been other studies on Arctic aerosols and their role in the climate (Schmale et al., 2021), and there have been other studies on fluorescent particles on the Arctic and other sites, mainly focused on the possible sources of this fluorescent aerosols (Fu et al., 2015; Moallemi et al., 2021; Šantl-Temkiv et al., 2019). But the gap in knowledge that this study aims to fill is how the fluorescent particles concentration changes throughout the year in the Arctic, in order to have a better understanding on the differences in climate over the year.

2. Methods

2.1. Campaign Description

The Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) is the most extensive expedition in the central Arctic Ocean. Embarked on the German research vessel RV Polarstern, the largest polar expedition in history took place from September 2019 to October 2020. The icebreaker set sail from Tromsø, Norway, to spend a year drifting through the Arctic Ocean - trapped in ice (MOSAiC Consortium, 2016) ending the expedition in October 2020 in Bremerhaven, Germany. The drift track is shown in figure 1.

The scientific work in MOSAiC was executed on the ship itself and on the ice around the ship. A full annual cycle of observations was accomplished.

The comprehensive program to document and characterize all aspects of the Arctic atmospheric system in unprecedented detail, from multiple perspectives, and across multiple scales, was implemented and designed by an international team.

These measurements were coordinated with other observational teams to explore cross-cutting and coupled interactions with the Arctic Ocean, sea ice, and ecosystem through a variety of physical and biogeochemical processes.

The research program was organized into four sub-groups: atmospheric state, clouds and precipitation, gases and aerosols, and energy budgets (Shupe et al., n.d.).

During the expedition a laboratory container was running on the ship, equipped with several instruments, to measure aerosol concentration, size distribution and their chemical composition. A research camp was set up on the ice around the ship.

The data that was collected during the 13 months experiment is used for this Master's Thesis.

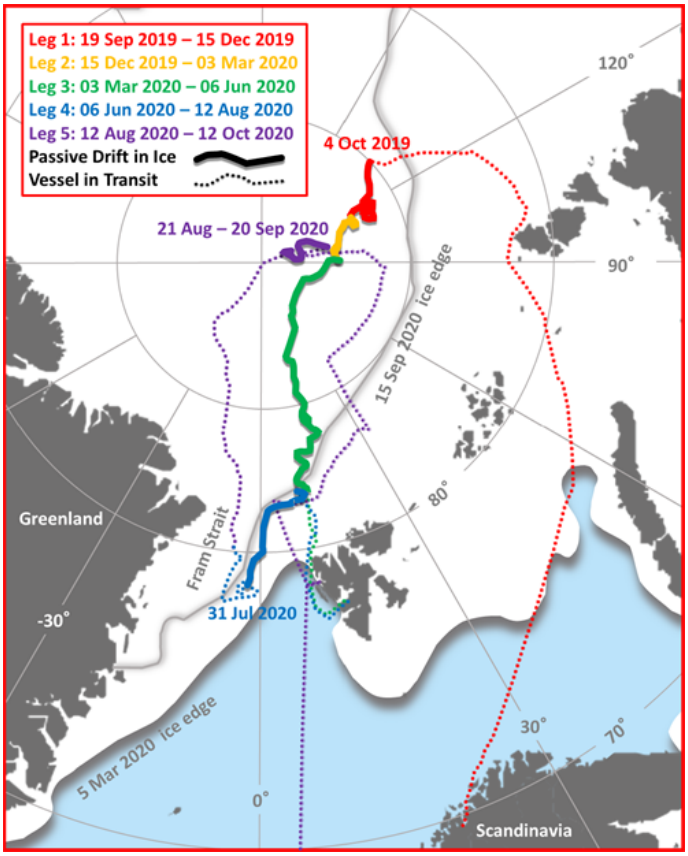

Figure 1. MOSAiC expedition route (Adapted from *Shupe et al., n.d.)*.

2.2. Fluorescent Aerosol Measurements

We used a Wideband Integrated Bioaerosol Sensor-New Electronics Option (WIBS-NEO - originally developed by the University of Hertfordshire and is licensed to and manufactured by Droplet Measurement Technologies) to measure fluorescent aerosol particles on a single-particle basis. Furthermore, the WIBS measures aerosol optical diameter in a range of sizes between 0.5 and 50 µm, and asymmetry factor (AF) which is a measure of aerosol morphology. In order to excite fluorescence in individual particles, the instrument uses UV xenon flashlamp sources. Contrasting with UV lasers, the UV xenon flashlamp sources allow for the precise selection of particular UV wavebands. These wavebands were selected to optimize the detection of the common bioaerosol components (Tryptophan and nicotinamide adenine dinucleotide - NADH) on the WIBS-NEO. This instrument is also a cost-effective alternative to other bioaerosol measurement instruments considering that Xenon source is far less expensive than a UV laser (Droplet Measurement Technologies, n.d.).

The single-particle fluorescence sensor uses a central optical chamber, around which are arranged the following components: a continuous-wave 635nm diode laser employed in the

detection of particles and the determination of particle size and shape, a forward-scattering quadrant photomultiplier tube (PMT) used in the determination of particle size and shape, two pulsed xenon UV sources emitting at different wavebands, two fluorescence detection channels, FL1 and FL2, detecting intrinsic particle fluorescence across two wavebands (Droplet Measurement Technologies, n.d.).

2.2.1. The Container

The *Swiss Container* was placed on the D-deck of the ship to monitor the aerosol and gas phase atmospheric composition. Aerosols and trace gases were sampled from two different inlets: a whole air inlet (total inlet) which allowed sampling all particles and droplets up to 40 µm and an interstitial inlet which is equipped with a cyclone to cut off particles larger than 1 µm, designed to sample particles that do not activate in cloud and fog. The total inlet was built after the Global Atmosphere Watch recommendations (World Meteorological Organization, 2016). An automated valve inside the container switched hourly between the total and interstitial inlets to allow instruments connected behind the valve to sample from each of the inlets alternately. The flow of the inlets was kept constant at 10 (total inlet) and 16.7 L/min (interstitial inlet). The inlets of the container had a length of 1.5 m and sampled at a height of approximately 15 m above sea level. The temperature inside the *Swiss Container* was kept constant at 20 °C. The sampled air was dried when entering the container due to the strong temperature gradient between outside and inside, but additional inline heating was applied when necessary. Relative humidity (RH) in the inlet lines was continuously measured and maintained below 40%.

2.2.2. Fluorescent Particle Detection

The aerosol fluorescent measurements are conducted by exciting particles with two Xenon flash lamps at wavelengths of 280 and 370 nm and then detecting the fluorescent light intensity in the wavebands from 310-400 nm and 420–650 nm. This results in three different excitation wavelength (ExWL) and emission waveband (EmWB) configurations: channel A (ExWL 280 nm and EmWB 310–400 nm), channel B (ExWL 280 nm and EmWB 420–650 nm) and channel C (ExWL 370 nm and EmWB 420–650 nm). The fluorescent detection threshold in each channel is determined based on the background signal measured during a “forced triggering” (FT) procedure. Moreover, we use the classification scheme introduced by Perring et al. (2015). In this method, the fluorescent particles are divided into 7 different classes (A, B, C, AB, AC, BC, and ABC) based on the logical combination of emitted signals in the three fluorescent channels (Moallemi et al., 2021).

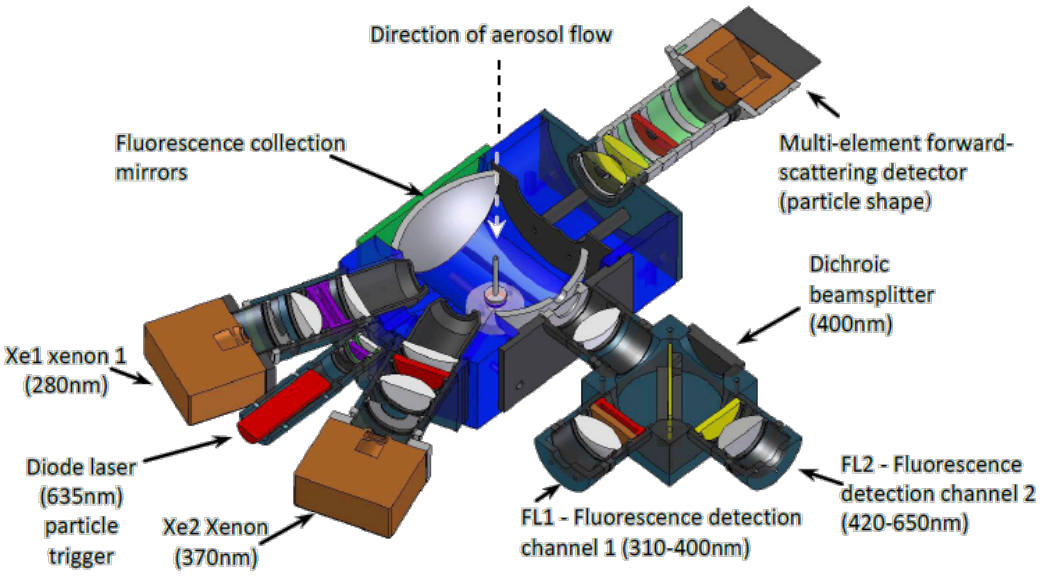

Figure 2. Wideband Integrated Bioaerosol Sensor (Adapted from *Droplet Measurement Technologies, n.d.)*.

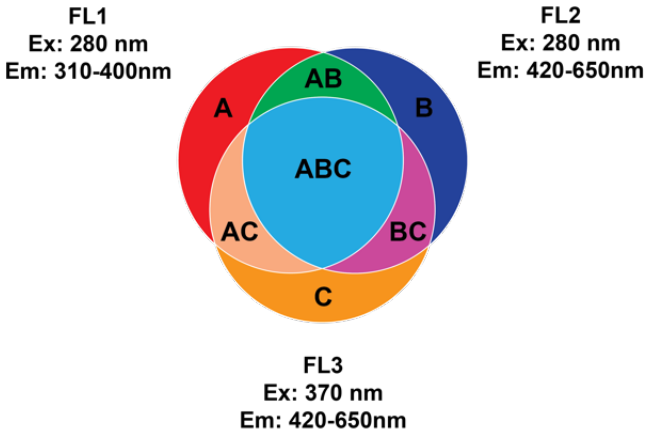

Figure 3. Fluorescent particles classes (Adapted from *Droplet Measurement Technologies, n.d.)*.

2.2.3. Particle Size

WIBS-NEO is a optical particle counter (OPCs) that, like most OPCs, uses a particle size calibration based on a theoretical curve that assumes the particles are spherical and of a specified refractive index (Mie theory). The calibration curve is based on aerosols of standard monodisperse polystyrene latex (PSL) microspheres. The refractive index of these spheres is quoted as 1.58 +/-0.2. Considering that this calibration curve is based on PSL spheres, the reported size should be taken only as an estimate when measuring spherical particles of different refractive index (e.g., water droplets) or non-spherical solid particles (Droplet Measurement Technologies, n.d.).

2.2.4. Particle Shape

WIBS-NEO incorporates an analysis of the forward scattered light captured by the Quadrant PMT to determine an index of particle shape, or more accurately, scattering asymmetry.

The instrument records the scatter intensity values received by each quadrant and determines the root-mean-square variation around the mean value to yield an Asphericity Factor (AF) (Droplet Measurement Technologies, n.d.). A perfect sphere would correspond to AF = 0, and a high aspect ratio fiber to an AF approaching 100 (Moallemi et al., 2021). Since it exists electronic and optical noise, results in spherical particles are measured with values between approximately ~2 to 6, rather than zero (Droplet Measurement Technologies, n.d.).

2.2.5. WIBS-NEO Toolkit

The WIBS-NEO Toolkit is data analysis software for the WIBS-NEO Instrument, developed using IGOR. This tool is used to load, process, visualize, and inspect data generated by the WIBS-NEO.

Functionality includes loading single and multiple raw data files containing particle-by-particle fluorescence, size, asymmetry factor and other particle-relevant data. The software then converts particle-by-particle data into time-resolved particle concentrations, and size distributions. The basic toolkit also has some plotting capability (Droplet Measurement Technologies, n.d.).

In this study, the toolkit was used to load raw data files that were organized in monthly periods and extract .csv monthly files with different particle data.

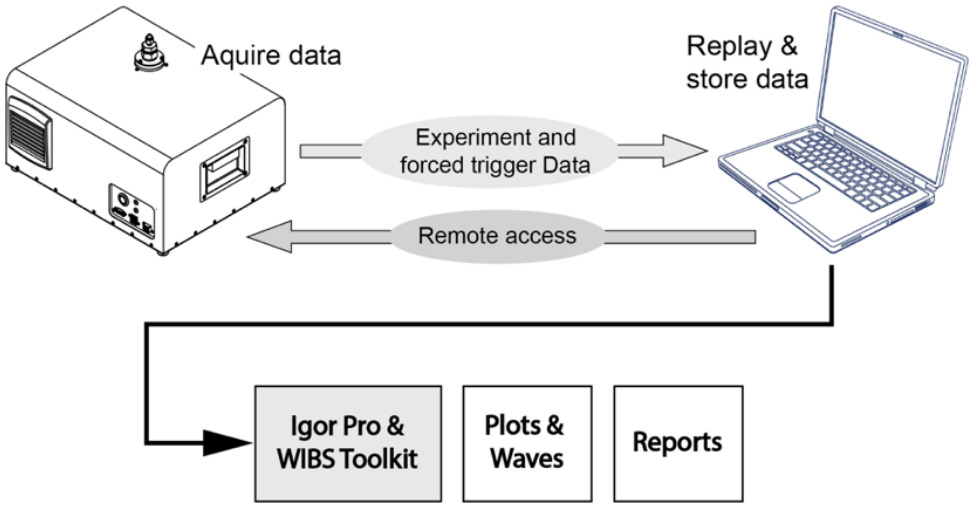

Figure 4. Flow diagram for WIBS-NEO data acquisition and evaluation (Adapted from *Droplet Measurement Technologies, n.d.)*.

2.3. Data Analysis

Concerning the data analyzis, I first used IGOR PRO 8, version 8.0.4.2, 64 bit, by WaveMetrics, along with the WIBS-NEO toolkit, to process the WIBS data.

Firstly, I joined all the raw data files and forced trigger files in different folders, separated by month. The toolkit provides options for loading all files in a single directory or for loading a single file within a directory. These folders were loaded in IGOR on the *Load data and Background* window, using the option *Load all files in folder* for both the raw data and the forced trigger background. Subsequently, the data was saved as a text file by selecting the button on the main Toolkit screen. The monthly text files (.csv) provided data for average asphericity, average size (μm) and average concentration (cm^3), for all types of particles, in one second resolution. These text files were then concatenated into a single dataset using Python.

Afterward, a pollution mask developed by Beck et al. (in preparation) was applied to the dataset. This pollution mask uses Condensation Particle Counter (CPC) data. The periods that have really high concentrations or concentration above the median or the gradient, are deleted. When applied to the WIBS dataset, 0 corresponds to polluted periods and 1 to clean periods. The dataset was then separated into polluted data and clean data.

Figure 5 is a representation of a timeseries plot: the above panel has the pollution mask applied and the bottom one has untreated data.

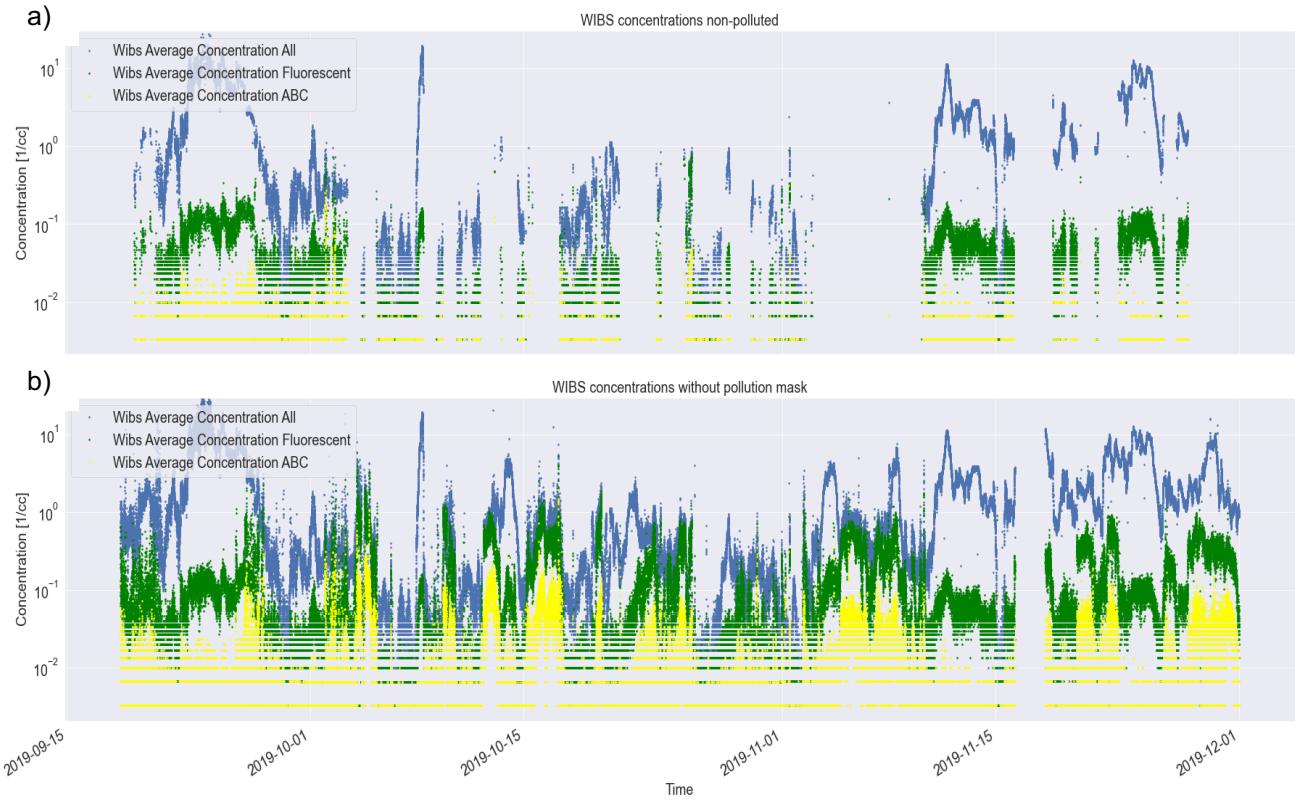

Figure 5. Application of the pollution mask. a) Clean period. b) No pollution mask applied.

The weather is also an important factor which influences aerosol concentrations. Wind data was collected during the entire expedition on Polarstern. Wind speed and direction were measured with a 2D sonic anemometer on the main mast of the vessel. The wind dataset was used in a time resolution of 1 minute in this study.

3. Results and Discussion

3.1. Seasonality of fluorescent aerosol

In this chapter, total and fluorescent particle concentrations, particle size, particle fractions and potential sources of these particles will be analyzed and discussed. The main focus of the discussion will be the seasonality of fluorescent particles, shown through monthly boxplots and stacked barplots, created using python. Subsequently and in order to better understand the connection between wind and the changes in particle concentration, a binned boxplot of concentration as function of wind speed was made in python.

3.1.1. Fluorescent particle concentrations throughout the year

In this first part of the chapter, seasonal particle concentrations will be approached.

Figure 6 shows the seasonal variation of both total and fluorescent particle concentration in a logarithmic axis. The difference between total concentration and fluorescent concentration is demonstrated in figure 6 and it can be observed that the first can reach sometimes two orders of magnitude higher than the second.

For total particle concentration, highest medians (medians give us information about the mid-point of the data, meaning that 50% of the data will be below that point and the other 50% will be above) occur from November to May and the greatest concentrations happen in January and February. In summer months and October, there are lower medians and concentrations, reaching its lowest values in August and October. As for the fluorescent particle concentrations, they follow the same pattern as the total particle concentrations, the peaking in January and February and having their lowest medians and concentrations in August and October.

When considering the Arctic Haze phenomenon, these results make sense since this phenomenon is amplified in winter and spring seasons, when the long-range transport of pollutant aerosols is higher. Even though in summer months there are more forest fires, this is not enough to surpass what happens in winter or spring seasons (Quinn et al., 2007).

To complement the previous hypothesis, studies on back trajectory data could be made, in order to understand where the particles could be coming from and also, looking into some specific case studies (like storms, for example) in deeper detail to have an idea of how they can influence the concentrations of fluorescent and non-fluorescent particles.

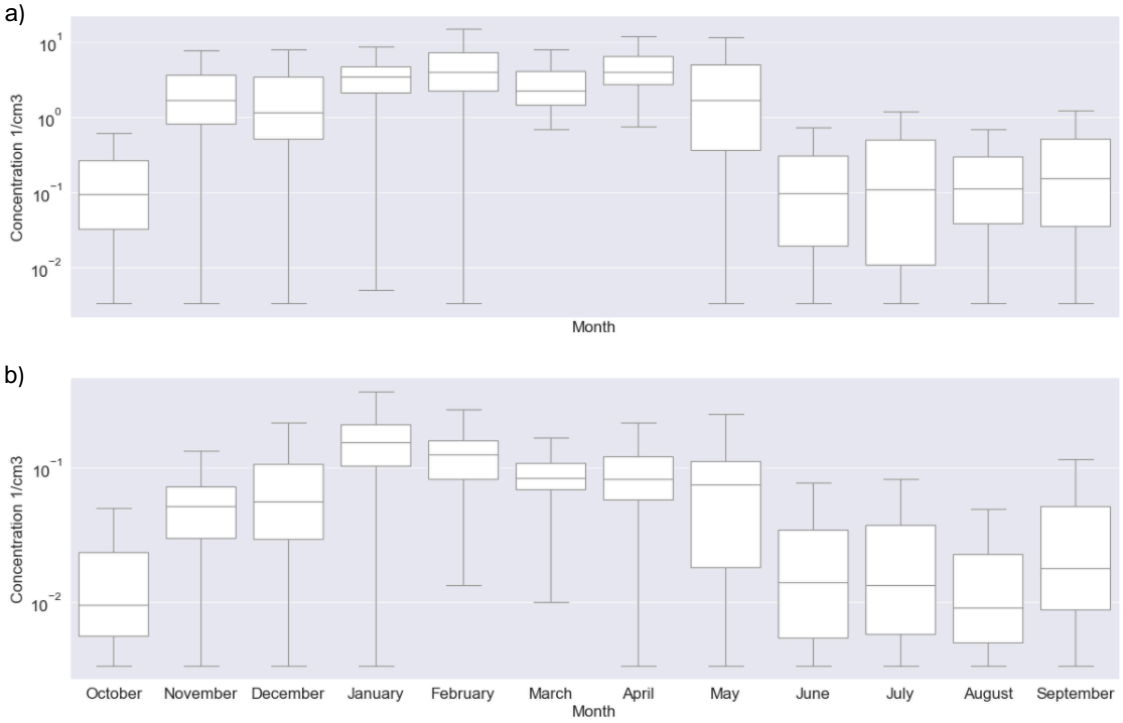

Figure 6. Seasonal variation in logarithmic axis. a) Total particle concentration. b) Fluorescent particles concentration as measured by the WIBS.

In order to look more carefully into the concentrations, the same boxplot was plotted with a linear axis instead (figure 7). It can be observed that the higher concentrations happen during winter and spring seasons, confirming that the peaks in concentration are on January and February.

The highest concentration of total particles is about 15 particles per cm^3 in February, reaching in the same month the highest median as well, with a value of 4 particles per cm^3. Contemplating the fluorescent particle concentrations, the maximum concentration is around 0.40 particles per cm^3 in January and the median is 0.15 particles per cm^3.

On the other hand, summer and autumn seasons have low concentrations, both in total and fluorescent particles. October and August reach concentrations of almost 0 particles per cm^3.

Once again, these results are according to the literature about the Arctic Haze phenomenon but when contemplating fluorescent concentrations, one would expect to have higher concentrations in summer, since, in principle, these should be the months when biological activity is more intense and therefore, bioaerosols would be in greater representation. One hypothesis is that, during winter/springtime, particles that can interfere with the fluorescence detection like dust or black carbon, are more dominant. To confirm this, one could look into pollution data (black carbon data, for example) and try to relate this concentrations with the previous data, in each month.

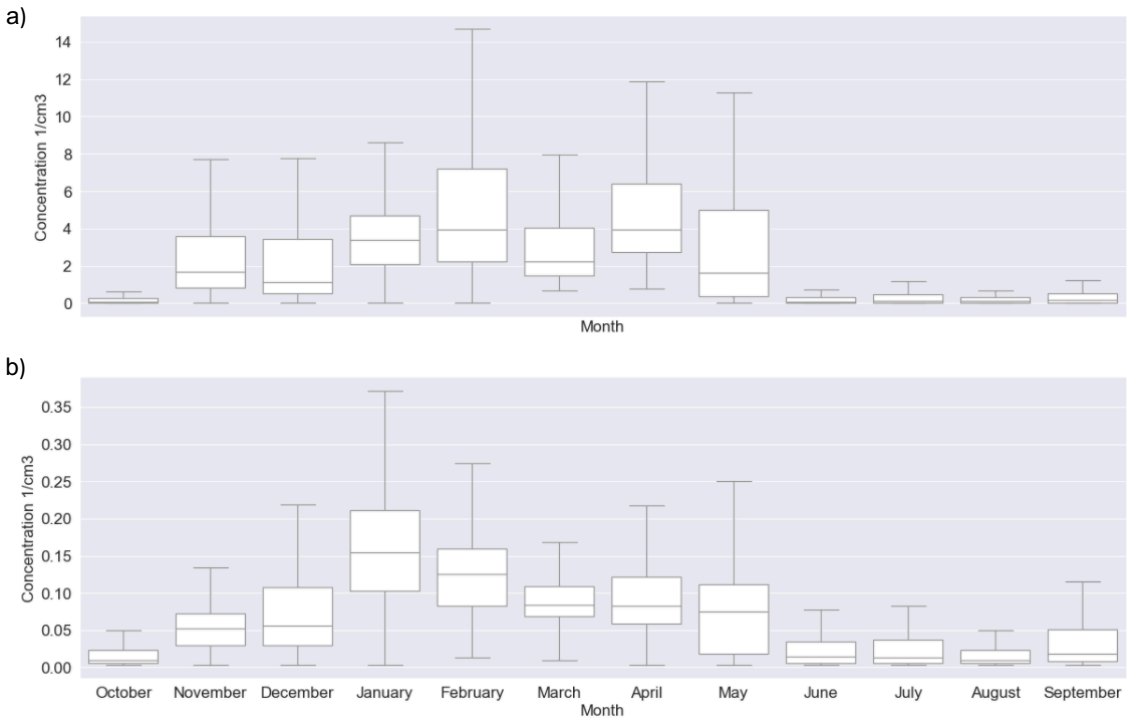

Figure 7. Seasonal variation in linear axis. a) Total particle concentration. b) Fluorescent particles concentration as measured by the WIBS.

3.1.2. Seasonality of particles size

These second chapter has the purpose of discussing the difference in sizes over the year. For that, the boxplots in figure 8 were plotted.

In the boxplots below, it is possible to see that the median sizes are similar throughout the year, even though, in winter and spring, the maximum sizes are smaller than in summer and autumn. The variation of sizes is minor in winter and in spring and in summer and autumn, it is bigger. The largest particles in figure 8 a) measure around 1.27 μm in June and the smallest around 0.5 μm (this is the lower limit of the instrument). As for the sizes of the fluorescent particles, they are generally the same size as the rest of the particles. The biggest particles measure about 1.9 μm in October and the smaller ones are again 0.5 μm. These results suggest that non-fluorescent particles have a smaller size distribution than the fluorescent ones.

Dust and combustion particles have a greater range of sizes than biogenic particles. However, since the variation of sizes in the absence of biological activity (winter months) and keeping in mind that the maximum sizes are smaller in the winter and spring, one of the possible hypotheses is that bioaerosols are generally bigger than other types of particles in the Arctic.

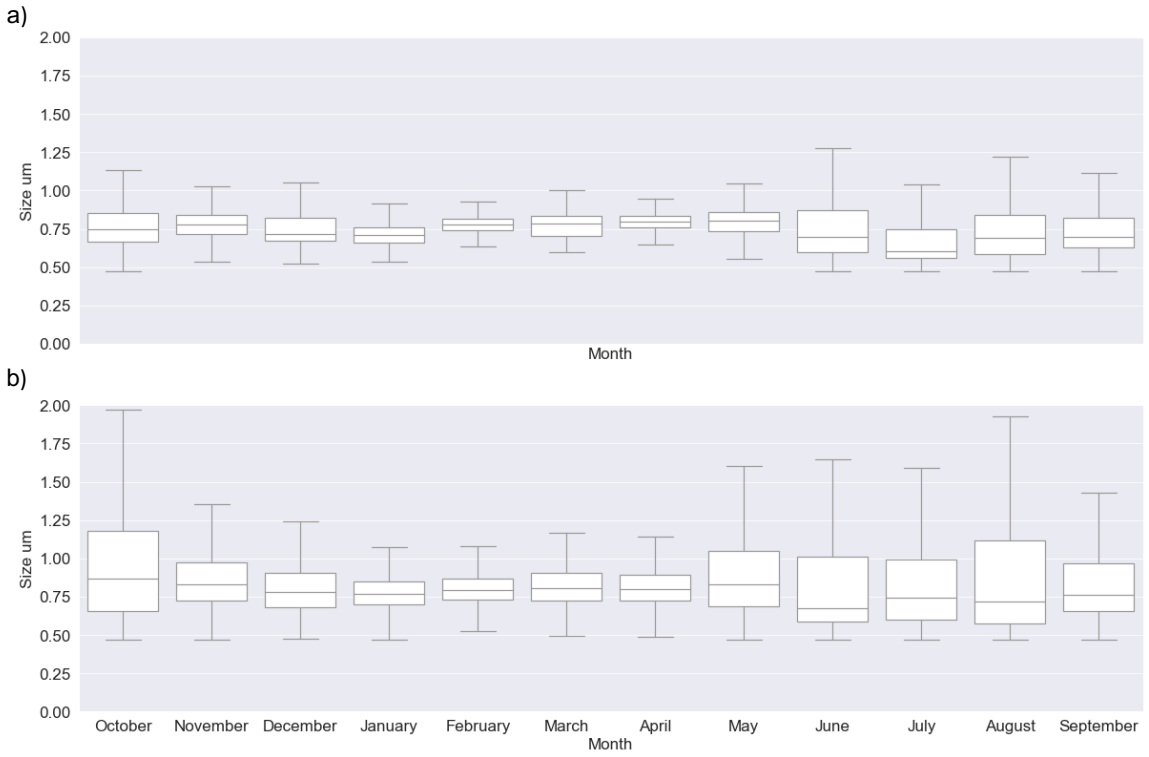

Figure 8. Seasonal variation in linear axis. a) Total particle size. b) Fluorescent particles size as measured by the WIBS.

Figure 9 aims the deep clarification of the boxplots above with the binning of the different sizes.

In figure 9 it can be seen that sizes between 0.7 and 0.9 μm are dominant, meaning they have higher concentrations, with medians of approximately 0.09 particles per cm^3 and maximums ranging between 0.27 and 0.29 particles per cm^3. These two size ranges are the sizes with more datapoints (table 1), meaning they are more representative.

The typical size range of bacteria is between 0.5-6 μm. In figure 9, the size range of the particles is exactly this size range, leading to the conclusion that these results reflect a majority of biogenic particles.

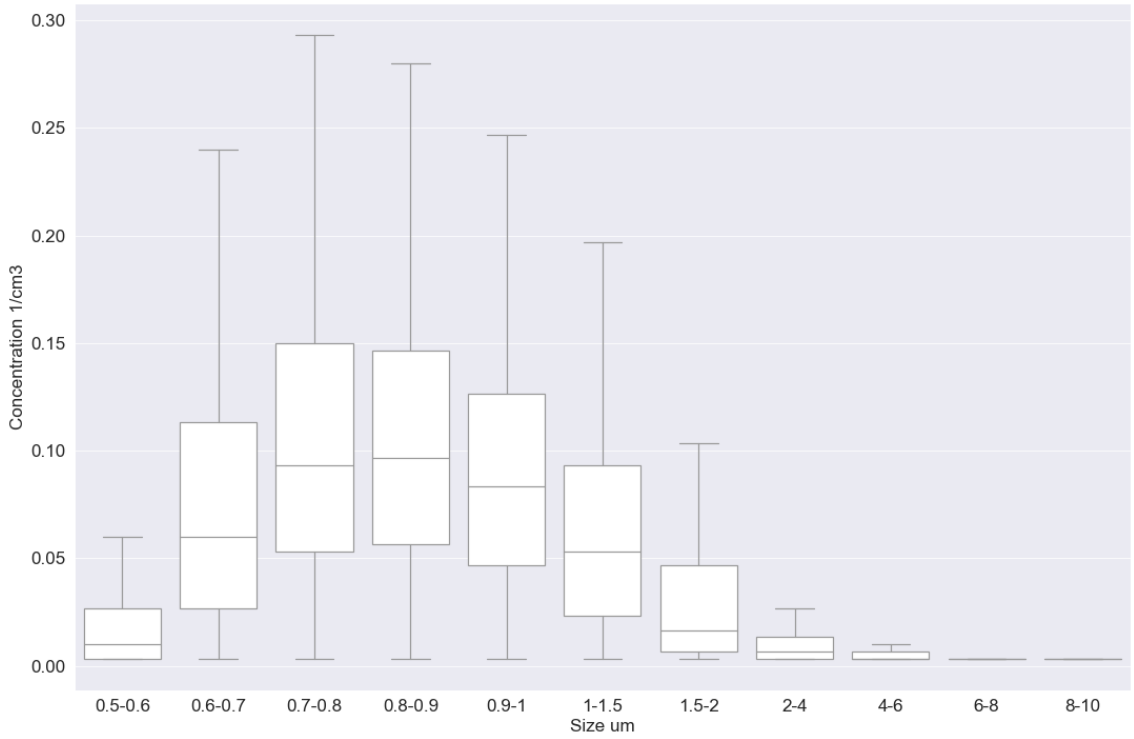

Figure 9. Variation of sizes of fluorescent particles.

Table 1. Number of datapoints on each size bin of fluorescent particles.

Size Range	Number of datapoints
0.5 – 0.6	13343
0.6 – 0.7	28885
0.7 – 0.8	41285
0.8 – 0.9	32164
0.9 – 1.0	17685
1.0 – 1.5	20585
1.5 – 2.0	3360
2.0 – 4.0	2210
4.0 – 6.0	234
6.0 – 8.0	66
8.0 – 10.0	26

In figure 10 it can be seen that sizes between 0.8 and 1 μm are dominant, meaning they have higher concentrations, with medians of approximately 4.5 and 2.5 particles per cm^3 and maximums ranging between 15 and 16.1 particles per cm^3. The three size ranges with more datapoints (table 2) and therefore, more representative are the sizes between 0.6 – 0.9 μm.

In this case, the results in question are for all particles data, suggesting this way that when mixing non-fluorescent particles with fluorescent particles, the concentrations are higher in each size range but there are fewer size ranges.

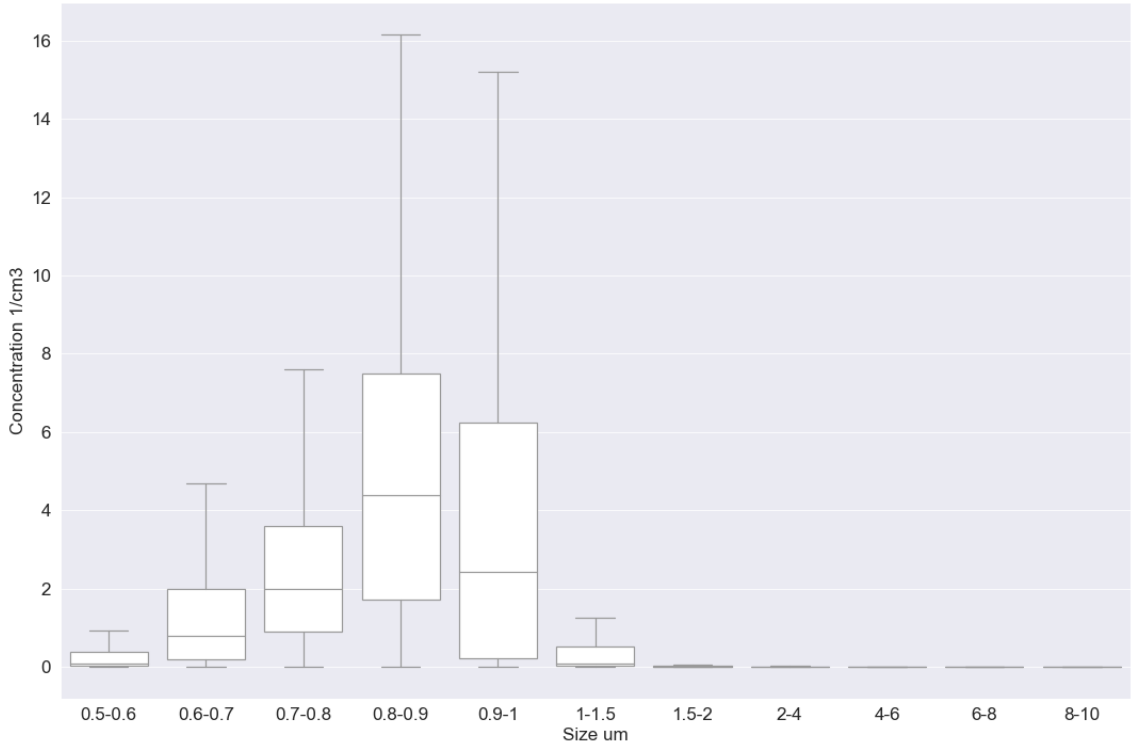

Figure 10. Variation of sizes of total particles.

Table 2. Number of datapoints on each size bin of total particles.

Size Range	Number of datapoints
0.5 – 0.6	15569
0.6 – 0.7	41577
0.7 – 0.8	62555
0.8 – 0.9	42983
0.9 – 1.0	9896
1.0 – 1.5	4961
1.5 – 2.0	539
2.0 – 4.0	426
4.0 – 6.0	65
6.0 – 8.0	23
8.0 – 10.0	11

3.1.3. Particle fractions

In order to understand if the different particle types have different patterns over the year, two graphs were plotted: one for clean periods only (figure 11) and one for polluted periods only (figure 12). In these graphs, the colors correspond to fluorescent particles of different types.

The first thing that figure 11 shows is that the AB and B type particles seem to be overall the dominant types in clean periods. Both types are more preeminent during the summer months. The AC particles are the ones with the smallest fraction.

The biggest AB fraction and the smallest B fraction happen in June and the biggest B fraction and smallest AB fraction happen in January.

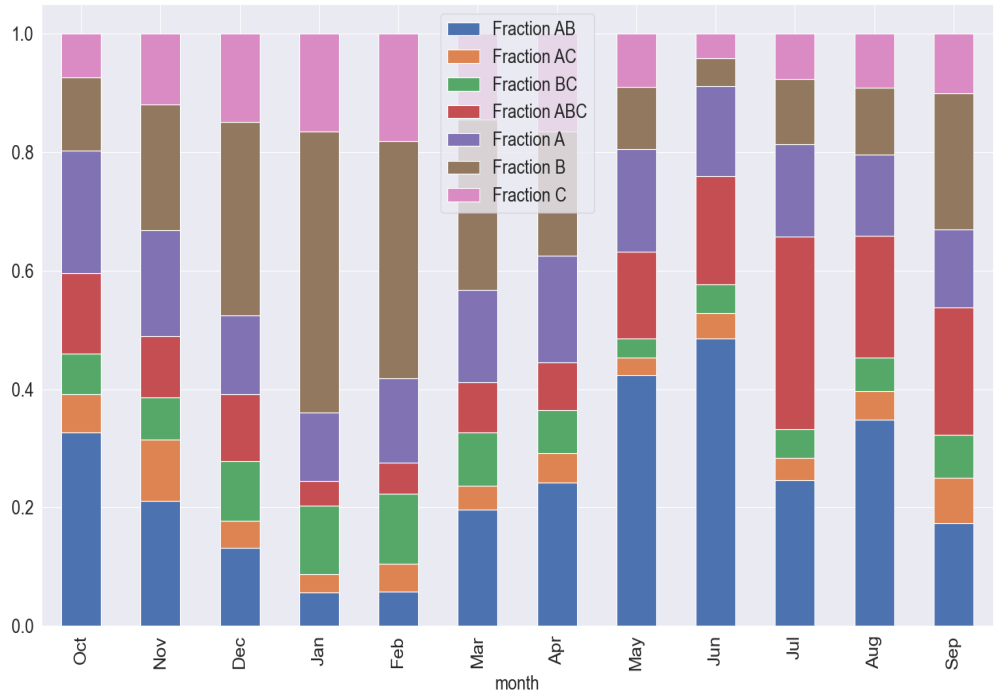

Figure 11. Seasonal variation of fractions of the different types of particles during clean periods.

When looking at figure 12, there is a clear conclusion: AB particles are the dominant particles in pollution periods and they are more present throughout the year. Then again, the AC

particles are the ones with the negligible fraction. Figure 12 establishes that pollution is clearly a source of fluorescent particles. December, January, February and March are the months with less AB type particles. April is month with more AB type particles.

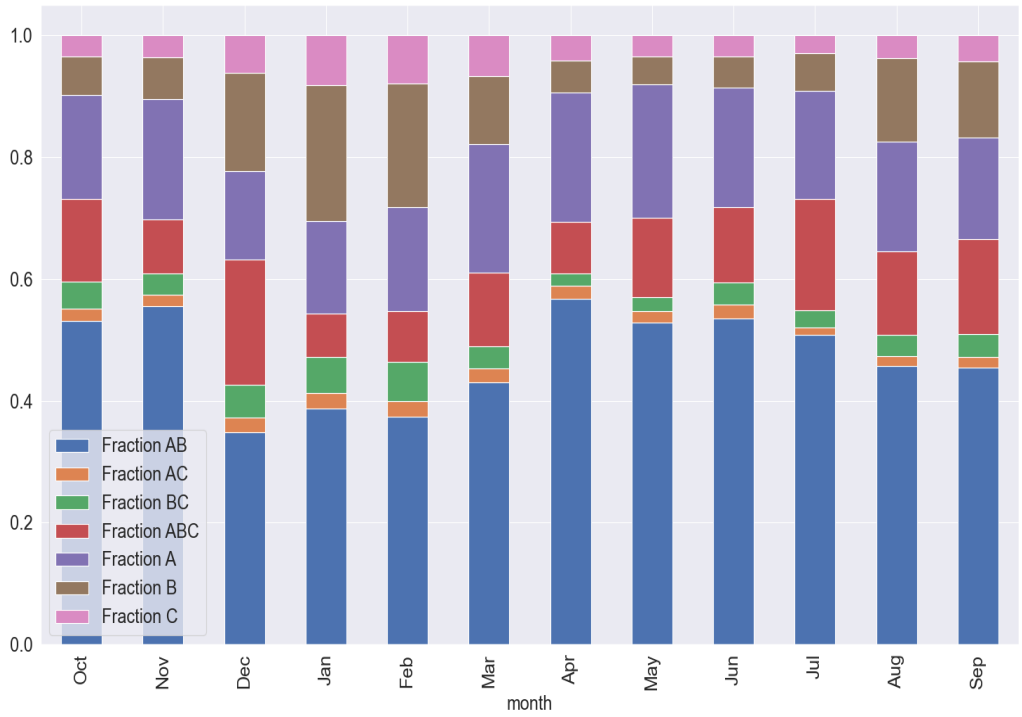

Figure 12. Seasonal variation of fractions of the different types of particles during polluted periods.

To support the hypothesis (Moallemi et al., 2021) that AB particles are dominant in pollution, the following equations were calculated:

$$\frac{Mean\ Concentration\ of\ AB\ particles\ clean}{Mean\ Concentration\ of\ All\ particles\ clean} = \frac{0.0193}{2.9772} = 0.0065$$

$$\frac{Mean\ Concentration\ of\ AB\ particles\ polluted}{Mean\ Concentration\ of\ All\ particles\ polluted} = \frac{0.1655}{2.6041} = 0.0636$$

As shown, the presence of AB particles in pollution in one order of magnitude higher then clean periods. This hypothesis would benefit from more studies in different regions of the planet, to confirm that in pollutes sites, there's always a dominance of AB type particles over the other types.

3.2. Potential Sources

It is proven now that pollution is one of the potential sources for fluorescent particles. But it is conceivable that other sources may exist. These sources can be snow (the wind as an uplifting mechanism of snow) or other local sources and possible regional sources.

The boxplots from figures 13 – 17 intend to show the influence of the wind in the concentration of fluorescent particles.

Figure 13 focus on the September to November season (SON). In this season, the concentrations change a lot with the increasing of the wind speed, meaning that, with higher wind speeds, there are higher concentrations and with lower wind speeds, lower concentrations. Implying that wind has a lot of influence in concentrations in this season.

These results can possibly mean that, in this season, local sources are a potential focus for next studies.

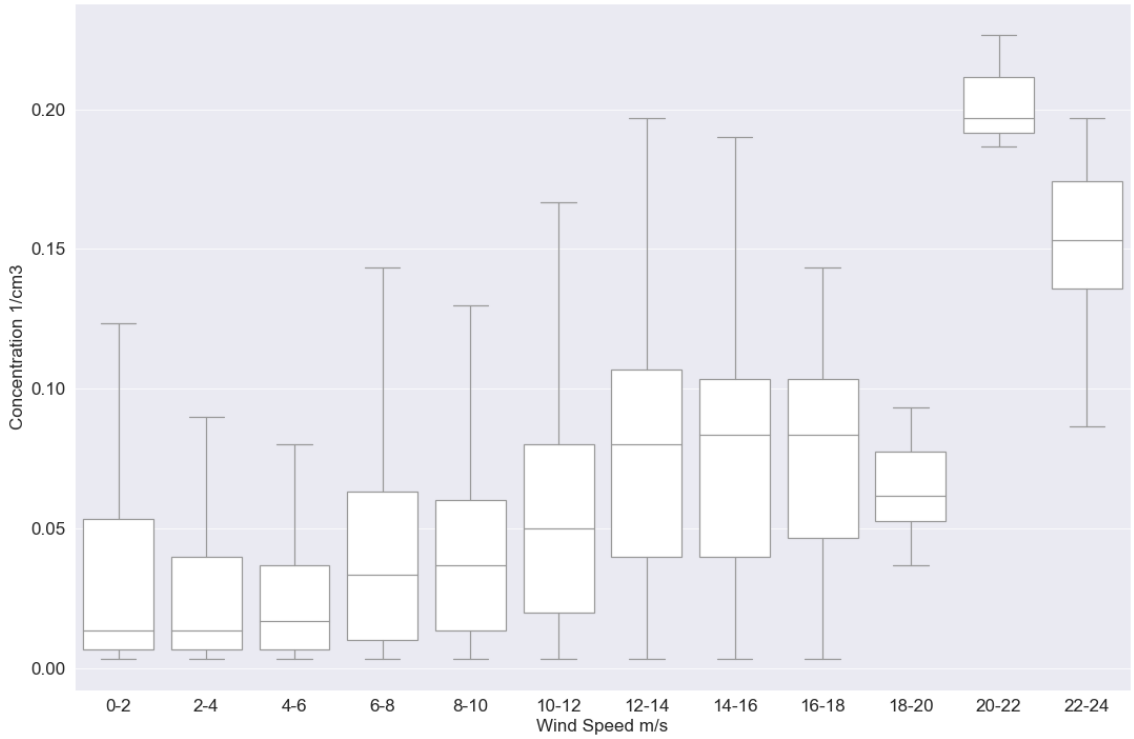

Figure 13. Concentration of fluorescent particles as a function of wind speed (September-November).

When looking at figure 14 (representing December to February season), wind does not reach such high speeds as the previous season. However, it still has some influence in the concentrations since the concentrations still change with the wind speed. In this season, speeds only go up to 20 m/s instead of 24 m/s and the median concentrations are higher than the previous season.

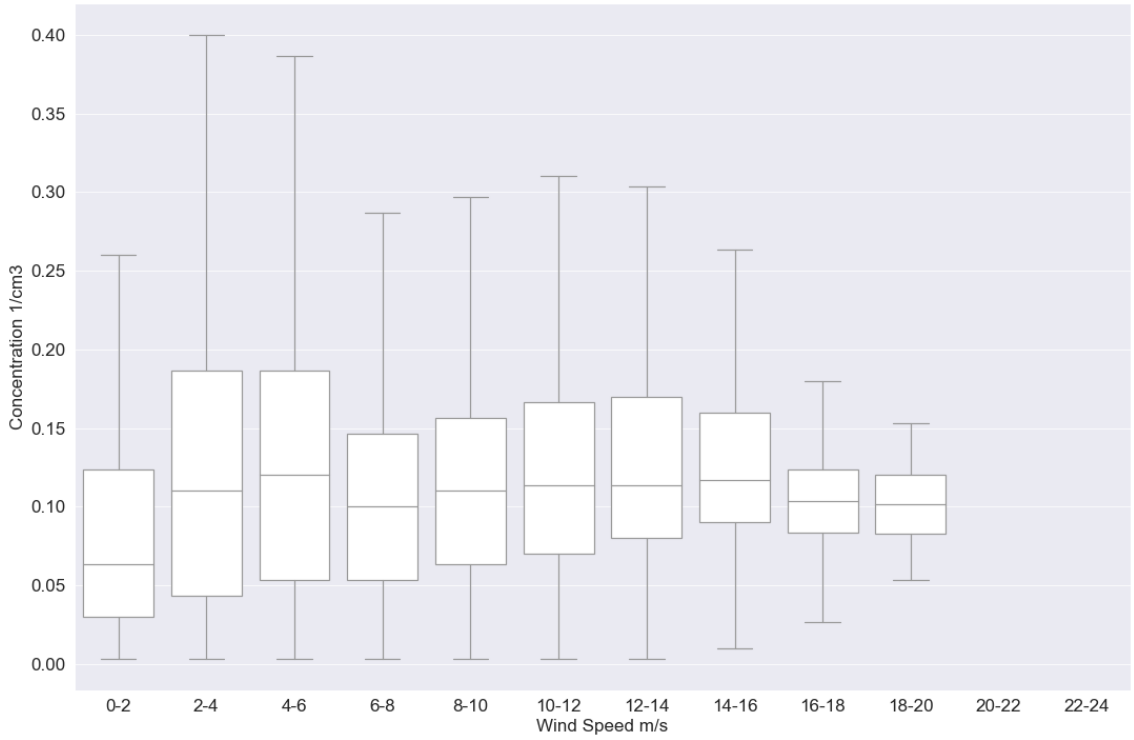

Figure 14. Concentration of fluorescent particles as a function of wind speed (December-February).

Figure 15 (March – May season) follows a different pattern as the previous one but reaching speeds up to 24 m/s this time. The concentrations are lower than the previous season. Wind has, again, some influence in the concentration of fluorescent particles.

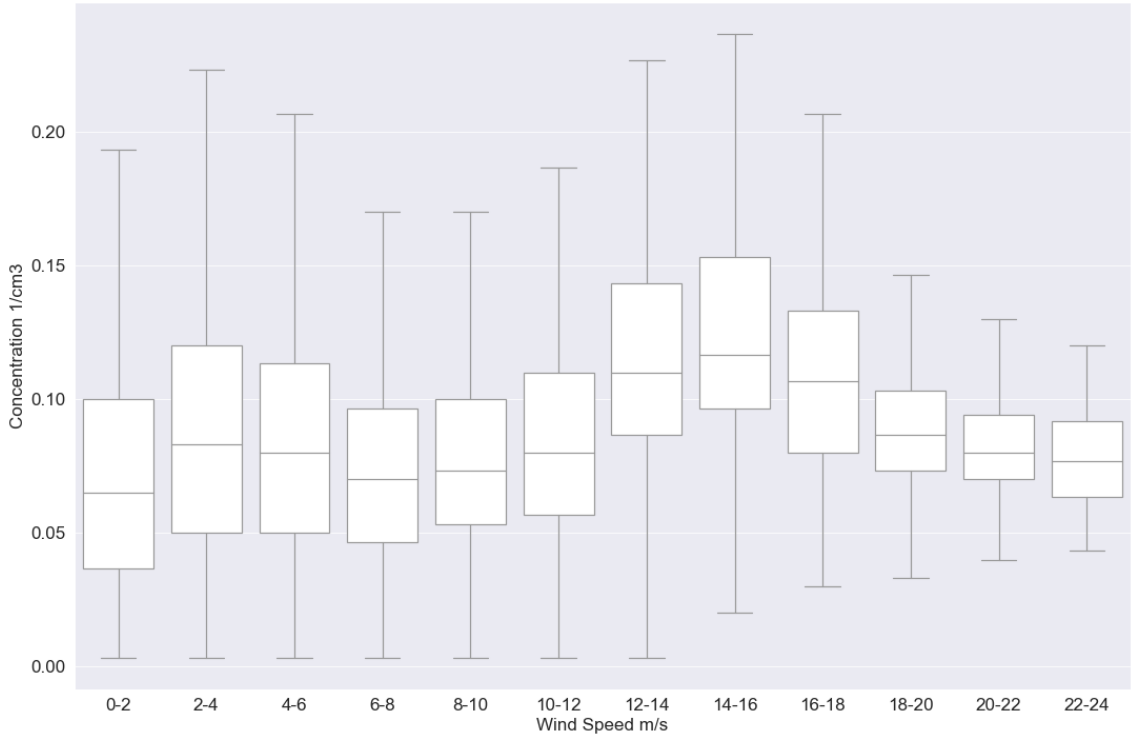

Figure 15. Concentration of fluorescent particles as a function of wind speed (March-May).

Figure 16 shows the summer season (June – August) which seems to be an abnormal season when compared to the others. The wind speeds only go up to 16 m/s and the concentrations in each bin are low, except for the 10 – 12 m/s speed range, where the concentrations are much higher than at other speeds. This could be an isolated event where speeds in this range lifted a lot of biological particles.

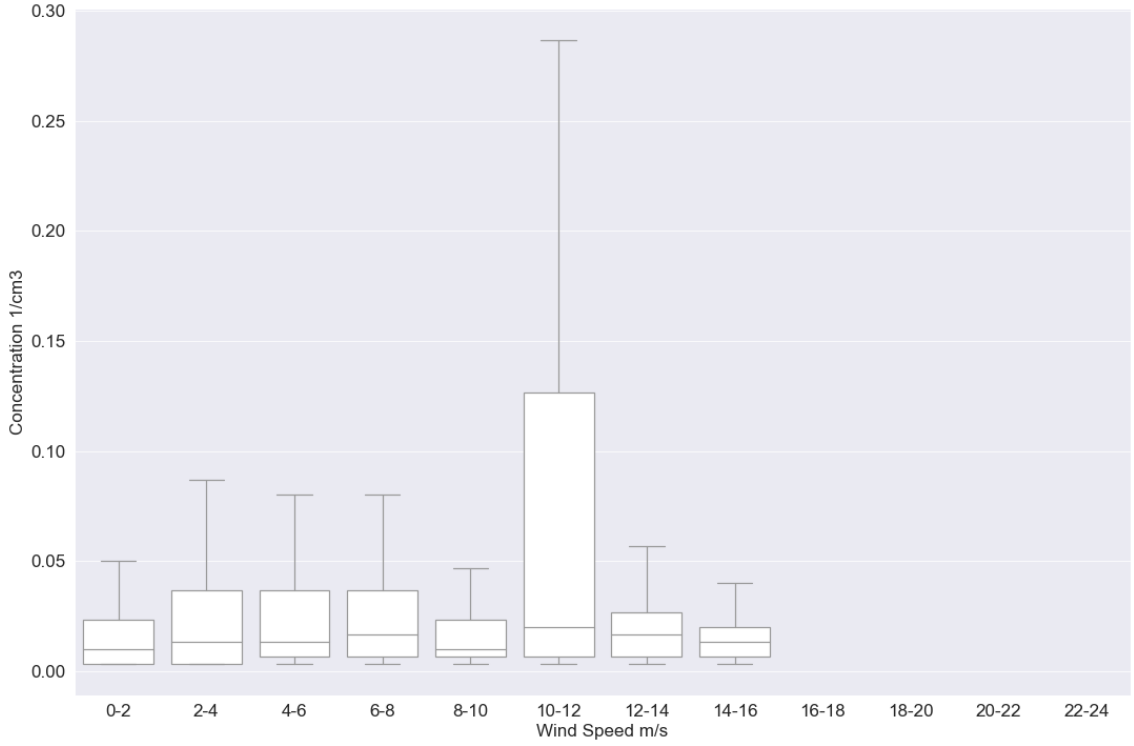

Figure 16. Concentration of fluorescent particles as a function of wind speed (June-August).

Figure 17 represents the full year cycle and it sums up the ideas of the figures above. The medians increase with the wind speed until the 16 m/s and then they start to decrease, meaning that wind has in fact some effect in the increasing of concentrations.

Most of the datapoints (table 3) are from the lower concentrations, meaning that lower speeds are much more represented than higher speeds.

All the previous discussed results are related to a possible local source, using wind as a mechanism but, to really have the full extent of the possible sources, back trajectory data would have to be used to evaluate regional sources, instead of just local, since there's also a high probability of particles coming to the Arctic from the north of Europe.

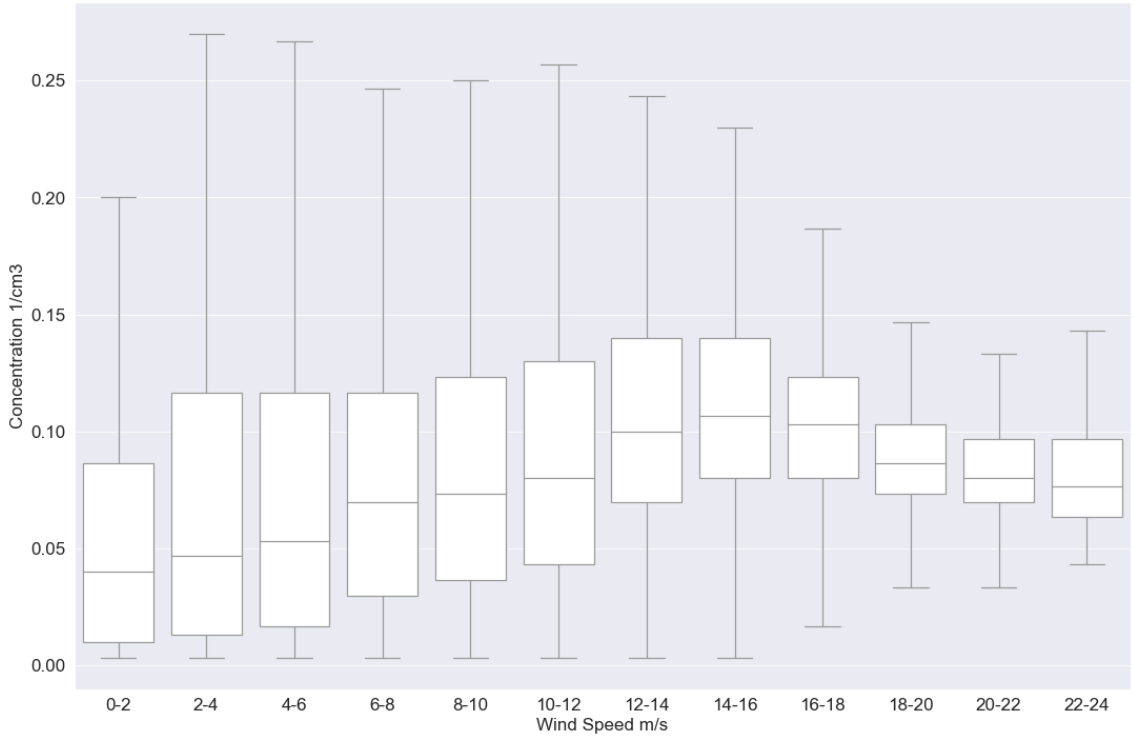

Figure 17. Concentration of fluorescent particles as a function of wind speed (full year cycle).

Table 3. Number of datapoints on each wind speed bin (full year).

Wind Speed Range	Number of datapoints
0 – 2	7921
2 – 4	29209
4 – 6	41008
6 – 8	46260
8 – 10	28839
10 – 12	18101
12 – 14	11048
14 – 16	5239
16 – 18	1507
18 – 20	417
20 – 22	279
22 – 24	85

4. Conclusions

In the lights of this thesis' topic and all that was previously discussed, some conclusions can be made regarding the seasonality of fluorescent aerosol.

Total and fluorescent particle concentrations were discussed and led to the inference that total particle concentrations are usually two orders of magnitude higher than the fluorescent particle concentrations. Moreover, both total and fluorescent particles have their highest median values of concentration in the December-January-February (DJF) and March-April-May (MAM) seasons, and the concentration peaks are reached in January and February. August and October are months with low values of concentrations. This can be explained by the Arctic Haze phenomenon, although the expectation was that the summer would have higher concentrations of fluorescent particles due to the increased biological activity.

Regarding particle sizes, the median sizes are similar throughout the year, but June-July-August (JJA) and September-October-November (SON) seasons have the bigger maximum sizes. Considering the size measures, the conclusion to be taken is that fluorescent particles are generally the same size as non-fluorescent particles. The most common sizes for fluorescent particles are 0.7 to 0.9 μm and the total range of sizes is between 0.5 and 6 μm. A high probability of these results reflecting a majority of biogenic particles was discussed.

Particle fractions were talked through and the main insights that we gained were that AB and B type particles are dominant through the entire year, in clean periods, and AB type particles are clearly dominant in polluted periods, supporting a previously published hypothesis.

Finally, wind was seen as a possible mechanism to lift snow and sea particles, making them possible local sources of bioaerosols.

Further studies need to be made in the bioaerosols field to better understand their influence on cloud formation processes and their sources.

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