Stockholm University

Understanding the long-term trends and seasonality of Arctic atmospheric aerosol

Through the lens of black carbon and new particle formation

Dominic Heslin-Rees

Doctoral Thesis in Environmental Sciences at Stockholm University, Sweden 2025

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Academic dissertation for the Degree of Doctor of Philosophy in Environmental Sciences at Stockholm University to be publicly defended on Friday 11 April 2025 at 10.00 in De Geersalen, Geovetenskapens hus, Svante Arrhenius väg 14 and online via Zoom, public link is available at the department website.

Abstract

The Arctic region is witnessing changes on an unprecedented level. Surface air temperatures have increased at a rate four times the global average. Two of the main climate forcers that are responsible for perturbing the radiative balance in the Arctic are greenhouse gases and atmospheric aerosols. Aerosols are tiny solid or liquid particles suspended in the atmosphere that range in size from a few nanometres to tens of microns. These particles affect the climate by interacting with radiation and influencing cloud formation, brightness, and longevity. The work presented in this thesis aims to improve our understanding of the drivers and mechanisms involved in controlling both the seasonal variations and the long-term changes in Arctic aerosols, and analyse the general aerosol lifecycle. In a changing Arctic, both the emissions of anthropogenic and natural aerosol particles have and are expected to continue to change. For one, the long-range transport of anthropogenic aerosols is likely to continue to decline with reductions in emissions.

Measurements of Arctic aerosols were carried out at a research observatory on Svalbard. In this thesis, a variety of instrumentation and measurements were used to assess seasonal and long-term changes in various aerosol-related variables.

The work in this thesis shows that the concentration of light-absorbing aerosol particles has decreased significantly over the past two decades, with the largest decrease in contributions from northern Siberia. This thesis argues that a quarter of the overall reduction is due to changes to the removal processes via wet scavenging. In this thesis, the changes in environmental parameters along the transport pathway to the site are explored. From this perspective, precipitation is shown to act as both a source and a sink, impacting the number of particles depending on their size, whilst solar radiation is shown to promote an increase in the number of aerosol particles over the entire size spectrum. Furthermore, using the first long-term time series measuring light-absorbing particles inside and outside of clouds, the process of nucleation scavenging is explored. Increased uptake of light-absorbing particles into cloud droplets is presented from April until October. Incorporation of these particles into cloud droplets is shown to be dependent on temperature and cloud water content. Lastly, the frequency in the production of small particles, barely a nanometre in diameter, in the vicinity of Svalbard is shown to be heavily influenced by solar radiation and the total surface area of pre-existing aerosol particles. The Greenland Sea is shown to be a relatively larger source of these small particles compared to neighbouring seas. Its shown that the total surface area of pre-existing aerosols within airmasses is reduced through cloud and precipitation events, setting the stage for new particle formation and the replenishment of aerosol particles in the presence of solar radiation.

Understanding how these findings can be broadened and applied across a larger geographical region remains to be answered. Additionally, the overall effect these mechanisms and changes can have on the radiative balance in the Arctic requires further exploration.

Keywords: *Arctic, aerosol, scavenging, sinks, precipitation, new particle formation, black carbon, trends, seasonality.*

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Cover picture of Ny-Ålesund research village taken by the author on April 8 2022 close to 11pm.

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The Arctic region is witnessing changes on an unprecedented level. Surface air temperatures have increased at a rate four times the global average. Two of the main climate forcers that are responsible for perturbing the radiative balance in the Arctic are greenhouse gases and atmospheric aerosols. Aerosols are tiny solid or liquid particles suspended in the atmosphere that range in size from a few nanometres to tens of microns. These particles affect the climate by interacting with radiation and influencing cloud formation, brightness, and longevity. The work presented in this thesis aims to improve our understanding of the drivers and mechanisms involved in controlling both the seasonal variations and the long-term changes in Arctic aerosols, and analyse the general aerosol lifecycle. In a changing Arctic, both the emissions of anthropogenic and natural aerosol particles have and are expected to continue to change. For one, the long-range transport of anthropogenic aerosols is likely to continue to decline with reductions in emissions.

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spectrum. Furthermore, using the first long-term time series measuring light-absorbing particles inside and outside of clouds, the process of nucleation scavenging is explored. Increased uptake of light-absorbing particles into cloud droplets is presented from April until October. Incorporation of these particles into cloud droplets is shown to be dependent on temperature and cloud water content. Lastly, the frequency in the production of small particles, barely a nanometre in diameter, in the vicinity of Svalbard is shown to be heavily influenced by solar radiation and the total surface area of pre-existing aerosol particles. The Greenland Sea is shown to be a relatively larger source of these small particles compared to neighbouring seas. Its shown that the total surface area of pre-existing aerosols within airmasses is reduced through cloud and precipitation events, setting the stage for new particle formation and the replenishment of aerosol particles in the presence of solar radiation.

Understanding how these findings can be broadened and applied across a larger geographical region remains to be answered. Additionally, the overall effect these mechanisms and changes can have on the radiative balance in the Arctic requires further exploration.

Sammanfattning

Klimatet i Arktis genomgår förändringar utan tidigare motstycke. Temperaturerna i ytskiktet har ökat mer än fyra gånger så snabbt som det globala medelvärdet. Två av de huvudsakliga orsakerna till den förändrade strålningsbalansen i Arktis kan härledas till växthusgaser och atmosfäriska aerosoler. Dessa aerosoler är mycket små fasta partiklar eller vätskedroppar suspenderade i atmosfären och återfinns inom storleksintervallet mellan några få nanometer upp till tiotals mikrometer. Dessa partiklar påverkar klimatet genom både direkt interaktion med strålning och genom inverkan på molnens bildning, reflektans och livstid. Det arbete som presenteras i denna avhandling syftar till att öka vår förståelse rörande drivande processer och mekanismer som är involverade i regleringen av både säsongsvariation och långsiktiga förändringar av den Arktiska aerosolen samt analysera aerosolens generella livscykel. I det föränderliga Arktis har emissionerna av både antropogena och naturliga aerosolpartiklar genomgått förändringar över tid, och dessa förväntas fortgå. Till exempel kan nämnas att långväga transport av antropogena aerosoler till Arktis fortsatt kommer minska till följd av ytterligare emissionsreduktioner.

Observationer av den Arktiska aerosolen har genomförts vid en forskningsstation på Svalbard. I denna avhandling har flertalet olika instrument och mätningar tillämpats för att utvärdera olika variabler kopplade till aerosoler vad gäller både säsongsvariation och förändring över längre tidsskalor. Detta arbete kan visa på att en signifikant minskning i koncentrationen ljusabsorberande partiklar skett de senaste två årtiondena, varav den största minskningen kan härledas till bidrag från norra Sibirien. Avhandlingen gör även gällande att en fjärdedel av den totala minskningen kan relateras till förändrade sänkprocesser kopplade

till våtdeposition. Vidare studeras hur olika miljövariabler förändrats under transporten av luftmassor till mätstationen. Utifrån detta perspektiv kan visas att nederbörd både kan fungera som källa och sänka med en storleksberoende påverkan på partikelantalet, medan solstrålning påvisats främja en ökning av antalet partiklar över hela storleksintervallet. Vidare, genom analys av den första långsiktiga mätserien av ljusabsorberande partiklar i och utanför moln, studeras upptaget av partiklar i molndroppar genom så kallad ”nucleation scavenging”. Ett ökat upptag av dessa ljusabsorberande partiklar i molndroppar noteras mellan april-oktober. Hur effektivt upptaget sker beror av både temperatur och mängd molnvatten. Slutligen visar avhandlingen på att nybildning av små partiklar, knappt 1nm stora, i Svalbards närområde kraftigt påverkas av solstrålning och den omgivande aerosolens ytarea. Grönlandshavet visas vara en relativt sett större källa till dessa partiklar jämfört med angränsande hav. Analysen visar vidare att den totala ytarean hos de omgivande, redan existerande partiklarna, reduceras av moln och nederbörds processer i luftmassor. Detta i sin tur skapar förutsättning för nypartikelbildning och återskapande av aerosolpopulationen vid soligt väder.

Gällande generalisering och förståelse för hur dessa resultat kan appliceras på större geografiska områden så lämnas frågan öppen. Vidare kräver en kvantifiering av hur de påvisade mekanismerna och förändringarna påverkar strålningsbalansen i Arktis ytterligare forskningsinsatser.

List of papers

The list of following papers are included within this doctoral thesis:

I. **Heslin-Rees, D.**, Tunved, P., Ström, J., Cremer, R., Zieger, P., Riipinen, I., Ekman, A. M. L., Eleftheriadis, K., and Krejci, R.: Increase in precipitation scavenging contributes to long-term reductions of light-absorbing aerosol in the Arctic, Atmos Chem Phys, 24, 2059–2075, https://doi.org/10.5194/acp-24-2059-2024, 2024.

D.HR. processed the light-absorption coefficient data set, analysed all the data, and wrote the manuscript with input from co-authors.

II. Khadir, T., Riipinen, I., Talvinen, S., **Heslin-Rees, D.**, Pöhlker, C., Rizzo, L., Machado, L. A. T., Franco, M. A., Kremper, L. A., Artaxo, P., Petäjä, T., Kulmala, M., Tunved, P., Ekman, A. M. L., Krejci, R., and Virtanen, A.: Sink, Source or Something In-Between? Net Effects of Precipitation on Aerosol Particle Populations, Geophys Res Lett, 50, https://doi.org/10.1029/2023GL104325, 2023.

D.HR. helped analyse the size distribution and back trajectory data concerning the Zeppelin Observatory, and provided input into the manuscript.

III. Zieger, P., **Heslin-Rees, D.**, Karlsson, L., Koike, M., Modini, R., and Krejci, R.: Black carbon scavenging by low-level Arctic clouds, Nat Commun, 14, https://doi.org/10.1038/s41467-023-41221-w, 2023.

D.HR. performed the back trajectory calculations, helped analyse the data, and along with P.Z. wrote the manuscript with input from all co-authors.

IV. **Heslin-Rees D.**, Tunved P., Aliaga D., Lampilahti J., Riipinen I., Ekman A. M. L., Park K. T., Mazzini M., Gilardoni S., Thakur R., Park K., Yoon Y. J., Lee K., Sipilä M., Mazzola M., Krejci R.: Drivers governing the seasonality of new particle formation in the Arctic. Aerosol Res., (submitted).

D.HR. set up the NAIS instrument at Zeppelin Observatory with R. K., analysed the data, performed the back trajectory calculations, and wrote the manuscript with input from co-authors.

List of papers not included in the thesis**:**

I. Aliaga, D., Sinclair, V. A., Krejci, R., Andrade, M., Artaxo, P., Blacutt, L., Cai, R., Carbone, S., Gramlich, Y., Heikkinen, L., **Heslin-Rees, D.**, Huang, W., Kerminen, V.-M., Koenig, A. M., Kulmala, M., Laj, P., Mardoñez-Balderrama, V., Mohr, C., Moreno, I., Paasonen, P., Scholz, W., Sellegri, K., Ticona, L., Uzu, G., Velarde, F., Wiedensohler, A., Worsnop, D., Wu, C., Xuemeng, C., Zha, Q., and Bianchi, F.: New particle formation dynamics in the central Andes: contrasting urban and mountaintop environments, Aerosol Research, 3, 15–44, https://doi.org/10.5194/ar-3-15-2025, 2025.

II. Freitas, G. P., Adachi, K., Conen, F., **Heslin-Rees, D.**, Krejci, R., Tobo, Y., Yttri, K. E., and Zieger, P.: Regionally sourced bioaerosols drive high-temperature ice nucleating particles in the Arctic, Nat Commun, 14, https://doi.org/10.1038/s41467-023-41696-7, 2023.

III. Freitas, G. P., Kopec, B., Adachi, K., Krejci, R., **Heslin-Rees, D.**, Yttri, K. E., Hubbard, A., Welker, J. M., and Zieger, P.: Contribution of fluorescent primary biological aerosol particles to low-level Arctic cloud residuals, Atmos Chem Phys, 24, 5479–5494, https://doi.org/10.5194/acp-24-5479-2024, 2024.

IV. Gilardoni, S., **Heslin-Rees, D.**, Mazzola, M., Vitale, V., Sprenger, M., and Krejci, R.: Drivers controlling black carbon temporal variability in the lower troposphere of the European Arctic, Atmos Chem Phys, 23, 15589–15607, https://doi.org/10.5194/acp-23-15589-2023, 2023.

V. **Heslin-Rees, D.**, Burgos, M., Hansson, H. C., Krejci, R., Ström, J., Tunved, P., and Zieger, P.: From a polar to a marine

environment: Has the changing Arctic led to a shift in aerosol light scattering properties?, Atmos Chem Phys, 20, 13671–13686, https://doi.org/10.5194/acp-20-13671-2020, 2020.

VI. Kommula, S. M., Buchholz, A., Gramlich, Y., Mielonen, T., Hao, L., Pullinen, I., Vettikkat, L., Ylisirniö, A., Joutsensaari, J., Schobesberger, S., Tiitta, P., Leskinen, A., **Heslin-Rees, D.**, Haslett, S. L., Siegel, K., Lunder, C., Zieger, P., Krejci, R., Romakkaniemi, S., Mohr, C., and Virtanen, A.: Effect of Long-Range Transported Fire Aerosols on Cloud Condensation Nuclei Concentrations and Cloud Properties at High Latitudes, Geophys Res Lett, 51, https://doi.org/10.1029/2023GL107134, 2024.

VII. Platt, S. M., Hov, Ø., Berg, T., Breivik, K., Eckhardt, S., Eleftheriadis, K., Evangeliou, N., Fiebig, M., Fisher, R., Hansen, G., Hansson, H. C., Heintzenberg, J., Hermansen, O., **Heslin-Rees, D.**, Holmén, K., Hudson, S., Kallenborn, R., Krejci, R., Krognes, T., Larssen, S., Lowry, D., Myhre, C. L., Lunder, C., Nisbet, E., Nizzetto, P. B., Park, K. T., Pedersen, C. A., Pfaffhuber, K. A., Röckmann, T., Schmidbauer, N., Solberg, S., Stohl, A., Ström, J., Svendby, T., Tunved, P., Tørnkvist, K., Van Der Veen, C., Vratolis, S., Yoon, Y. J., Yttri, K. E., Zieger, P., Aas, W., and Tørseth, K.: Atmospheric composition in the European Arctic and 30 years of the Zeppelin Observatory, Ny-Ålesund, Atmos Chem Phys, 22, 3321–3369, https://doi.org/10.5194/acp-22-3321-2022, 2022.

VIII. DeMott P. J., Swanson B. E., Creamean J. M., Tobo Y., J. Hill T. C., Barry K. R., Beck I. F., Frietas G. P., **Heslin-Rees D.,** Lackner C. P., Schmale J., Krejci R., Zieger P., Geerts B., and Kreidenweis S. M.: Ice nucleating particle sources and transports between the Central and Southern Arctic regions during winter cold air outbreaks, Elementa: Science of the Anthropocene, (in review)

IX. Herrero-Anta S., Eckhardt S., Evangeliou N., Gilardoni S., Graßl S., **Heslin-Rees D.**, Krejci R., Mateos D., Mazzola M., Ritter C., Stebel K: Exceptional high AOD over Svalbard in Summer 2019: A multi-instrumental approach, (to be submitted early 2025)

X. Mazzini M., Gysel Beer M., Brem B. T., Modini R. L., Aliaga D., Lamphilati J., **Heslin-Rees D.**, Cristofanelli P., Bianchi F. and Marinoni A.: Aerosol size distribution and new particle formation in high mountain environments: a comparative analysis of long-term observation at Monte Cimone and Jungfraujoch GAW Global stations (to be submitted early 2025)

Abbreviations

AA	Arctic Amplification
ACI	Aerosol-cloud interaction
ARI	Aerosol-radiation interaction
AR	Aerosol Radiation
BC	Black carbon
CCN	cloud condensation nuclei
CPC	condensation particle counter
CS	Condensation Sink
DMA	differential mobility analyser
DMPS	differential mobility particle sizer
DMS	Dimethyl Sulphide
eBC	Equivalent Black carbon
ERF	Effective Radiative Forcing
FeBC	Scavenged fraction of Black Carbon
GCVI	(ground-based) counterflow virtual impactor
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory model
INP	Ice nucleating particle
MAAP	multi-angle absorption photometer
MAC	Mass absorption cross-section
NAIS	neutral cluster and air ion spectrometer
NPF	New particle formation
PNSD	Particle number size distribution
SA	Sulphuric acid
SI	supplementary information
WBF	Wegener–Bergeron–Findeisen (process)
ZEP	Zeppelin Observatory

Contents

"All I've ever wanted is to see this crazy, beautiful world that little bit clearer,"

"You just want to fill it with smoke."

Dr Ben Santer talking to Don Pearlman, Kyoto, London's Westend, 2025

Introduction

notes on a journey into the Arctic…

Let me introduce you to the absorbing and scintillating world of Arctic aerosols. For this, we will need to wrap up warm as we prepare ourselves to travel beyond the Arctic circle and to the archipelago of Svalbard, where our scientific journey takes place.

Here, in the first part of this thesis, we hope to confer all the necessary tidbits of information necessary to contextualise the studies which follow. It is not easy descending into this world, especially when we cannot see what we are looking for – generally speaking aerosol particles, immersed in the atmosphere, are so small they cannot be seen with the naked eye.

We will familiarise you with the Arctic and the changes taking place there, before switching gears to discuss aerosols, and how they relate to the changing Arctic. For this, we will delve into the climatic importance of aerosols, and the fact that they are an essential ingredient for cloud formation. We will also discuss the typical lifecycles of Arctic aerosols from the perspective of source to sink, how we are able to measure them in the first place, and what we have learnt from long-term monitoring.

After setting the scene, we will discuss the four main studies that make up this thesis, all of which are built upon observations from our measurement site in the Arctic, Zeppelin Observatory (hereafter, referred to as ZEP). The observatory is sat 474 metres above sea level along the ridge of Zeppelinfjellet, overlooking the old mining village of Ny-Ålesund and the Kongsfjord, which is dammed by glaciers. The thesis is composed of the following 4 papers:

- **Paper I** explores the long-term changes in light-absorbing aerosol particles. In **Paper I**, we examine the potential role of not just sources but also sinks, in determining trends.
- **Paper II** expands on the role of sinks to investigate whether precipitation not only acts as a sink, but if it can also be a source of particles.
- For **Paper III**, we look in detail at cloud scavenging, a part of the sink process, by which particles are incorporated into clouds.
- Finally, in **Paper IV**, we set our sights on natural secondary aerosol particles formed in situ, (as opposed to those originating from direct

emission) and explore the environmental conditions governing this process of formation.

In the final part, we will place the findings in this thesis into a broader scientific context and discuss how they fit in to the bigger picture of aerosol and climate science. The prevailing motivation for the thesis is to understand processes and changes on various temporal and spatial scales related to one of the main Arctic climate forcing agents, atmospheric aerosols. The Arctic climate is changing at an unprecedented rate due to the main climate forcers of greenhouse gases and aerosols. Understanding the changes in terms of the amount and in regard to the underlying lifecycle processes governing aerosol concentrations forms the backbone of thesis. For a detailed list of research questions refer to Sect. 5.

The Arctic

The Arctic is considered by many climate scientists to act as a "canary in the coal mine", essentially becoming an early warning system for climatic changes. The Arctic region is witnessing several unprecedented changes, including ongoing and significant increases in surface temperatures and dramatic reductions in summertime sea ice coverage and thickness, to name just a couple (Meredith et al., 2019; AMAP., 2021; Kim et al., 2023). Its status as a rapidly changing region is a far cry from the once previous perceived notion that it was somehow unimpacted by human activities. The future of the Arctic is a concern of global importance just on its one, but even so there is growing evidence to suggest that the changes taking place in the Arctic are not restricted to the region itself, but can also affect the midlatitudes (Francis and Vavrus, 2012; Coumou *et al.*, 2018; Ding *et al.*, 2024). The Arctic region has been the target of several monitoring and assessment programmes including the Global Atmosphere Watch (WMO/GAW), Integrated Carbon Observation System (ICOS), The Aerosol, Clouds, and Trace Gases Research Infrastructure (ACTRIS) and the co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe *(*EMEP). As a result, vast amounts of atmospheric data, in many cases on climatological relevant timescales has been gathered; this data has served as a treasure trove for

process, source-receptor relationship, life-cycle and trend analysis studies. It is upon parts of these data that this thesis is built.

2.1 Ny-Ålesund, Svalbard

"it's either scientists

Or people who are mining

Or dog sledge people"

Dry Cleaning, taken from the song Anna Calls From the Arctic, 2022

Svalbard (74-81°N) is an archipelago situated in between the Arctic Ocean, Norwegian, Greenland and Barents seas. Importantly for the purposes of this thesis it is home to the research village of Ny-Ålesund (see Fig. 1).

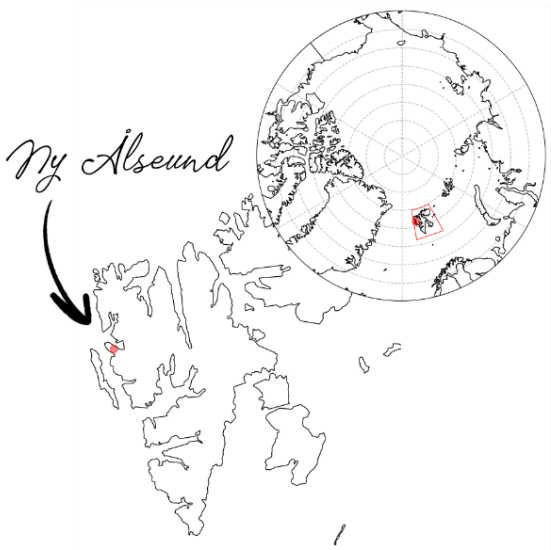

Fig 1: The location of Svalbard, halfway between the northern coast of Norway and the North Pole. The red dot signifies the location of the research village of Ny-Ålesund, whilst the red rectangle outlines the location of the archipelago of Svalbard.

Established in the early 20th century due to its coal deposits, the village of Ny-Ålesund struggled for some time, contending with unprofitability, disasters,

and strikes, until mining operations eventually ended in 1962. Nowadays, Ny-Ålesund (see Fig. 2), is home to the most well-equipped Arctic research base thanks to the joint efforts of eighteen countries from across the globe, specialising in research fields such as oceanography, glaciology and atmospheric science (https://nyalesundresearch.no/). It cannot go, however, unnoticed that there is an odd sense of irony in the fact that this research facility, which primarily focuses on the effects of climate change, came about because of coal mining. The research carried out in and around the village is extensive, world-leading, and vital to long-term environmental monitoring. However, it is perhaps important to bear in mind the geopolitical significance of the Arctic; it can be argued that research stations such as Ny-Ålesund have enabled countries to maintain a semblance of interest in the region.

Fig 2: The snowy research village of Ny-Ålesund, captured on 5 May 2023, with Zeppelin Observatory and the world's northernmost post office highlighted.

2.2 Arctic Climate

If one were to stand on the Earth at 66° 34'N during the winter solstice, the Sun would fail to rise above the horizon. If one were to return to that line on the summer solstice, the Sun would not set. The polar night describes the period

when the Sun is continuously below the horizon, and the terms polar day or midnight sun refer to the period of ever-present sunlight. This line of latitude defines the Arctic circle and serves as one of many definitions for the Arctic region. Beyond this line, we enter the Arctic.

The Arctic, like all other places, receives the same amount of daylight hours (averaged over the course of a year). However, the short growing seasons (defined agriculturally) and harsh, cold, and dark winters that arise from these extreme shifts in daylight hours make the Arctic a unique and fascinating environment to study. Another characteristic feature of the Arctic is that it is primarily ocean—albeit an ocean covered by ice; much of the Arctic is covered by ice for long periods of the year. The sea ice acts as a natural thermostat, helping to regulate the Earth's climate by reflecting back incoming solar radiation. Despite the fact that the region beyond the Arctic Circle accounts for only 4% of the Earth's surface, changes to the Arctic climate can have profound impacts on sea level rise, weather, ocean currents and biodiversity.

2.3 Arctic Clouds

The Arctic surface temperature is significantly influenced by clouds; thus, the presence of clouds and surface temperature are inextricably linked. In the Arctic, low-level mixed-phase clouds (MPCs), a mixture of liquid droplets and solid ice crystals—are fairly common. Typically, MPCs are present 30-50% of the time in the Arctic, and for Svalbard in and around Ny-Ålseund, the occurrence of MPCs is above the Arctic average (i.e., 55%) (Mioche *et al.*, 2015). Characteristically, for Svalbard, these clouds are mainly brought about via westerly winds, which bring with them more moist air masses (Gierens et al., 2020). MPCs are the most important cloud type in terms of influencing surface temperature (Shupe and Intrieri, 2004). To comprehend the changes taking place in the Arctic, one must understand the influence Arctic clouds can impart on the radiative balance of the Arctic.

Clouds directly influence the atmospheric radiative fluxes by providing a downward infrared flux and reflecting shortwave radiation. Cloud radiative effects depend on both cloud macro and microphysical properties, as well as

the underlying surface properties. Generally speaking, outside the Arctic, low-level clouds tend to cool the planet (Sedlar *et al.*, 2011; Kay *et al.*, 2016; Sledd and L'Ecuyer, 2019, 2021), however, in the Arctic the opposite is the case. Clouds have an overall warming effect on the surface of Svalbard for example; negative in the summer and positive the rest of the year (Curry *et al.*, 1996; Ebell *et al.*, 2020).

One example highlighting the importance of understanding future changes in cloud cover in our continually perturbed climate concerns the potential consequences of an ice-free Arctic. Due mainly in part to the large changes in surface albedo, an ice-free Arctic would add an additional annual global radiative heating of 0.71 Wm^{-2}. Equivalent to emitting an additional one trillion tones of CO_2 (Pistone, Eisenman and Ramanathan, 2019). For comparison, to date, a total of 1.77 trillion tones of CO_2 have been emitted into the atmosphere since preindustrial times (Global Carbon Budget., 2023). The relevance of this to clouds is that this conclusion assumes constant cloudiness and a major source of uncertainty in this prediction is how cloud cover over the Arctic Ocean may change in the future, particularly in response to retreating sea ice.

2.4 Atmospheric circulation in regards to the Arctic

Climatological global circulation patterns describe the air movements that emerge once you begin to average weather systems over many years, whilst atmospheric large-scale circulation patterns are instantaneous phenomena. The prevailing circulation patterns arise from differences in the amount of heat different parts of the globe receive. For our purposes, we are mainly interested in how atmospheric circulation patterns can influence the Arctic climate and also affect the transport of airborne material up to the Arctic.

The Arctic Oscillation (AO) describes the differences in pressure between the Arctic and the mid-latitudes, and is somewhat similar to the North Atlantic Oscillation (NAO) phenomenon. When the AO is in its positive phase, the air pressure over the Arctic is lower than average, while air pressure over the mid-latitudes is higher than average. The positive AO phase results in the jet stream being shifted further northwards, with the consequence that there are fewer cold outbreaks across the mid-latitudes. Conversely, the negative phase of the

AO, consists of higher than average air pressures over the Arctic and lower than average air pressures over the mid-latitudes. During this phase the jet stream becomes more meandering, allowing Arctic air masses to encroach into the mid-latitudes, bringing with them colder polar air and Arctic weather. The AO transitions from positive to negative phases continuously. In recent years, comparing 1993-2003 to 2004-2014, we have seen a shift to more negative AO indices, leading to more meridional transport (south-to-north or vice versa) (Maturilli and Kayser, 2016). Previous studies have shown that transport through the North Atlantic has become more frequent in the last decades (Mewes and Jacobi, 2019). Furthermore, it has been suggested that long-term trends in the NAO can influence long-term trends in Arctic pollution (Stohl, 2006)

The volume of cold polar air above the Arctic is often referred to as the polar dome, with the Arctic front as its boundary (Bozem *et al.*, 2019). The Arctic front acts a transport barrier for the warmer air masses at lower latitudes, decoupling the Arctic from the influence of the mid latitudes. Only air masses which have experienced diabatic cooling (i.e. external forcing) are able to enter the polar dome (Stohl, 2006). The position of the boundary varies over time and space, which enables air masses influenced by pollution sources further south to become a part of the polar dome. Typically, the polar front advances in the winter to include more emissions coming from Eurasia. In the summertime, the polar front retreats, and ZEP is found more often outside of the polar front receiving mainly marine air masses from the Atlantic Ocean (Dallósto *et al.*, 2017).

2.5 Arctic amplification

"places that alter their albedo through the progression or regression of snow-cover, should in turn feel more of an effect than those places at lower latitudes"

Arrhenius, 1896

Arctic amplification (AA) is the phenomenon describing the increased rates of warming observed in the Arctic, relative to the global average. The degree of AA depends on the time window in question and the latitude (see Fig. 3 taken

from Rantanen et al. (2022)). Regions further north have typically experienced higher warming rates. With respect to our time window (i.e. using data from the early 2000s) and the latitude of Ny-Ålesund (i.e. 79°N), we can expect an AA of between 4.5 and 5.1 (see Fig. 3). In absolute terms, Ny-Ålesund has exhibited an annual mean surface temperature increase of approximately 1.3 K ± 0.7 K per decade (Maturilli, Herber and König-Langlo, 2015). Winter, in particular, witnessed the largest seasonal rate of increase at +3.1 ±2.4K per decade.

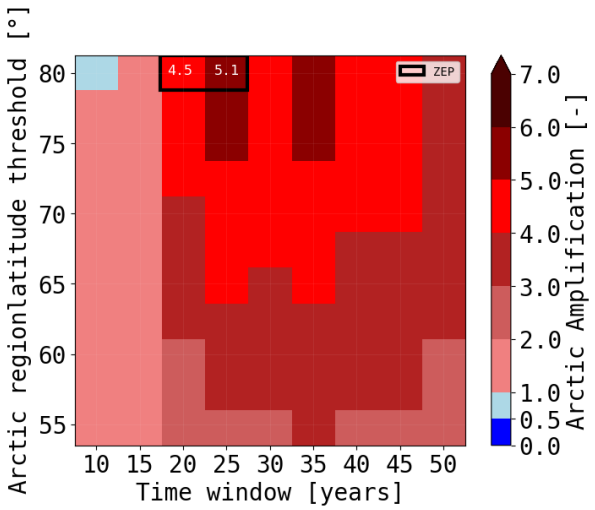

Fig 3: Sensitivity of Artic Amplification (AA) to latitude and period of interest. Grid boxes are coloured based on the near-surface (2m) air temperature rate of increase compared to the global average rate of increase. The grid box representing the Zeppelin Observatory latitude and the measurements that have taken place since the early 2000s/late 1990s (i.e. 79°N & 20-25 years) is highlighted by a black rectangle. Adapted from Rantanen et al. (2022) and based on Coupled Model Intercomparison Project phase 6 (CMIP6).

GHGs and aerosols constitute two of the most significant climate forcers, with GHGs playing the role of the warming agent and aerosols generally acting as the cooling agent. The radiative forcing that results from changes in the emissions of these two agents, give rise to certain Arctic-specific responses, some of which can exacerbate the near-surface air temperature increases, thus giving rise to AA. Examples of Arctic-responses and mechanisms that help to explain AA include the surface-albedo feedback, the limited convection in the

Arctic, and the transport of water vapour and heat into the Arctic from lower latitudes (Taylor *et al.*, 2022); all of these responses can be perturbed by the aforementioned climate forcers. The surface-albedo feedback is considered the most well-known, describing the reduction in surface albedo through decreasing snow and ice, and the enhanced solar heating which follows.

One particularly interesting climate simulation that demonstrates the complexity of AA showed that even with no sea ice present the Arctic displayed amplified warming, with sea-surface temperatures warmed by 1.5 times the global average (Henry and Vallis, 2021). This finding, suggests that AA cannot solely be explained by the surface-albedo feedback.

Aerosols, the Arctic & clouds

3.1 The climatic importance of aerosols

"If there was no dust in the air there would be no fogs, no clouds, no mists, and probably no rain"

John Aitken, 1881

To begin, let's establish why one should care about aerosols in the context of a changing world—and more specifically, in a changing Arctic.

Greenhouse gases, especially carbon dioxide (CO_2), are considered to be the main driver increasing the Earth's surface temperature. According to the most recent IPCC report total human-caused global surface temperatures have risen by approximately 1.07°C (0.8 – 1.3°C) since the preindustrial era (IPCC, 2021). GHGs are estimated to have contributed 1-2°C, whilst aerosols have acted as a cooling counterforce, offsetting this warming by approximately 0.0 – 0.8°C (IPCC, 2021). These two climate forcers lead to opposing effects, which suggests that there are competing interests at play when it comes to improving air quality. Reductions in the emission of particles and their precursors can abate related health issues, but reducing aerosol emissions could at same time

exacerbate the climate problem. It could even be argued that further clean air policies and continued reductions of anthropogenic aerosols make the case for increased GHG emission control.

Effective radiative forcing (ERF) is the term used to describe the balance between incoming and outgoing radiation, where a positive ERF leads to warming and a negative ERF leads to a cooling. Aerosol ERF includes both direct (i.e. aerosol-radiation interaction, ARI) and indirect (i.e. aerosol-cloud interaction, ACI). Aerosol direct effects describe the interaction these particles have with radiation, either by absorbing or reflecting short and longwave radiation; warming aerosols, such as black carbon, generally absorb energy, whereas cooling aerosols (e.g. sulphate, nitrate and organics) reflect incoming shortwave radiation. It is important to note that no single aerosol is purely absorbing or scattering, instead they incorporate elements of both of these optical properties, and the absorption and scattering efficiency vary with size, chemical composition and the wavelength of incident radiation. Aerosol-cloud interactions arise from the ability of aerosols to act as cloud condensation nuclei (CCN) or ice nucleating particles (INPs), seeding cloud droplets and ice crystals respectively.

Aerosol particles act as a critical ingredient for cloud formation because they provide a surface for water to condense on, which occurs much more readily compared to the homogenous nucleation of water droplets. The ability of particles to act as cloud seeds is dependent on their size and their chemical composition, generally speaking, large, soluble particles make the best CCNs. Understanding how size and chemistry impacts the formation of cloud droplets, and the resulting impact on cloud physics and dynamics, as well as precipitation efficiency, are regarded as questions of central importance within atmospheric aerosol science. We have known for some time that perturbing the number of aerosols, impacts the number of CCN, which in turn can alter certain cloud properties including brightness and the lifetime of clouds; more CCNs results in brighter (i.e. 1st indirect effect or Twomey effect; S. Twomey, 1977) and more long-lived (Albrecht effect; Albrecht, 1989) clouds. Some recent studies have shown effectively how, for example, enhanced aerosol concentrations from volcanic eruptions can induce increased cloud cover (Chen *et al.*, 2022, 2024). The uncertainties surrounding the effect aerosols have on the climate is large, with most of the uncertainties pertaining to their indirect effects on clouds.

3.1.1 From an Artic perspective

Like the rest of the climate, the Arctic is sensitive to short-lived pollutants such as aerosols. Attempts have been made to examine the influence GHGs and anthropogenic aerosols exert on the Artic, and to tease out exactly how the Arctic responds to changes in their emissions (Shindell and Faluvegi, 2009; Navarro *et al.*, 2016; Stjern *et al.*, 2019; Ren *et al.*, 2020).

Wu et al. (2024) suggest that AA induced by anthropogonic aerosols was stronger than the AA induced by GHGS, during the period when there was the strongest increase in anthropogonic aerosol concentrations (i.e. 1955-1984). Since the 1980s, sulphate emissions have reduced across Europe and North America resulting from the implementation of major air quality regulations and clean air policies (e.g. US Clean Air Act). Navarro et al. (2016) argued that this reduction in sulphate has warmed the Arctic and can explain a significant fraction of the overall AA. It is therefore possible to imagine that recent reductions in anthropogenic aerosol concentrations (see Aas et al. (2024) for European trends since 2000) could have important implications, and help explain the most recent warming of the Arctic and the strengthening of AA (Chylek *et al.*, 2022).

If only anthropogenic aerosols that act as warming agents (e.g. black carbon, BC) are considered, any mitigation will most likely lead to negative radiative forcing, helping to stem the extent of AA (especially in terms of direct aerosol forcing). However, the exact impact will depend on the vertical and seasonal distribution of BC. From an Arctic perspective, the altitude at which the warming agents reside in the atmosphere is important. BC in the lowest layer of the atmosphere causes a very strong surface warming, as a result of cloud and sea-ice feedbacks (Flanner, 2013). BC at higher altitudes has been shown to cool the surface, by increasing cloud cover beneath (AMAP, 2015). The altitude at which BC resides in the Arctic after being emitted and transported depends on the latitude of its origin; generally speaking BC emitted at lower latitudes which reaches the Arctic is more likely to reside at higher altitudes (AMAP, 2015). Combining these ideas, it can be ascertained that BC emissions within the Arctic have a greater impact per tonne on the surface air temperature

(Sand *et al.*, 2013). Nonetheless, local emissions of BC, despite increasing, remain a small fraction of the overall within-Arctic BC (AMAP, 2015). A striking example and slight expansion of these ideas comes from Jacobson et al. (2012), who suggested that re-routing flights away from the Arctic would have a net benefit. This is because BC would be removed more easily from the atmosphere as lower latitudes tend to induce more wet scavenging.

3.2 Atmospheric Aerosols

"Science is the systematic classification of experience"

George Henry Lewes

Now, we know that aerosols do indeed influence our climate, and that the Arctic is particularly sensitive to changes in this climate forcer. But what do we know about atmospheric aerosols, and where do we need to improve our knowledge to better assess their effects?

To begin with, it is important to recognise that not only does the climate interact with aerosol particles, but we also do as humans. Aerosol particles pose as the most important environmental problem in relation to human health, with an estimated 7 million premature deaths ascribed to air pollution every year (Lelieveld *et al.*, 2015). This thesis does not relate to the health effects of aerosols; however, it is worth bearing in mind that we are constantly interacting with aerosol particles with every breath we take. We exchange approximately 500 cm^3 of air (i.e. your average 0.5 litre water bottle) with each breath. Depending on, where in the world you live the concentration of particles present per an average cubic centimetre of air can vary dramatically; in Stockholm perhaps it is 10,000, whilst in some of the most polluted corners of the planet number concentrations could reach 10,000,000 – this makes for a lot of particles exchanged.

Atmospheric aerosols are defined as liquid or solid particles suspended in the air. Aerosol particles range in size from a few nanometres up to hundreds of micrometres, spanning over several orders of magnitude. Mass and size,

however, do not scale linearly. To put the size range into perspective, it's worth noting that 1 billion 10 nm particles are equivalent in mass to one 10-micron particle.

The lifetime of aerosol particles within the lower part of the Earth's atmosphere called the troposphere is controlled by an intricate balance between sources and sinks. This leads to a relatively short lifetime – within the timescale of days to weeks. Atmospheric circulation is incapable of homogenously mixing aerosols within such a short period, therefore, compared to long-lived GHGs, concentrations of particulate matter are more variable.

Aerosol particles are categorised on the basis of their size, their source either being natural or anthropogenic, and the processes behind their presence in the atmosphere. Sources of both natural and anthropogenic aerosols are wide ranging; examples of natural aerosols include windblown desert dust, sea salt aerosol produced from breaking waves and bubble bursting, and sulphuric acid formed via the oxidation of vapours emitted by phytoplankton. On the flip side, anthropogenic aerosol particles include soot released from incomplete combustion, and the now ubiquitous nanoplastics released into the atmosphere directly or as a result of resuspension.

The other subdivision used to characterise aerosol particles is based on whether they were directly emitted (i.e. primary aerosol) or whether their production was indirect (i.e. secondary). Secondary aerosols are those which are formed in the atmosphere through a process of gas-to-particle conversion. From the aforementioned examples dust would constitute a primary aerosol, whilst sulphuric acid formed via oxidation processes would be deemed secondary.

3.2.1 Particle number size distributions

Central to aerosol science is the concept of particle number size distributions (PNSDs). It is important we count particles based on their size, as size largely controls the lifetime, optical properties, and CCN efficiency of aerosol particles. It is not feasible to both count and measure the diameters of every single

particle, and so instead, particles are divided based on their size into discrete intervals, i.e. size bins, and then the number of particles present in each bin is counted. This gives rise to the PNSD, which one can essentially think of as a histogram. A typical PNSD will have the x-axis representing diameter (Dp) (i.e. size, given in nanometres or microns), and the y-axis will reflect the concentration of particles (i.e. given in number of particles per cubic centimetre). Each distribution depends on the binning methods and/or the instrument used. One trick utilised by aerosol scientists to compare measurements of different distributions is to normalise the PNSDs by the size bin used. PNSD data generally comes in the form of dN/dlogDp (i.e., normalised, conveying the number of particles per unit logarithmic diameter interval). By finding the width of each size bin, and multiplying the given dN/dlogDp by dlogDp we can get the particle concentration in a bin (dN).

Countless studies have demonstrated that PNSDs can be divided up into modes. By studying many aerosol size distributions, it becomes apparent that common and general patterns are present. Simply put, particles are not homogeneously spread over the various size bins, but tend to be present in modes of different sizes. These modes are the result of various processes controlling the lifecycle of atmospheric aerosols. Different PNSDs arise as a result of diverse emission sources coupled with a range of atmospheric processes. The size distribution is a kind of fingerprint that, in a way connects to the sources and processes that have been most active. The PNSD can be used to see if aerosols have been recently formed, or if the measured aerosols have been processed by clouds or have undergone significant aging.

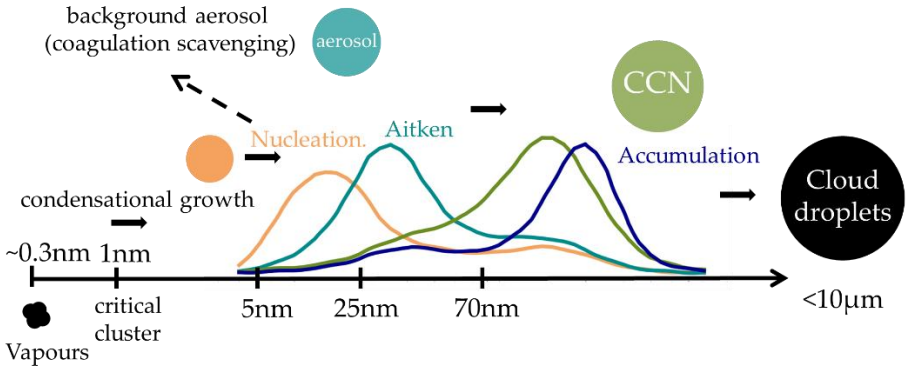

Fig 4: Examples of common particle number size distributions measured in the Arctic. The orange curve represents the nucleation mode particles. The cyan curve accounts for the Aitken mode particles (which have typically grown from nucleation mode particles). The purple curves consist of a large number of accumulation mode particles. DMPS data from ZEP was used to generate curves. For the PNSDs the y-axis (not shown) represents the particle number concentration, with units cm^{-1}. Figured adapted from Westervelt et al. (2013).

We can characterise the different PNSDs into a set of common modes namely and in order of size the nucleation, Aitken, accumulation and coarse mode. The nucleation mode (<10nm) comes about from the fresh production of particles that nucleate out of the gas phase (orange PNSD curve in Fig 4). Aitken mode particles (10-100nm) can represent growing nucleation mode particles primarily emitted particles in different stages of growth (cyan PNSD curve in Fig 4). Both nucleation and Aiken mode particles have a relatively short lifetime and coagulate easily. The accumulation mode (0.1 to 1 micron) typically represents the upper range to which particles can grow by condensation, and a dominant accumulation mode signifies an aerosol in typically aged, polluted airmasses (purple PNSD curve in Fig 4). As the name suggests, accumulation mode particles have a relatively long lifetime, as these particles are the least prone to dry scavenging, and loss by diffusion – generally only through clouds can accumulation mode particles be removed. Finally, the coarse mode (>1 micron) generally consists of particles which have been produced mechanically such as dust, pollen and sea salt. Coarse mode particles, given their size, typically have a shorter lifespan as then are more susceptible to gravitational settling.

3.3 Lifecycle of Arctic aerosols

The lifecycle of atmospheric aerosols in the Arctic is important to understand, and will help place this thesis into the larger context.

Figure 5 tries to portray some of the central lifecycle processes affecting Arctic aerosols, and we will refer back to it throughout this subsection. To simplify things, we have chosen to focus on two of the main pathways perturbing the Arctic aerosol budget namely local emissions of gaseous precursor vapours within the Arctic (left-hand side of Fig. 5), which lead to the formation of particles in situ in the Arctic environment, and the direct emission of primary particles from biomass burning or industry from lower latitudes (right-hand side of Fig. 5), which age and grow en-route to the Arctic.

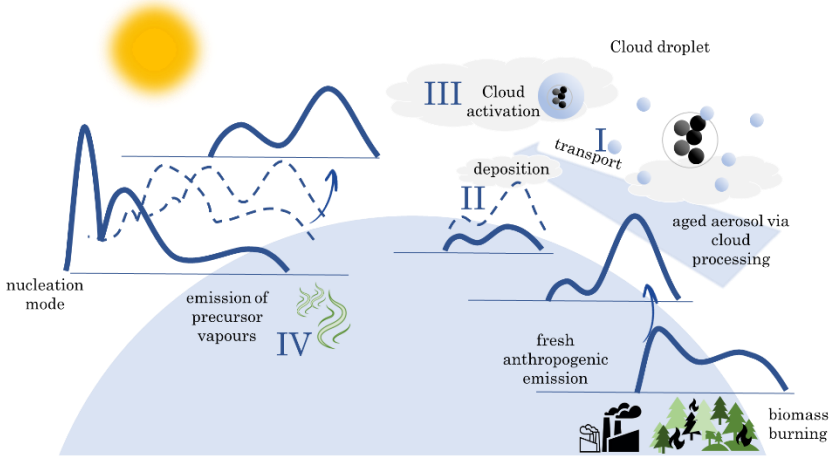

Fig. 5: Here, we have chosen to focus on particular sources of aerosols, given the content of this thesis, and the reader should bear in mind that this is of course an oversimplification of the sources of particles observed in the Arctic; we are not mentioning for example sea spray aerosol and mineral dust. The roman numerals highlight the papers which are centred on or feature the aerosol processes (**Papers I-IV**).

We can say that the life of a particle begins, when it is introduced into the atmosphere, either by being emitted as a primary aerosol particle e.g. the fresh anthropogenic emissions from industry, vegetation or the Earth's surface or by

being formed in the atmosphere as a result of gas-to-particle conversion, also known as nucleation.

Once introduced into the atmosphere, the next step in this simplified version of the lifecycle of an aerosol is transport. By transport, we refer to the journey that aerosol particles take themselves on to end up in the Arctic, and like any good journey it changes the traveller. Particles can alter themselves depending on who they bump into and what they experience along the way.

For the particles formed through nucleation, they tend to grow through condensation or can coagulate with larger, pre-existing aerosols. A large portion of freshly nucleated particles end up coagulating with the larger pre-existing aerosols, adding material to the predominant accumulation mode. In essence, these newly formed particles lose their identity (we can observe this through the transformation of the PNSD on the left-hand side of fig. 5).

Another way aerosol particles change is via a process commonly known as aging. Aging is a term that combines processes including gas condensation, dissolution, coagulation with other particles, and chemical reactions on and within particles (Carslaw, 2022). Aging processes can alter optical, physical, and chemical properties of the aerosol. Through aging, a particle's ability to uptake water changes and thus their ability to act as effective CCN.

Along the way to the Arctic, clouds play a pivotal role in the aerosol lifecycle. Aerosol particles are incorporated into water droplets via two main ways: either through nucleation scavenging or impaction scavenging (Pruppacher and Klett et al., 1978). By far the most important process is nucleation scavenging (Ohata *et al.*, 2016) and so we will focus only on that here. For nucleation scavenging, cloud droplets form through the activation of aerosol particles. When the conditions for cloud formation are met e.g. high supersaturation, the larger aerosol particles are activated first, becoming cloud droplets, and the smaller interstitial aerosol particles (i.e. unactivated) may diffuse or coagulate into the cloud droplets. Once activated, cloud droplets act as small chemical reactors, efficiently transforming, for example, sulphur dioxide into sulfuric acid. All this chemistry going on in these hydrometers plays a key role in transferring material from smaller particles (i.e. in the Aitken

mode) to the accumulation mode. Particles that travel from afar can undergo cloud processing multiple times (on average, 10 times) in which they are incorporated into cloud droplets, and then are continuously released back into the atmosphere through cycles of evaporation and condensation. When the cloud droplet evaporates at the end of one nonprecipitating cloud cycle the combined outcome of chemical reactions and in-cloud scavenging results in a residue that is larger and generally has a different chemical composition than the original activated particle. Through several iterations of nonprecipitating cloud cycles, the PNSD develops a minimum at the size determined by the supersaturation reached in the average cloud, i.e. essentially corresponding to the typical threshold for CCN-activation – this is known as the Hoppel minimum (Hoppel, Frick and Larson, 1986). For the Arctic, the Hoppel minimum presents itself at around 70 nm for a typical size distribution, but varies widely geographically depending on environmental factors. Essentially, we can say that the effect of nonprecipitating clouds gives rise to the distinctive PNSD we observe at ZEP, where we generally see two characteristic modes namely the smaller Aitken mode and the larger accumulation mode (we can observe this through the transformation of the PNSD on the right-hand side of fig. 5, as the accumulation mode increases at the expense of the Aitken mode).

One important process related to cloud formation which we will touch upon in a preceding paper concerns mixed-phase cloud, and is known as the Wegener–Bergeron–Findeisen (WBF) process. Even though this thesis does not concern itself with INPs, the WBF process affects the efficiency of cloud droplet activation. The presence of ice crystals, which form when INPs are present and the conditions are right, may affect the growth of liquid cloud droplets; because of the lower saturation vapour pressure over ice compared with liquid water, ice crystals grow at the expense of liquid cloud droplets.

The ultimate stage is when aerosols finally settle down and are deposited onto the surface. Deposition can happen via two main routes namely wet and dry scavenging, with wet scavenging being responsible for the majority of deposition (Hales, 1978). Deposition is important as it defines the overall lifespan of particles, and it can alter the albedo of the underlying surface. In the Arctic, for example, the deposition of light-absorbing particles on snow or ice

can lead to a positive radiative forcing, and further enhance the warming of the Arctic. The only effective sink of accumulation mode particles is in-cloud scavenging, while dry deposition is important for smaller and larger particles. We can think of the accumulation mode as a kind of terminal stage, and in order to "reset" the cycle this mode must be scavenged out (i.e. what we observe during the deposition in figure 5, as the accumulation decreases).

3.4 Seasonality of Arctic aerosols

Our site, ZEP, where we measure aerosol particles, exhibits what can be described as a typical Arctic aerosol seasonality, characterised by a sustained increase in the accumulation mode particles (0.05 – 0.5μm) during the late winter and early spring, followed by the appearance of smaller nucleation and Aitken mode particles (1.5nm – 0.05μm) in the summer. The Arctic Haze exhibits the largest mass of aerosol particles, whilst, in terms of number, the summer displays the highest concentrations.

To understand the seasonality of aerosol particles at ZEP, we build on the work done by Tunved, Ström and Krejci (2013). We can partition the annual cycle into three major seasons namely:

1. *Arctic Haze (Feb - May)*
2. *Summer (Jun - Sep)*
3. *Transition or the slow- build-up season (Oct -Jan)*

It is worth noting that most models struggle to faithfully reproduce the aerosol seasonality we observe in the Arctic (see AMAP, 2015 for BC and SO4 comparisons), however, there has been definite progress over the decades (see e.g. Shindell et al. (2008)). There may be many reasons for this, but one reason perhaps is an inability to accurately model fast processes, such as wet scavenging, which are difficult to parameterise, especially within coarse spatial grids (Garrett et al., 2011). Furthermore, the process of nucleation in the Arctic region is still not well understood, which hampers the capability of models to get the concentrations right.

3.4.1 Arctic Haze

We have all encountered hazy conditions, where your visibility is obscured. Haze is caused by particles suspended in the atmosphere that can efficiently scatter and attenuate sunlight, often giving the air a brown or yellowish tint. The Arctic is no different *pre se* and witnesses haze. The prominence of these hazy conditions in the Arctic has a distinctive seasonality, and as mentioned above peaks between February and May.

The Arctic haze phenomenon has been known about for decades, with air force pilots even coining the term "*Arctic haze''* back in the 1950s after reporting haze events of an "unknown origin" during flight campaigns (Shaw, 1995). The pilots even noted that these events were more common between March and May. Going even further back, both the Greenland expedition of Nordenskiöld and the 1882 expedition of Nansen onboard the *Viking*, realised that the surfaces of large ice sheets were not completely clean, but in fact often had a "dirty, greyish, or even brownish hue", with Nordenskiöld hypothesising about whether it was fallen "cosmic dust" (Nordenskiöld, 1874).

We now know though, through research carried out in the 1970s, that the Arctic haze is a result of polluting aerosols that travel thousands of kilometres to reach the Arctic (Shaw, 1995). Under favourable meteorological conditions, anthropogenic aerosol particles, generally originating from Eurasia (from the perspective of Svalbard), are effectively transported over long distances to the Arctic. The presence of these larger accumulation mode particles during the Arctic Haze season occurs every year, typically towards the end of winter and the beginning of spring. The season is characterised by low amounts of precipitation, efficiently reducing the sink of the accumulation mode, which results in high concentrations of accumulation mode particles.

In recent decades, the scientific community has once again renewed its interest in the Arctic haze as a way of explaining the changes taking place in the Arctic climate.

3.4.2 Summer and NPF

"The conclusion that was reached was that the Arctic is clean in summer primarily because it is warmer and wetter"

Timothy Garrett, 2011

In contrast to the Arctic haze season, the summer season is relatively clean, as concentrations of anthropogenic aerosol particles subside. Summer is characterised by a dramatic increase in solar insolation, coupled with increased photochemistry and an increased amount of precipitation. The summertime can experience two to three times higher precipitation rates, compared with winter (Tunved and Ström, 2019). It is the precipitation, in fact, that acts as one of the main controlling factors in regard to the distinct seasonality seen in the Arctic. Wet scavenging efficiently reduces the accumulation mode, and therefore reduces both the coagulation and condensation sinks.

Summertime witnesses the majority of new particle formation (NPF) events. Essentially, what happens is that increased solar insolation allows for more photochemical reactions to occur, and the increased biological activity in the neighbouring seas gives rise to the production of gaseous precursors, which are then oxidised to form condensable vapours. Nucleation of these vapours leads to the formation of stable clusters of molecules. When clusters reach a critical diameter, condensational growth becomes thermodynamically favourable, and these critical clusters subsequently grow via condensation and self-coagulation, becoming new particles (Curtius, Lovejoy and Froyd, 2006). The activation of these clusters can be described by nano-Köhler theory (Kulmala *et al.*, 2004), analogous to the more traditional Köhler theory explaining the activation of CCN. Summer is therefore characterised by the presence of smaller nucleation and Aitken mode particles, given the frequency of these NPF events and the growth through condensation that occurs afterward. The degree to which new particle formation, despite the slow growth witnessed in the Arctic, can influence the population of CCN is an open question. However, the effect on the CCN population is certainly non-negligible.

During the summer months, not only does the Arctic experience NPF, but there are also sporadic high-loading events caused by biomass burning events at lower latitudes, typically in the boreal forests of Russia and North America. We know that boreal forest fires are becoming more frequent and severe (Rogers *et al.*, 2020), despite a reduction in the number of global fires (Andela *et al.*, 2017). These events, coupled with effective northward transport, can lead to significant increases in the aerosol loading in the Arctic.

3.4.2 Transition or the slow- build up season

The transition period or slow-build up season as the name suggests witnesses an annual low both in terms of the number and mass of aerosol particles. The concentrations of both anthropogenic long-range pollutants and newly formed particles are reduced. The slow build up season experiences the start of the polar night when solar radiation in the Arctic begins to rapidly decline, this effectively puts a stop to the photochemical reactions that were present during the sunlight periods. Removal, mainly as a result of high precipitation rates, during this season still tend to be high and therefore the combination of few sources and a high sink, gives rise to particularly clean Arctic atmosphere.

3.5 Long-term trends

Researchers have studied the changes in concentrations of ambient anthropogenic aerosol particles in the Arctic for decades (Platt *et al.*, 2022; Schmale *et al.*, 2022), with sulphate and black carbon being two of the most common proxies for anthropogenic influence. Black carbon in the Arctic for example has gained renewed interest by the scientific community due to its potential warming effect, in particular altering the surface albedo of snow and ice. There have been numerous studies published which have reported on the long-term trends of anthropogenic ambient aerosols (Schmale *et al.*, 2022). Long-term monitoring of atmospheric pollutants has been going on at ZEP for

example since the late 90s, and this type of monitoring has been essential to performing any kind of trend analysis.

The consensus among studies is that the ambient atmosphere in the Arctic is indeed getting cleaner when it comes to anthropogenic aerosols throughout the past few decades. For example, Sharma et al. (2019) stated a 50% reduction in black carbon since 1990, and suggested that the economic decline in former Soviet Union countries, along with improvements in the air quality in Europe, were the main causes for reductions in the measured concentrations of ambient anthropogenic particulate matter in the Arctic (in this case Alert, Canada).

It is widely regarded that current observations, which mostly started after the 1990s, cannot reflect the most important declines in anthropogenic aerosol concentrations in the Arctic (Platt et al., 2022; Schmale et al., 2022). Historically, countries like the United States reached their maximum BC emissions for example in the 1920s, at the peak of coal use and prior to the great depression (Bond, 2010). Nonetheless, given the sensitivity of the Arctic to short-live climate forcers and the prospect of continued changes in the emissions of anthropogenic aerosols, long-term monitoring of these pollutants will remain vital.

Method

4.1 Zeppelin Observatory, Ny-Ålesund

"a bewildering array of instrumentation housed in iron tubes, glass cylinders, and steel bell jars. Some were clicking softly, some described an inky line on a spool of graph paper, and some just sat there looking menacing, coolly calibrated''

Sara Wheeler describing Zeppelin Observatory, taken from her book the Magnetic North: Notes from the Arctic Circle, 2009

In 1926, the airship *Norge* took off from Ny-Ålesund, aided by a 350m long mast (which still stands to this day). In just under 16 hours it had successfully helped humankind reach the North Pole for the first time, albeit 200 meters above the surface. Zeppelin Observatory takes its name from the mountain, Zeppelinfjellet, which in turn is eponymously named after this lighter-than-air type of airship from the 20th century that helped in this human-first.

Fig 6: Zeppelin Observatory on a clear sunny day on 6 April 2022. The observatory is situated 474 metres above sea level, not quite reaching the summit of the mountain. The whole-air inlet is labelled, through this is how we sample the air.

Zeppelin Observatory (see Fig. 6) was set up with the intention of monitoring the changes taking place in the Arctic; it was spurred on by a growing interest

in research surrounding Arctic haze, and the idea that dramatic changes to the Arctic, both in terms of large-scale human exploitation and enhanced warming, were just around the corner. ZEP was situated atop the Zeppelinfjellet with the idea that by minimising any local contamination, and being situated far from any major anthropogenic sources, it could act as a regional background station. Additionally, ZEP remains, for the majority of the time above the surface inversion, which in winter and spring has a depth of a few hundred metres (Hov and Holtet, 1987 see Platt et al. (2022). ZEP began carrying out background measurements in relation to atmospheric composition, both in terms aerosols and trace gases in 1989.

4.2 Measurements and air mass transport model

The sampling of air for the purpose of measuring various aerosol related parameters and/or cloud-properties, along with various other supporting measurements, is performed at the ZEP. A broad range of instrumentation has been used throughout this thesis, but for the sake of the introductory part of this thesis, we will only touch upon a bulk approach method for estimating black carbon, and two size segregation methods for counting the number of particles for different size ranges. In addition to these, we will also discuss the transport model used throughout the studies in question.

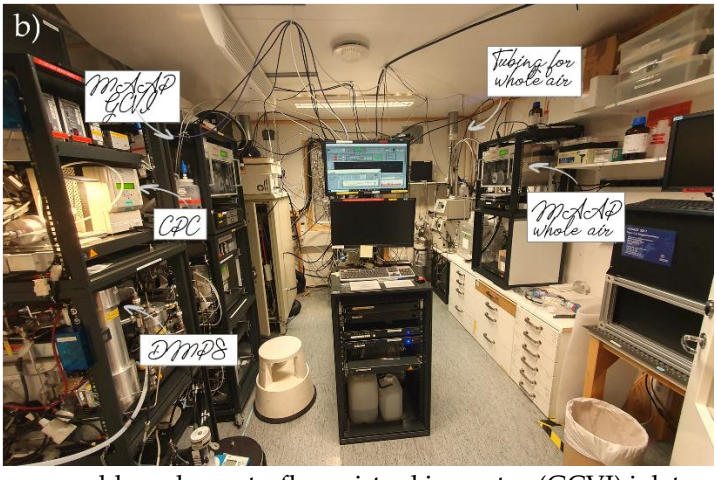

Fig 7: (a) The ground-based counterflow virtual impactor (GCVI) inlet and whole air inlet on top of the Zeppelin Observatory. (b) photo of inside the Stockholm University room at Zeppelin Observatory. The instruments and parts of the set up are labelled. From top-left going clockwise there are as follows: MAAP connected to the GCVI, tubing connected to the whole-air inlet situated on the roof (a), MAAP connected to the whole-air inlet, Differential Mobility Particle Sizer (DMPS), Condensation Particle Counter (CPC).

4.2.1 Light-absorbing aerosol measurements

Measuring light-absorbing aerosol particles works by utilising the ability of these particles to attenuate light. For **Papers I & III**, filter-based absorption photometers were used, namely the multi-angle absorption photometer (MAAP, Petzold and Schönlinner, 2004) and the particle soot absorption photometer (PSAP). These instruments work by measuring the change in the

light transmission induced by laden aerosols deposited on a filter (see Fig. 8). Through knowing the throughflow of air, the concentration of light-absorbing particles in the sampled air can be determined. The amount of light absorbed is described as the attenuation of light per given unit length, and is given in respect to the wavelength of the light source used in measuring the filter. The parameter we ultimately measure is the light-absorption coefficient ($\sigma_{ap}(\lambda)$). Using the $\sigma_{ap}(\lambda)$ we can also ascertain the equivalent black carbon concentration (eBC) by multiplying $\sigma_{ap}(\lambda)$ by a site-specific mass absorption cross-section. For ZEP a MAC of 10.6m^2g^{-1} (Ohata et al., 2021) was used.

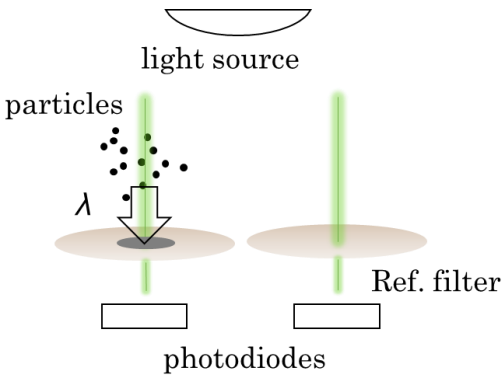

Fig 8: Aerosol particles are deposited on to a filter and the change in transmission, relative to a clean filter, is measured. Note that this is not an exact replication of how the MAAP nor PSAP works but rather a representation of the underlying concept.

4.2.2 Sampling cloud droplets: whole-air inlet and ground-based counterflow virtual impactor (GCVI)

To measure the concentration of cloud residuals, one must first be able to sample cloud droplets, and this is where the ground-based counterflow virtual impactor (GCVI; Brechtel Manufacturing Inc., USA, Model 1205) inlet comes into play. The GCVI works by sampling air and only allowing large particles above 6-7 microns (aerodynamic diameter) to pass the virtual stagnation plate, which separates particles based on this cut-off (Noone *et al.*, 1988; Shingler *et al.*, 2012). Particles greater than 6-7 microns are assumed to be cloud droplets

or ice crystals, and those smaller than this cut-off are assumed to interstitial (i.e., non-activated) aerosol particles. The GCVI is switched on to sample clouds, when the visibility sensor measures a visibility below 1 km for at least 5 minutes. The whole-air inlet as the same suggests samples all particles regardless of size.

For our purposes two identical MAAPs were connected to the inlets using a 3-way value (see Fig. 9). The valve allows us to measure light-absorbing particles that were either sampled through the GCVI or the whole-air inlet. All the concentrations that were measured were adjusted for loses and a sampling efficiency of 0.46 was applied when measuring via the GCVI (Karlsson et al., 2021).

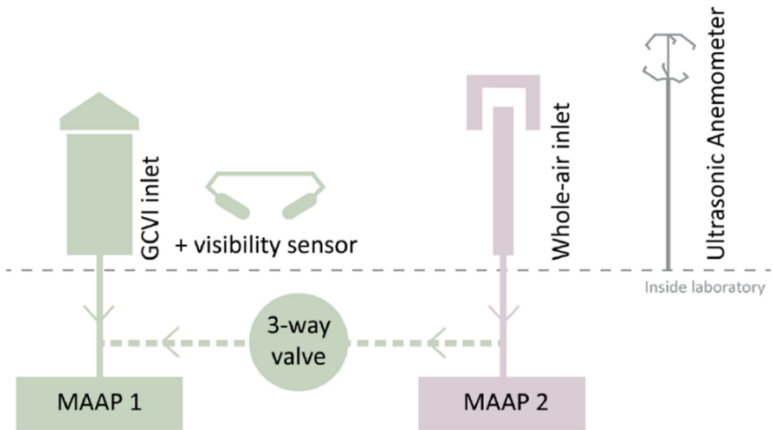

Fig 9: The setup for sampling eBC within cloud particles (residuals) and whole-air eBC (interstitial & cloud residuals). eBC is measured using two identical Multi Angle Absorption Photometers (MAAPs). The ultrasonic anemometer measures temperature and pressure, and the visibility which is used to infer cloudy or non-cloudy conditions is measured by the visibility sensor. Illustration taken from Zieger et al., 2023.

4.2.3 Particle and ion number and size distribution

Particle number size distributions (PNSDs) and ion number size distribution data were collected and utilised in **Papers II & IV**. Two sets of instrumentation were used throughout this thesis to measure the concentration of particles

within certain size ranges, namely the Differential Mobility Particle Sizer (DMPS) and the Neutral cluster and air ion spectrometer (NAIS).

4.2.3.1 Differential Mobility Particle Sizer (DMPS)

DMPS systems consist of a Differential Mobility Analyser (DMA) and a Condensation Particle Counter (CPC). Particles which enter the instrument are sorted by their size (electrical mobility), by first imparting an electrical charge on them, and then selecting them based on their electrical mobility which is related to their size and can be converted to an equivalent particle diameter. The concentration of particles, per size segregation, is then counted using a CPC after growing the particles using butanol vapours to optically-detectable sizes. The lowest cut off diameter (50% of particles pass though) for this DMPS setup was 5nm.

4.2.3.2 Neutral cluster and air ion spectrometer (NAIS)

The Neutral cluster and Air Ion Spectrometer (NAIS, developed by Airel Ltd., Estonia, see Fig. 10 a) for its appearance) is a mobility spectrometer, measuring both neutral particles and charged ions (Mirme and Mirme, 2013) that has been specifically designed to measure nanometre aerosols. Its ability to reliably and accurately measure down to 0.8nm for charged ions (both polarities) and 2.5nm for neutral particles (i.e. based electrical mobility) has made it a common instrument when researching the phenomenon of NPF. Along with that, its quick temporal resolution, ease of use and relatively low maintenance (e.g. not having to change butanol) are additional advantages over the more traditional DMPS instruments. The NAIS should be seen as serving to complement the DMPS measuring technique, as it is able to lower the limit for the detection of the smallest particles. However, the NAIS does not quite get down to measuring electrically neutral aerosol particles as small as 1-2nm, the point at which nucleation first begins, and it does not cover diameters greater than 40nm.

The design of the NAIS is based on an older instrument, the Air Ion Spectrometer (AIS), and like the AIS, the NAIS features two central multichannel electrical mobility analysers (see pink and blue main columns in Fig. 10 b)). The NAIS cycles through various modes in which it either measures all particles (including uncharged), and naturally charged particles (ions). When measuring particles, the ions generated by the charger must be removed (by the dischargers). The NAIS detects the particles which enter the two electrical mobility analysers, by detecting variabilities in the electrical field. It measures the whole mobility range of ions in both polarities and particles simultaneously. Particles are detected electrically and therefore we avoid the use of a CPC.

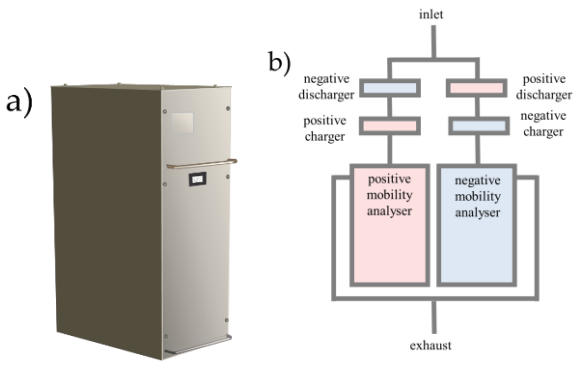

Fig 10: (a) model of the Neutral cluster and air ion spectrometer (NAIS) and (b) a simplified diagram of the main parts inside the NAIS.

4.2.4 Airmass history

'Essentially, all models are wrong, but some are useful'

George E. P. Box

Atmospheric dispersion models are used to help simulate the transport and dispersion of airborne pollutants. They constitute an invaluable asset in atmospheric science. One popular utilisation of dispersion models is to track the history of air parcels from source to receptor, both in terms of x,y,z coordinates, but also with respect to the prevailing meteorological conditions

during transport. Atmospheric dispersion models can be used in combination with surface properties, including pollution hotspots and land-use types to build upon the picture of the source-receptor relationships. The number of atmospheric dispersion models is numerous and wide-ranging of which the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler *et al.*, 1998; Stein *et al.*, 2015), the model used throughout this thesis work, is but one.

HYSPLIT may not be the most sophisticated, but it is user-friendly and fairly simple to run. Typically, air/fluid parcel models have either a Eulerian or a Lagrangian frame of reference, however, a key feature of HYSPLIT is that it blends these two different frames of reference. In the Eulerian view, the air parcel is viewed from a fixed point in space, as time passes, whereas in a Lagrangian perspective the observer follows the parcel through space and time.

Like most things, models are not infallible and exhibit certain limitations; many of these models, not just HYSPLIT, struggle when complex terrain topography is involved. They are also limited by the temporal and spatial resolution used in the meteorological fields. Furthermore, these models can find it difficult to actually estimate the height of the planetary boundary layer (PBL), i.e., mixing layer, an important parameter as it represents the lowest layer of the atmosphere, where turbulent wind dynamics are strong and the surface is able to influence the atmosphere.

Summary of papers

In this section, I want to take you through the various research papers that make up this thesis. The backdrop is that of a dramatically changing Arctic climate. The central focus of all 4 papers is the exploration of the lifecycle of Arctic aerosols, key players in the radiative budget. In this thesis we have looked at both anthropogenic and natural aerosols, namely light absorbing aerosols and DMS-derived newly formed particles. We have worked across a board range of diameters, from clusters of molecules barely 1nm in diameter to ~1μm, i.e. the accumulation mode. We have explored aspects of both long-range transported aerosols and particles produced in close proximity to the observatory. Lastly, we have investigated all these processes through the prism of various temporal scales, ranging from the short-term dynamics of an NPF day to seasonality and long-term trends.

This thesis comprises four main papers, **I-IV**. Each paper touches on certain aspects of the aerosol lifecycle. In **Paper I** we explored the emissions of light absorbing aerosol particles, their transport in the troposphere and one of the processes controlling the last stage of the aerosol lifecycle, i.e. removal by precipitation. In **Paper II,** we looked at how precipitation and solar radiation influence the concentration of different aerosol size modes. **Paper III** focused on how light-absorbing particles are activated into cloud droplets. Finally, **Paper IV,** explored new particle formation, the initial step in the aerosol life cycle. In each of the following subsections I state the motivating research questions, and then follow up with a brief summary of the main findings.

5.1 Paper I: *Increase in precipitation scavenging contributes to long-term reductions of light-absorbing aerosol in the Arctic.*

RQs relating to light-absorbing aerosols

RQ1a: How has the ambient concentration of light-absorbing particles changed over the course of the last two decades at ZEP?
RQ1b: How have different source regions changed in terms of their contribution to the light absorbing particles measured at ZEP?
RQ1c: What are the main factors which have influenced the overall trend?

RQ2a: Black carbon measured during the summer has been described as being dominated by biomass burning aerosols, is this a recent phenomenon?
RQ2b: Can biomass burning explain the recent stagnation in the long-term changes in black carbon, and do extreme events have the potential to influence long-term trends?

Paper I used the long-term observations at the Zeppelin Observatory to analyse the changes in the light-absorption coefficient (σ_{ap}) over the last couple of decades. It is important to note that σ_{ap} reflects the concentration of all light-absorbing particles, and not only black carbon. The basis of the paper was to understand the extent to which σ_{ap} was changing, and to delve into the possible factors governing the long-term observed trends in σ_{ap}.

We found that σ_{ap} at ZEP exhibited a statistically significant decreasing trend of -0.8% per year from 2002 to 2023, including decreasing trends across all three Arctic aerosol seasons (see Fig. 11). Moreover, each season showed an overall reduction in σ_{ap}, implying that the anthropogenic influence had reduced for all parts of the year, including the generally more polluted Arctic Haze season. This finding was in line with numerous other studies that described a reduction in the concentration of anthropogenic aerosol particles in the Arctic (Bodhaine and Dutton, 1993; Sharma *et al.*, 2013; Collaud Coen *et al.*, 2020; Schmale *et al.*, 2022). Special care was taken to ensure the long-term trends estimated in **Paper I** were statistically robust.

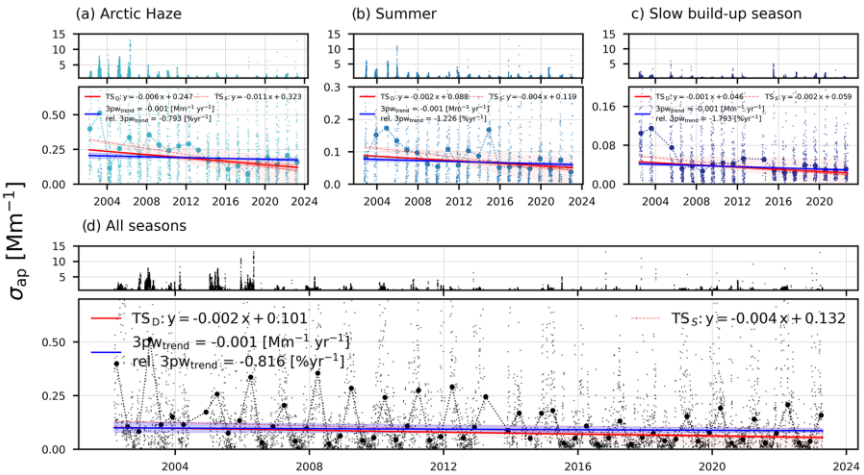

Fig. 11: Harmonised time series for the absorption coefficient (σ_{ap}) measured at Zeppelin Observatory for the (a) Arctic haze season (February to May), (b) summer (June to September), (c) slow-build-up season (October to January), and (d) the whole time series. The panels are split into two to display values greater than the maximum of the panel below. Both daily and seasonal medians are displayed in their respective colours; a colour represents each season, whilst black represents all seasons present. Trends for both daily (solid) and seasonal (dashed) medians are presented as red lines.

We were able to find the source regions that contributed most to σ_{ap} at ZEP throughout the last 22 years, using HYSPLIT back trajectories and the concentration weighted trajectory (CWT) approach (Hsu, Holsen and Hopke, 2003). The CWT approach allowed us to estimate the average σ_{ap} for air masses arriving from different locations. By weighting the concentrations by the residence times of air masses in the grid cells and then summing them up, we arrived at a CWT value for each respective grid cell. With the CWT mappings, we were able to identify some of the most significant extreme biomass burning events that had affected ZEP. These included the forest fires in the Yukon-Koyukuk region of Alaska, 10-17 July 2015 (Markowicz et al., 2016) and the Eastern Europe agricultural fire in May 2006 (Stohl *et al.*, 2007) (see Fig. 12).

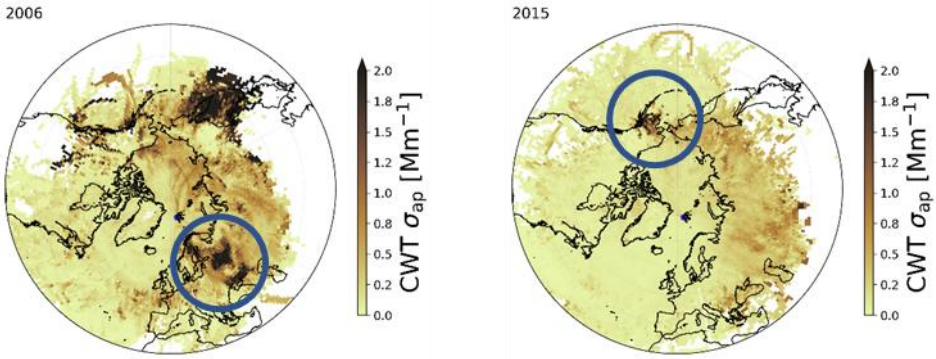

Fig. 12: Location of extreme biomass burning events: Concentration Weighted Trajectory (CWT) mappings for 2006 and 2015. CWT values for σ_{ap} calculated according to Hsu et al. (2003). Locations of emissions are circled.

By combing together 21 years of CWT mappings, we were able to analyse the spatial trend in regard to the contribution of σ_{ap} from various source regions (see Fig. 13). We were able to effectively show that the largest reductions in terms of contributions to σ_{ap} at ZEP could be found in Northern Siberia, and in particular the Yamal-Nenets gas flaring region in central Russia.

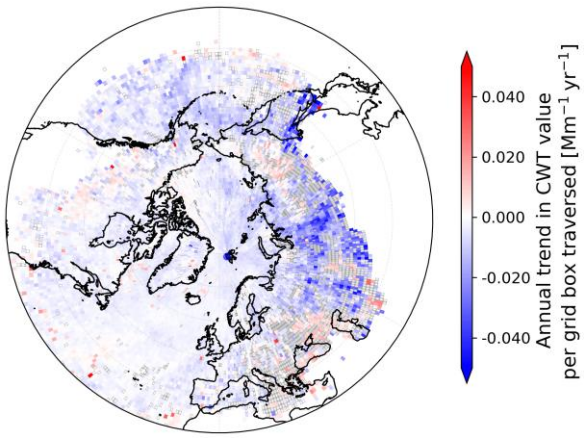

Fig. 13: Long-term trend in the annual CWT values for σ_{ap} for the years 2002–2022 (the last year is not included as it is not a full year). A threshold of 15 data points for each $1° \times 1°$ grid cell is applied to the mapping. All grid cells are statistically significant ($p<0.05$), unless the grid cell includes a grey.

After estimating the long-term trend in σ_{ap} at ZEP, temporally and spatially, we were particularly interested in finding out what controlled this trend and

to what degree did sources, sinks and transport play in the overall reduction of σ_{ap}.

It is widely credited that precipitation is one of the main controlling factors behind the seasonality of anthropogenic aerosol particles observed at ZEP (Garrett et al., 2011). Some model-based studies have speculated that a wetter and warmer Arctic could result in a cleaner Arctic (Jiao and Flanner, 2016), however, few trend studies have focused on the sinks of aerosols.

In **Paper I,** I explored the link between precipitation scavenging and the observed σ_{ap}. I used what we knew about the relationship between ambient σ_{ap} and precipitation amount to try and model the role played by sinks. I showed that the Arctic had witnessed a slight positive trend in precipitation, and by combining this with the relationship I obtained between precipitation and σ_{ap}, I was able to replicate the distinct and typical Arctic aerosol seasonality, with respect to σ_{ap}. Furthermore, I estimated that the increasing trend in precipitation could account for an estimated 25% of the overall reduction observed in σ_{ap}. The rest stems from reductions in emissions.

Paper I also commented on emissions from biomass burning events and their potential influence on σ_{ap}. I was able to show that extreme events from biomass burning had indeed increased in frequency; however, these events had not impacted the overall trend. Put simply, the way in which I calculated the long-term trends in σ_{ap} using pre-whitening techniques, along with seasonal and daily medians meant that these long-term trends were robust and desensitised to extreme events. However, I could see an impact from these extreme events if the arithmetic means for σ_{ap} were used.

5.2 Paper II *Sink, Source or Something In-Between? Net Effects of Precipitation on Aerosol Particle Populations.*

RQs relating to particle number size distributions

RQ3a: How is the particle number size distribution (PNSD) at ZEP influenced by precipitation en-route? How does the timing with respect to precipitation en-route influence the PNSD?

RQ3b: Is there any evidence to suggest that precipitation might be a direct source of nucleation mode particles at ZEP (i.e. when precipitation occurs on site)?

RQ4: How does the combined effect of aerosol wet removal (precipitation) and photochemistry (solar radiation) influence the concentration of particles of different sizes?

Paper II makes use of ten years of particle number size distribution data to explore the links between precipitation and aerosol number particle concentrations. It also places ZEP into a wider geographical context by comparing the measurements to different environments, i.e., the tropical and the boreal regions. The work in **Paper II** attempts to elucidate the role of precipitation, either as a source or sink, when it comes to the in situ-formed secondary aerosol particles in the nucleation and Aitken modes. It also attempts to examine and quantify the influence of both solar radiation and precipitation on the number concentrations of various aerosol modes.

Clouds and precipitation can increase the number concentration of small particles, either directly or indirectly. The indirect effect stems from precipitation removing the sink for ultrafine particles (i.e., CS). Precipitation effectively removes the larger accumulation mode particles, reducing the total surface area of pre-existing aerosol particles. This, in turn, increases the survivability of gaseous precursors required for nucleation, and the newly formed nucleation mode particles themselves. The direct production of ultrafine particles during or shortly after precipitation is related to particles formed in the vicinity of the clouds. The direct effect has been observed during targeted field campaigns, where newly formed particles were measured in the outflow of deep convective clouds, in tropical environments (Krejci *et al.*, 2003; Braga *et al.*, 2022). The indirect and direct effects relate to the two different production mechanisms connected to precipitation, and are the two mechanisms **Paper II** explores in detail.

The method adopted in **Paper II** explored the history of precipitation prior to arrival at ZEP, correlating precipitation rates with particle concentrations (see Figure 14). This analysis allowed us to reveal signals of both the direct and indirect production of ultrafine particles.

We showed clearly that the number concentration of particles with diameters greater than 70 nm correlated negatively with precipitation, regardless of when the rainfall occurred. For diameters less than 70 nm, the story is a little more complicated, showing both positive and negative correlations. We posit that this shift from a potential source to a dominating sink at 70 nm reflects the size of particles most prone to scavenging by precipitation. Below 70 nm, the effect of indirect and direct production mechanisms become relevant.

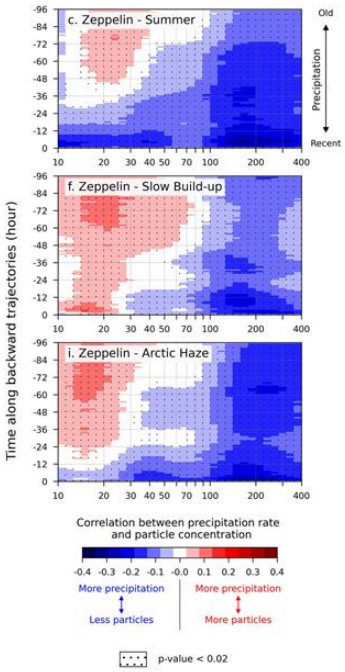

Fig. 14: Correlations between precipitation intensity and particle number size distribution at ZEP (c, f, i). The colour scale depicts the Spearman correlation coefficient between precipitation intensity at a given point of the 96 hr trajectories and the aerosol particle number size distribution. Stippling indicates statistical significance to the 98% level. The seasons represented are c. summer (Jun-Sep), f. slow-build up season (Oct-Jan) and i. Haze (Feb-May).

For ZEP, it was shown that for all seasons, at some point along the air mass history, there was a positive correlation between precipitation and the number concentration of particles smaller than 70 nm ($N_{<70nm}$) (see Fig. 14). The positive

correlations reflect the aforementioned indirect effect (i.e., the generation of NPF-favourable conditions due to reductions in CS and the coagulation sink). Interestingly, we observed that during the slow-build-up season (i.e. the transitional season), there was a positive correlation between $N_{10\text{-}20nm}$ and precipitation on-site at ZEP. This potentially signals that Aitken mode particles could be linked to recent precipitation events.

Paper II also tried to quantify and compare the influence of both precipitation and solar radiation on the total aerosol budget across various modes. For ZEP, it was found that solar radiation was by far a more dominant factor than precipitation when it came to influencing the number concentration of particles less than 50nm ($N_{<50nm}$). The solar radiation response on $N_{<50}$ seemed to stem from photochemically-induced NPF. Solar radiative flux is a good proxy for photochemical production potential, and it is widely acknowledged that photochemistry plays a pivotal role in the production of nucleating and condensing gases required for NPF. $N_{<50}$ responded weakly to changes in precipitation, which makes sense given the size of the particles (<50nm), many are not quite large enough to be wet scavenged. For particles greater than 100nm ($N_{>100nm}$), the responses to solar radiation and precipitation were essentially equal in magnitude, but displayed opposing signs. The precipitation response to $N_{>100nm}$ reflected the typical wet scavenging impact on aerosol populations. However, the positive response to solar radiation for $N_{>100nm}$ perhaps represents the continuous availability and condensation of vapours onto pre-existing larger particles during sunlight periods (an aspect we observed again in **Paper IV** through the growth of the condensation sink during NPF events, however, remains to be elucidated further).

5.3 Paper III: *Black carbon scavenging by low-level Arctic clouds.*

RQs relating to nucleation scavenging of black carbon aerosol particles

RQ5a: Can we describe the annual partitioning of BC between the interstitial particle phase (i.e. not activated) and the condensed phase (i.e. cloud droplets and ice crystals)?
RQ5b: How is the activation of BC impacted by environmental parameters e.g. cloud water content, total BC concentration, and temperature?

Paper III looked on 4 years' worth of observational eBC data from inside and outside of clouds. The unique instrumental setup at ZEP (as outlined in Sect. 4.2.2) was utilised to measure both directly activated and interstitial (inactivated) particles. The aim of **Paper III** was to determine how BC partitions between the interstitial particle phase and the condensed phase (i.e. cloud droplets) and to identify the environmental parameters that impact the proportion of BC activated into cloud droplets. **Paper III** focused on nucleation scavenging, the process by which aerosol particles are activated and become cloud droplets; we were unable to distinguish between nucleation and impaction scavenging; however, nucleation scavenging represents the predominant mechanism (Ohata *et al.*, 2016) and so it seemed reasonable to assume this was the main process we were exploring.

One of the major results from **Paper III** was the estimation of the scavenging fraction of eBC (F_{eBC}), which is the ratio of the concentration of cloud residuals to the concentration of eBC measured from the whole air inlet. The findings from **Paper III** showed a lower F_{eBC} during the Arctic Haze season, compared to a summer F_{eBC} that was close to unity. **Paper III** was able to show that the vast majority of eBC in the summer (March till October) is taken up and activated into cloud droplets.

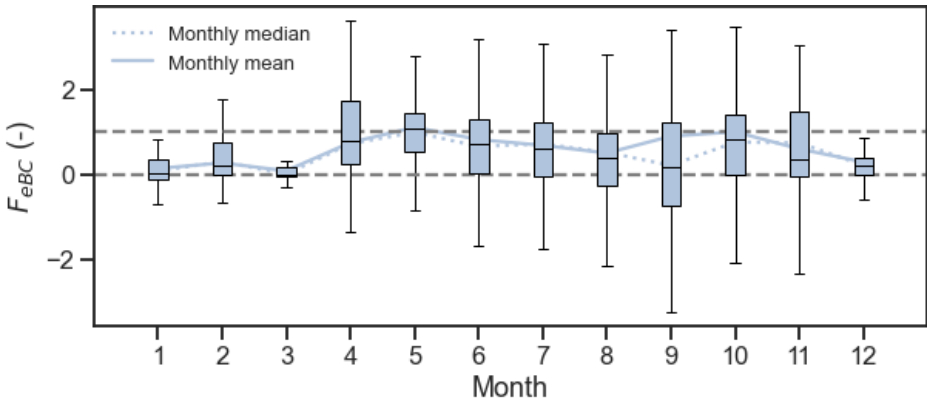

Fig. 15: Box plots of the monthly scavenged fraction of eBC (F_{eBC}). Note that F_{eBC} can exceed 1 and fall below 0 due to the high-resolution data often close to the detection limit. Extremely small eBC concentrations are used to calculate F_{eBC} and leading to large uncertainties.

The F_{eBC} at ZEP was higher compared to measurements performed at lower latitudes (e.g. $F_{scav,\ BC} \approx 0.61$ at Jungfraujoch by Cozic et al. (2007)). We speculated that the longer transport times to the Arctic, led to more aged and hygroscopic aerosols, making it easier for eBC to be effectively activated into cloud droplets, and thus resulting in a higher F_{eBC} value.

In addition to the seasonality of FeBC, **Paper III** looked at the relationships between environmental parameters and FeBC. We found that FeBC was clearly dependent on the cloud water content (CWC) such that clouds with higher CWC tended to scavenge more eBC. When the CWC was low, scavenging was limited by the availability of condensable water vapour. Another interesting finding was the dependency FeBC had with temperature. There appeared to be a shift in the calculated FeBC from close to zero at temperatures below -5°C to close to unity at temperatures greater than 5°C. We argued in **Paper III** that the reduction in FeBC at colder temperatures could be explained by the presence of ice in mixed-phased clouds. Ice crystals can grow at the expense of liquid cloud droplets due to the Wegener-Bergeron-Findeisen (WBF) process. The growth of ice crystals can therefore lead to the evaporation of liquid droplets, leaving BC in the interstitial phase, and lowering FeBC. Lastly, we found that the source regions for eBC cloud residuals tended to originate more from marine regions compared to eBC measured during non-cloudy periods.

We speculated that a possible source of eBC cloud residuals was related to emissions from shipping or flaring activities in the Norwegian Sea.

5.4 Paper IV: *Drivers governing the seasonality of new particle formation in the Arctic*

RQs relating to new particle formation

RQ6a: How frequent and how strong are NPF events over the course of a year, as measured from ZEP
RQ6b: What are the processes and parameters that can explain the NPF occurrence and strength?

RQ7: To what degree does NPF influence the CCN budget?

The focus of **Paper IV** was on new particle formation (NPF) observed at ZEP. For **Paper IV**, I investigated, from the perspective of sinks and sources and with the aid of a Neutral cluster and Air and Ion spectrometer (NAIS), the controlling factors determining nucleation and the subsequent growth of newly formed particles

I showed that NPF has a very distinct annual cycle. The NPF begins in April and continues throughout the summer months, with the final events occurring as late as November. I speculated that the NPF events occurring in late October and November, during the polar night when the Sun no longer crossed the horizon at ZEP, were linked to advected air masses from the south that had descended from high altitudes, where they were exposed to solar radiation. Furthermore, we detailed the likely geographic origins of air masses associated with nucleation observed at ZEP. I showed that NPF events were considerably more likely to originate from the marine regions off the western edge of Svalbard, particularly the Greenland Sea.

In **Paper IV**, we showed that using only two main environmental parameters—recently experienced solar insolation and the total surface area of pre-existing aerosol particles (i.e. condensation sink, CS)—I was able to predict the frequency of NPF event days, fairly robustly (see Fig. 16).

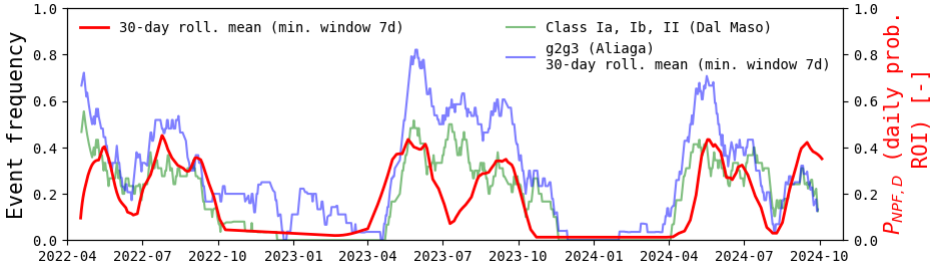

Fig. 16: The red line is the estimated daily likelihood of a measurements occurring in the region of interest (ROI). The ROI is based on the relation between CS and 6-hour solar accumulation defined in **Paper VI**. The green and blue lines represent the frequency of NPF events using the Dal Maso et al. (2005) or Aliaga et al. (2023) classification.

Final remarks and outlook

In this section, we will briefly outline the main successes of this thesis in relation to the research questions, try to connect them to the current state of Arctic climate and aerosol science, and consider whether it is possible to go full circle and speculate on how this thesis relates to the original motivation surrounding Arctic Amplification. We argue that the papers in this thesis were successful in shedding new light on certain Arctic aerosol knowledge gaps. We were able to tackle the list of research questions we posed; however, there were of course shortcomings, and I will touch upon what could have been done differently or improved. Moreover, the research that has emerged from this thesis has also attracted additional points of discussion, and thus I will discuss emerging research foci—both in general and as a direct consequence of the findings.

Throughout the studies presented in this thesis, we were able to use the setup of one of the best-equipped atmospheric research stations in the Arctic. I had access to unique data sets comprising of one of the longest sets of measurements in the Arctic of light-absorbing aerosol concentrations just to mention an example. Long-term time series data (i.e. decades) enables both advanced process studies as well as trend analysis. In addition, I was able to collaborate with colleagues and fellow researchers from a broad range of other institutes to further probe the originally posed research questions. Utilisation of long-term observations represents a different way of doing scientific research compared to more targeted experimental approaches or campaign-driven research agendas, as for a large part of this work, the data came first before a research plan was created; in other words, the research questions were not devised and the objectives of what we wanted to answer were not asked, before beginning the measurements. This serves to reflect the different ways science and research can be performed. Using data from long-term monitoring and accessing large data sets enables researchers to ask questions that simply cannot be considered when conducting more campaign-oriented studies. The changes taking place in the Arctic are on a climatic scale and only by analysing

data also on that time scale can we begin to understand the changes taking place.

In regards to **Paper I**, the main success was being able to use the long-term data sets at ZEP to show that the concentration of light-absorbing aerosol particles had declined over the past two decades (RQ1). We argued that approximately one quarter of this decline is likely due to an increase in precipitation scavenging, and that the rest arose from changes in emissions (RQ1). One discussion point that arises from this finding is the potential impact changes in precipitation may have on the overall radiative forcing in the Arctic. It could be speculated that reducing the concentration of ambient light-absorbing particles in the Arctic would bring about a reduction in positive forcing from direct aerosol effects (i.e. ARI), thereby reducing the amount of near-surface warming. However, overall, increases in wet scavenging are likely linked also to increased cloudiness and more water vapor; both of which will likely have larger effects on the radiative balance than just changes in BC. **Paper I** also hints at the possibility that deposition of BC may have increased, due to enhanced wet scavenging, which, considering the impact BC deposition has on sea ice and snow, could, in turn, produce a positive radiative forcing due to reductions in surface albedo. The key factor here would be to understand at which point along the route to the Arctic does deposition occur most readily – are the majority of anthropogenic aerosol particles removed before air masses reach the Arctic, or does the increased precipitation lead to a larger portion of light absorbing particles being deposited on Arctic surfaces? The effects of both transport and deposition in the Arctic are complex and hard to assess, and in order to evaluate the net impact, we would require both long term observations and the use of global climate models (GCMs). The latter is not utilised in this thesis.

In addition to this, if we broaden the scope to consider all anthropogenic particles, both warming and cooling agents, and assume fairly that they are all scavenged, and not just BC, then what could be the overall impact on the radiative forcing? At lower latitudes, perhaps, we could expect the increased scavenging and rainout of particles to reduce the reflectivity of the planet, as aerosol particles typically act as a cooling component of the climate, scattering

solar radiation back to space. In the Arctic, the increased scavenging and reduction in ambient aerosol concentrations could enhance AA, like what was modelled by Navarro *et al.* (2016). The overall effect on the radiative forcing becomes more complicated when we involve aerosol-cloud interactions. We know thanks to **Paper III** that BC is effectively scavenged and that BC particles are able to act as CCN. In **Paper I**, I only infer this from measurements collocated with back trajectories; however, in **Paper III** we measure this directly. Understanding the effect, the long-term reduction in the concentration of BC particles could have on cloud properties and thus Arctic climate is challenging to answer and beyond the scope of this work, but one I will touch on again when discussing the results from **Paper III** below. It is however important to note that substantial changes have been taking place during the last decades, and that it is reasonable to assume that this will have had an impact on the Arctic radiative budget and, in extension, any future climate projections.

Paper II showed that clouds, and more specifically precipitation, can be a process facilitating both the production and removal of particles. **Paper II** succeeded in demonstrating the effectiveness of precipitation in removing larger accumulation mode particles (>70nm) (RQ3). It also showed the key differences between the Aitken and the accumulation modes when it comes to precipitation. For all particles in the accumulation mode, precipitation acted as a sink, and there was a negative correlation between precipitation and particle number concentrations for all diameters greater than 70nm. The Aitken mode, however, showed a mixed response (RQ3); we could see the signal behind the indirect production of nucleation mode particles i.e. the reduction of the condensation sink, helping to provide the conditions for NPF (RQ6, and a process which is discussed in more detail in **Paper IV**). **Paper II** demonstrated the importance of investigating how different processes influence different aerosol modes. It would be interesting to further research the signal that we observed, pertaining to the direct production of nucleation mode particles from precipitation (RQ4).

The main finding from **Paper III** was to demonstrate, through in-situ measurements, that light absorbing particles (i.e. eBC) could be activated into

cloud droplets. **Paper III** is the first time a fairly long-term observational data set, measuring actual cloud residuals, had been utilised to estimate the F_{eBC} in the Arctic. We were also able to show that F_{eBC} varied depending on the month and other environmental factors (RQ5). It could be argued that the findings from **Paper III** are applicable to other types of aerosol particles of a similar size because the physical mechanisms of aerosol scavenging are highly dependent on size, and less on the chemical composition. **Paper III** shows nicely that during the months with low accumulation mode concentrations, F_{eBC} is large, and the Arctic is potentially CCN-limited and therefore susceptible to changes in CCN concentrations (RQ5). Clean regions like the Arctic are where aerosol-mediated effects on cloud radiative properties, like the Twomey effect, will be most pronounced. If we combine together the findings from **Papers I and III,** we can speculate about how continued reductions in anthropogenic aerosols could impact clouds; in other words what influence will the decreasing trends in eBC for example have on the availability of aerosol particles to be scavenged? We know from **Paper III** that the accumulation mode particles are preferentially scavenged (RQ5), so perhaps a cleaner Arctic could in fact make the region even more CCN-limited, and thus even more efficient in removing the remainder of the particles. This is but pure speculation, but nonetheless, it would be interesting to investigate a future, more pristine, Arctic, resembling more pre-industrial conditions.

Now, with regard to NPF and the influence it has on Arctic aerosols, I have been able to show in **Paper IV** that NPF indeed influences the overall Aitken mode budget and can increase the concentration of particles with the potential to act as CCN (RQ7). Importantly, this production occurs during periods where the atmosphere is particularly clean and cloud formation is potentially limited by CCN availability. CCN during the periods when we have NPF will most likely also come from the Aitken mode, and not just the accumulation mode, providing the possibility that NPF events will perturb the overall CCN budget. Changes in the CCN-budget may lead to alterations in the cloud microphysics that may have considerable effects on the Arctic climate given the importance of Arctic clouds to the radiative budget (as laid out in Sect 2.3). It would be interesting to investigate further the seemingly self-regulating

qualities behind the repopulation of Aitken mode particles during the Arctic NPF season.

One aspect of **Paper IV** that also applies to **Paper I,** which could be improved upon, is the somewhat circular nature of the simplified models that were developed. It is to some extent analogous to models solely run on training data sets; it is important to test how these crude models fare with new measurements.

Papers I-IV all touch on the notion that the ratio of anthropogenic to biogenic aerosols could change over time in the Arctic. Mitigation policies to improve air quality at lower latitudes, transition to a wetter and warmer Arctic as well as other changes in the Arctic due to climate change are all factors which may impact this ratio between anthropogenic and biogenic aerosols. I observed a reduction in anthropogenic aerosol particles across all seasons in **Paper I**, and saw that this reduction is dependent not only on sources, but sinks as well. I also saw, in **Paper IV**, that an important source of secondary biogenic aerosols is controlled by solar radiation and the total surface area of pre-existing aerosols (RQ6), meaning that the formation of secondary biogenic aerosols could potentially increase depending on these factors. The two trends are interconnected; a cleaner Arctic could encourage more nucleation for example. Moreover, changes in marine productivity, cloud coverage, and retreating sea ice will also have an influence on the concentration of biogenic aerosol particles. Continued sea ice retreat exposes more surface water to the atmosphere, and hence this could lead to increased emissions of biogenic gaseous precursors and also primary marine aerosol particles.

The focus of this thesis is on improving our knowledge of aerosol sources and sinks in the rapidly changing Arctic environment, a geographical area that is critical to the Earth's climate. This thesis helps to explore and analyse data from a part of the globe that has historically remained relatively under-explored and under-sampled. One interesting motivation to study pristine regions, but is out of the scope of this thesis, is to try to approximate preindustrial conditions, important when addressing questions around the total radiative forcing induced by anthropogenic aerosols (Carslaw *et al.*, 2013). Are we moving towards conditions more similar to the natural background in terms of

atmospheric trace constituents, and what will be the ultimate consequence of this?

One major limitation of this work though is that of scale; we only explore measurements from essentially one point. Hence, it is natural to ask the question of how representative Zeppelin Observatory is and is it possible to extrapolate our findings to the wider Arctic. A considerable amount of work has been done on trying to bring together the data from all the various Arctic measurement sites to see what the similarities and differences are (e.g. Freud et al., 2017). The work in this thesis should therefore be viewed as a part of this broader pan-Arctic research. There are also potential limitations specific to Zeppelin Observatory; given the local topography and features such as glacier-driven katabatic winds, there is a chance that certain aspects cannot be generalised due to the uniqueness inherit to the site. Only by using GCMs can we really begin to generalise over large pan-Arctic geographical regions. Through GCMs we can test some of the hypothesises and research questions laid out in this thesis on a wider scale.

Nonetheless, in-situ measurements remain a vital component of the overall research infrastructure. There are important and numerous advantages that in-situ measurements pose over other types of data. For example, in-situ measurements allow for aerosol and cloud properties to be measured simultaneously, as opposed to satellites, which require clear sky conditions to measure aerosol particles. Furthermore, direct, in-situ measurements are necessary because it is unlikely that accurate information about particle microphysical and chemical properties can be gained from remote sensing measurements alone. Data is also of vital importance when assessing how the models perform, and thus crucial for evaluation of process descriptions used in these models. Measurements and observations like those presented in this thesis can potentially help evaluate aerosol-cloud interactions in GCMs.

In all, the findings and measurements presented in this thesis serve as an important complement to the research done by the various other researchers at different institutes, using different techniques and methods, aiding the overarching goal of accurately describing the complex Arctic climate system. There is a great deal of good science that has been done and continues to be

done, and connecting it all together holistically to understand and frame the big picture remains a challenge.

References

Books:

Carslaw, K. S., Aerosols and Climate Elsevier, 2022, ISBN 9780128197660, https://doi.org/10.1016/B978-0-12-819766-0.09996-X

Pruppacher, H.R. and J. D. Klett, Microphysics of clouds and precipitation pp. 714, D. Reidel, Hingham, Mass., 1978.

Articles:

Aas, W. *et al.* (2024) 'Trends in Air Pollution in Europe, 2000–2019', *Aerosol and Air Quality Research*, 24(4). Available at: https://doi.org/10.4209/aaqr.230237.

Albrecht, B.A. (1989) 'Aerosols, Cloud Microphysics, and Fractional Cloudiness', *Science*, 245(4923), pp. 1227–1230. Available at: https://doi.org/10.1126/science.245.4923.122.

Andela, N. *et al.* (2017) 'A human-driven decline in global burned area', *Science*, 356(6345). Available at: https://doi.org/10.1126/science.aal4108.

Arctic Monitoring and Assessment Programme (AMAP) (2015) *AMAP Assessment 2015: Black carbon and ozone as Arctic climate forcers. Arctic Monitoring and Assessment Programme (AMAP)*. Olso, Norway. Available at: www.amap.no.

Arctic Monitoring and Assessment Programme (AMAP). (2021) *Arctic climate change update 2021: Key trends and impacts. Summary for policy-makers.*

Arrhenius, S. (1896) 'XXXI. *On the influence of carbonic acid in the air upon the temperature of the ground*', *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science*, 41(251), pp. 237–276. Available at: https://doi.org/10.1080/14786449608620846.

Bodhaine, B.A. and Dutton, E.G. (1993) 'A long-term decrease in Arctic haze at Barrow, Alaska', *Geophysical Research Letters*, 20(10), pp. 947–950. Available at: https://doi.org/10.1029/93GL01146.

Bond, T.C. (2010) *Testimony for 'Clearing the Smoke: Black Carbon Pollution' House Committee on Energy Independence and Global Warming*.

Bozem, H. *et al.* (2019) 'Characterization of transport regimes and the polar dome during Arctic spring and summer using in situ aircraft measurements', *Atmospheric Chemistry and Physics*, 19(23), pp. 15049–15071. Available at: https://doi.org/10.5194/acp-19-15049-2019.

Braga, R.C. *et al.* (2022) 'Detrainment Dominates CCN Concentrations Around Non-Precipitating Convective Clouds Over the Amazon', *Geophysical Research Letters*, 49(20). Available at: https://doi.org/10.1029/2022GL100411.

Carslaw, K.S. *et al.* (2013) 'Large contribution of natural aerosols to uncertainty in indirect forcing', *Nature*, 503(7474), pp. 67–71. Available at: https://doi.org/10.1038/nature12674.

Chen, Y. *et al.* (2022) 'Machine learning reveals climate forcing from aerosols is dominated by increased cloud cover', *Nature Geoscience*, 15(8), pp. 609–614. Available at: https://doi.org/10.1038/s41561-022-00991-6.

Chen, Y. *et al.* (2024) 'Substantial cooling effect from aerosol-induced increase in tropical marine cloud cover', *Nature Geoscience*, 17(5), pp. 404–410. Available at: https://doi.org/10.1038/s41561-024-01427-z.

Chylek, P. *et al.* (2022) 'Annual Mean Arctic Amplification 1970–2020: Observed and Simulated by CMIP6 Climate Models', *Geophysical Research Letters*, 49(13). Available at: https://doi.org/10.1029/2022GL099371.

Collaud Coen, M. *et al.* (2020) 'Multidecadal trend analysis of in situ aerosol radiative properties around the world', *Atmospheric Chemistry and Physics*, 20(14), pp. 8867–8908. Available at: https://doi.org/10.5194/acp-20-8867-2020.

Coumou, D. *et al.* (2018) 'The influence of Arctic amplification on mid-latitude summer circulation', *Nature Communications*. Nature Publishing Group. Available at: https://doi.org/10.1038/s41467-018-05256-8.

Cozic, J. *et al.* (2007) *Atmospheric Chemistry and Physics Scavenging of black carbon in mixed phase clouds at the high alpine site Jungfraujoch*, *Atmos. Chem. Phys*. Available at: www.atmos-chem-phys.net/7/1797/2007/.

Curry, J.A. *et al.* (1996) 'clim-1520-0442_1996_009_1731_ooacar_2_0_co_2', *Journal of Climate*, 9(8), pp. 1731–1764. Available at:

https://doi.org/https://doi.org/10.1175/1520-0442(1996)009<1731:OOACAR>2.0.CO;2.

Curtius, J., Lovejoy, E.R. and Froyd, K.D. (2006) 'Atmospheric ion-induced aerosol nucleation', *Space Science Reviews*, 125(1–4), pp. 159–167. Available at: https://doi.org/10.1007/s11214-006-9054-5.

Dallósto, M. *et al.* (2017) 'Arctic sea ice melt leads to atmospheric new particle formation', *Scientific Reports*, 7(1). Available at: https://doi.org/10.1038/s41598-017-03328-1.

Ding, S. *et al.* (2024) 'A Review on the Arctic–Midlatitudes Connection: Interactive Impacts, Physical Mechanisms, and Nonstationary', *Atmosphere*. Multidisciplinary Digital Publishing Institute (MDPI). Available at: https://doi.org/10.3390/atmos15091115.

Draxler, R.R. *et al.* (1998) *An Overview of the HYSPLIT_4 Modelling System for Trajectories, Dispersion, and Deposition*, *Australian Meteorological Magazine*.

Ebell, K. *et al.* (2020) 'Radiative effect of clouds at ny-Ålesund, svalbard, as inferred from ground-based remote sensing observations', *Journal of Applied Meteorology and Climatology*, 59(1), pp. 3–22. Available at: https://doi.org/10.1175/JAMC-D-19-0080.1.

Flanner, M.G. (2013) 'Arctic climate sensitivity to local black carbon', *Journal of Geophysical Research Atmospheres*, 118(4), pp. 1840–1851. Available at: https://doi.org/10.1002/jgrd.50176.

Francis, J.A. and Vavrus, S.J. (2012) 'Evidence linking Arctic amplification to extreme weather in mid-latitudes', *Geophysical Research Letters*, 39(6). Available at: https://doi.org/10.1029/2012GL051000.

Freud, E. *et al.* (2017) 'Pan-Arctic aerosol number size distributions: Seasonality and transport patterns', *Atmospheric Chemistry and Physics*, 17(13), pp. 8101–8128. Available at: https://doi.org/10.5194/acp-17-8101-2017.

Garrett, T.J. *et al.* (2011a) 'The role of scavenging in the seasonal transport of black carbon and sulfate to the Arctic', *Geophysical Research Letters*, 38(16). Available at: https://doi.org/10.1029/2011GL048221.

Garrett, T.J. *et al.* (2011b) 'The role of scavenging in the seasonal transport of black carbon and sulfate to the Arctic', *Geophysical Research Letters*, 38(16). Available at: https://doi.org/10.1029/2011GL048221.

Gierens, R. *et al.* (2020) 'Low-level mixed-phase clouds in a complex Arctic environment', *Atmospheric Chemistry and Physics*, 20(6), pp. 3459–3481. Available at: https://doi.org/10.5194/acp-20-3459-2020.

Hales, J.M. (1978) 'Wet removal of sulfur compounds from the atmosphere', 12(1–3), pp. 389–399. Available at: https://doi.org/10.1016/0004-6981(78)90222-6.

Henry, M. and Vallis, G.K. (2021) 'Reduced High-Latitude Land Seasonality in Climates with Very High Carbon Dioxide', *Journal of Climate*, pp. 1–38. Available at: https://doi.org/10.1175/jcli-d-21-0131.1.

Hoppel, W.A., Frick, G.M. and Larson, R.E. (1986) *EFFECT OF NONPRECIPITATING CLOUDS ON THE AEROSOL SIZE DISTRIBUTION IN THE MARINE BOUNDARY LAYER*.

Hsu, Y.-K., Holsen, T.M. and Hopke, P.K. (2003) *Comparison of hybrid receptor models to locate PCB sources in Chicago*, *Atmospheric Environment*.

Intergovernmental Panel on Climate Change (IPCC) (2023) 'The Earth's Energy Budget, Climate Feedbacks and Climate Sensitivity', in *Climate Change 2021 – The Physical Science Basis*. Cambridge University Press, pp. 923–1054. Available at: https://doi.org/10.1017/9781009157896.009.

Jacobson, M.Z. *et al.* (2012) 'The effects of rerouting aircraft around the arctic circle on arctic and global climate', *Climatic Change*, 115(3–4), pp. 709–724. Available at: https://doi.org/10.1007/s10584-012-0462-0.

Jiao, C. and Flanner, M.G. (2016) 'Changing black carbon transport to the Arctic from present day to the end of 21st century', *Journal of Geophysical Research*, 121(9), pp. 4734–4750. Available at: https://doi.org/10.1002/2015JD023964.

Karlsson, L. *et al.* (2021) 'A long-term study of cloud residuals from low-level Arctic clouds', *Atmospheric Chemistry and Physics*, 21(11), pp. 8933–8959. Available at: https://doi.org/10.5194/acp-21-8933-2021.

Kay, J.E. *et al.* (2016) 'Recent Advances in Arctic Cloud and Climate Research', *Current Climate Change Reports*. Springer, pp. 159–169. Available at: https://doi.org/10.1007/s40641-016-0051-9.

Kim, Y.H. *et al.* (2023) 'Observationally-constrained projections of an ice-free Arctic even under a low emission scenario', *Nature Communications*, 14(1). Available at: https://doi.org/10.1038/s41467-023-38511-8.

Krejci, R. *et al.* (2003) 'Evolution of aerosol properties over the rain forest in Surinam, South America, observed from aircraft during the LBA-CLAIRE 98 experiment', *Journal of Geophysical Research: Atmospheres*, 108(18). Available at: https://doi.org/10.1029/2001jd001375.

Kulmala, M. *et al.* (2004) 'Organic aerosol formation via sulphate cluster activation', *Journal of Geophysical Research: Atmospheres*, 109(4). Available at: https://doi.org/10.1029/2003jd003961.

Lelieveld, J. *et al.* (2015) 'The contribution of outdoor air pollution sources to premature mortality on a global scale', *Nature*, 525(7569), pp. 367–371. Available at: https://doi.org/10.1038/nature15371.

Maturilli, M., Herber, A. and König-Langlo, G. (2015) 'Surface radiation climatology for Ny-Ålesund, Svalbard (78.9° N), basic observations for trend detection', *Theoretical and Applied Climatology*, (120), pp. 331–339. Available at: https://doi.org/https://doi.org/10.1007/s00704-014-1173-4.

Maturilli, M. and Kayser, M. (2016) 'Arctic warming, moisture increase and circulation changes observed in the Ny-Ålesund homogenized radiosonde record', *Theor Appl Climatol* [Preprint]. Available at: https://doi.org/10.1007/s00704-016-1864-0.

Meredith, M. *et al.* (2019) *IPCC special report on the ocean and cryosphere in a changing climate*.

Mewes, D. and Jacobi, C. (2019) 'Heat transport pathways into the Arctic and their connections to surface air temperatures', *Atmospheric Chemistry and Physics*, 19(6), pp. 3927–3937. Available at: https://doi.org/10.5194/acp-19-3927-2019.

Mioche, G. *et al.* (2015) 'Variability of mixed-phase clouds in the Arctic with a focus on the Svalbard region: A study based on spaceborne active remote sensing', *Atmospheric Chemistry and Physics*, 15(5), pp. 2445–2461. Available at: https://doi.org/10.5194/acp-15-2445-2015.

Mirme, S. and Mirme, A. (2013) 'The mathematical principles and design of the NAIS - A spectrometer for the measurement of cluster ion and nanometer aerosol size distributions', *Atmospheric Measurement Techniques*, 6(4), pp. 1061–1071. Available at: https://doi.org/10.5194/amt-6-1061-2013.

Navarro, J.C.A. *et al.* (2016) 'Amplification of Arctic warming by past air pollution reductions in Europe', *Nature Geoscience*, 9(4), pp. 277–281. Available at: https://doi.org/10.1038/ngeo2673.

Noone, K.J. *et al.* (1988) 'Design and calibration of a counterflow virtual impactor for sampling of atmospheric fog and cloud droplets', *Aerosol Science and Technology*, 8(3), pp. 235–244. Available at: https://doi.org/10.1080/02786828808959186.

Nordenskiöld, A.E. (1874) 'On the cosmic dust which falls on the surface of the earth with the atmospheric precipitation', *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science*, 48(321), pp. 546–547. Available at: https://doi.org/10.1080/14786447408641166.

Ohata, S. *et al.* (2016) 'A key process controlling the wet removal of aerosols: New observational evidence', *Scientific Reports*, 6. Available at: https://doi.org/10.1038/srep34113.

Ohata, S. *et al.* (2021) 'Estimates of mass absorption cross sections of black carbon for filter-based absorption photometers in the Arctic', *Atmospheric Measurement Techniques*, 14(10), pp. 6723–6748. Available at: https://doi.org/10.5194/amt-14-6723-2021.

Petzold, A. and Schönlinner, M. (2004) 'Multi-angle absorption photometry - A new method for the measurement of aerosol light absorption and atmospheric black carbon', *Journal of Aerosol Science*, 35(4), pp. 421–441. Available at: https://doi.org/10.1016/j.jaerosci.2003.09.005.

Pistone, K., Eisenman, I. and Ramanathan, V. (2019) 'Radiative Heating of an Ice-Free Arctic Ocean', *Geophysical Research Letters*, 46(13), pp. 7474–7480. Available at: https://doi.org/10.1029/2019GL082914.

Platt, S.M. *et al.* (2022) 'Atmospheric composition in the European Arctic and 30 years of the Zeppelin Observatory, Ny-Ålesund', *Atmospheric Chemistry and Physics*, 22(5), pp. 3321–3369. Available at: https://doi.org/10.5194/acp-22-3321-2022.

Rantanen, M. *et al.* (2022) 'The Arctic has warmed nearly four times faster than the globe since 1979', *Communications Earth and Environment*, 3(1). Available at: https://doi.org/10.1038/s43247-022-00498-3.

Ren, L. *et al.* (2020) 'Source attribution of Arctic black carbon and sulfate aerosols and associated Arctic surface warming during 1980-2018', *Atmospheric Chemistry and Physics*, 20(14), pp. 9067–9085. Available at: https://doi.org/10.5194/acp-20-9067-2020.

Rogers, B.M. *et al.* (2020) 'Focus on changing fire regimes: interactions with climate, ecosystems, and society', *Environmental Research Letters*. Institute of Physics Publishing. Available at: https://doi.org/10.1088/1748-9326/ab6d3a.

Sand, M. *et al.* (2013) 'The arctic response to remote and local forcing of black carbon', *Atmospheric Chemistry and Physics*, 13(1), pp. 211–224. Available at: https://doi.org/10.5194/acp-13-211-2013.

Schmale, J. *et al.* (2022) 'Pan-Arctic seasonal cycles and long-term trends of aerosol properties from 10 observatories', *Atmospheric Chemistry and Physics*, 22(5), pp. 3067–3096. Available at: https://doi.org/10.5194/acp-22-3067-2022.

Sedlar, J. *et al.* (2011) 'A transitioning Arctic surface energy budget: The impacts of solar zenith angle, surface albedo and cloud radiative forcing',

Climate Dynamics, 37(7–8), pp. 1643–1660. Available at: https://doi.org/10.1007/s00382-010-0937-5.

Sharma, S. *et al.* (2013) '16-year simulation of arctic black carbon: Transport, source contribution, and sensitivity analysis on deposition', *Journal of Geophysical Research Atmospheres*, 118(2), pp. 943–964. Available at: https://doi.org/10.1029/2012JD017774.

Sharma, S. *et al.* (2019) 'A Factor and Trends Analysis of Multidecadal Lower Tropospheric Observations of Arctic Aerosol Composition, Black Carbon, Ozone, and Mercury at Alert, Canada', *Journal of Geophysical Research: Atmospheres*, 124(24), pp. 14133–14161. Available at: https://doi.org/10.1029/2019JD030844.

Shaw, G.E. (1995) *The Arctic Haze Phenomenon*. Available at: https://doi.org/https://doi.org/10.1175/1520-0477(1995)076<2403:TAHP>2.0.CO;2.

Shindell, D. *et al.* (2008) *Atmospheric Chemistry and Physics Climate forcing and air quality change due to regional emissions reductions by economic sector*, *Atmos. Chem. Phys*. Available at: www.atmos-chem-phys.net/8/7101/2008/.

Shindell, D. and Faluvegi, G. (2009) 'Climate response to regional radiative forcing during the twentieth century', *Nature Geoscience*, 2(4), pp. 294–300. Available at: https://doi.org/10.1038/ngeo473.

Shingler, T. *et al.* (2012) 'Characterisation and airborne deployment of a new counterflow virtual impactor inlet', *Atmospheric Measurement Techniques*, 5(6), pp. 1259–1269. Available at: https://doi.org/10.5194/amt-5-1259-2012.

Shupe, M.D. and Intrieri, J.M. (2004) 'Cloud Radiative Forcing of the Arctic Surface: The Influence of Cloud Properties, Surface Albedo, and Solar Zenith Angle', *Journal of Climate*, 17(3).

Sledd, A. and L'Ecuyer, T. (2019) 'How much do clouds mask the impacts of arctic sea ice and snow cover variations? Different perspectives from observations and reanalyses', *Atmosphere*, 10(1). Available at: https://doi.org/10.3390/atmos10010012.

Sledd, A. and L'Ecuyer, T.S. (2021) 'Emerging Trends in Arctic Solar Absorption', *Geophysical Research Letters*, 48(24). Available at: https://doi.org/10.1029/2021GL095813.

Stein, A.F. *et al.* (2015) 'Noaa's hysplit atmospheric transport and dispersion modeling system', *Bulletin of the American Meteorological Society*. American Meteorological Society, pp. 2059–2077. Available at: https://doi.org/10.1175/BAMS-D-14-00110.1.

Stjern, C.W. *et al.* (2019) 'Arctic Amplification Response to Individual Climate Drivers', *Journal of Geophysical Research: Atmospheres*, 124(13), pp. 6698–6717. Available at: https://doi.org/10.1029/2018JD029726.

Stohl, A. (2006) 'Characteristics of atmospheric transport into the Arctic troposphere', *Journal of Geophysical Research Atmospheres*, 111(11). Available at: https://doi.org/10.1029/2005JD006888.

Stohl, A. *et al.* (2007) *Arctic smoke-record high air pollution levels in the European Arctic due to agricultural fires in Eastern Europe in spring 2006*, *Atmos. Chem. Phys*. Available at: www.atmos-chem-phys.net/7/511/2007/.

S. Twomey (1977) 'The Influence of Pollution on the Shortwave Albedo of Clouds', *Journal of the Atmospheric Sciences*, 34(7), pp. 1149–1152. Available at: https://doi.org/https://doi.org/10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2.

Taylor, P.C. *et al.* (2022) 'Process Drivers, Inter-Model Spread, and the Path Forward: A Review of Amplified Arctic Warming', *Frontiers in Earth Science*. Frontiers Media S.A. Available at: https://doi.org/10.3389/feart.2021.758361.

Tunved, P. and Ström, J. (2019) 'On the seasonal variation in observed size distributions in northern Europe and their changes with decreasing anthropogenic emissions in Europe: Climatology and trend analysis based on 17 years of data from Aspvreten, Sweden', *Atmospheric Chemistry and Physics*, 19(23), pp. 14849–14873. Available at: https://doi.org/10.5194/acp-19-14849-2019.

Tunved, P., Ström, J. and Krejci, R. (2013) 'Arctic aerosol life cycle: Linking aerosol size distributions observed between 2000 and 2010 with air mass transport and precipitation at Zeppelin station, Ny-Ålesund, Svalbard', *Atmospheric Chemistry and Physics*, 13(7), pp. 3643–3660. Available at: https://doi.org/10.5194/acp-13-3643-2013.

Westervelt, D.M. *et al.* (2013) 'Formation and growth of nucleated particles into cloud condensation nuclei: Model-measurement comparison', *Atmospheric Chemistry and Physics*, 13(15), pp. 7645–7663. Available at: https://doi.org/10.5194/acp-13-7645-2013.

Wu, Y.T. *et al.* (2024) 'Stronger Arctic amplification from anthropogenic aerosols than from greenhouse gases', *npj Climate and Atmospheric Science*, 7(1). Available at: https://doi.org/10.1038/s41612-024-00696-0.

Zieger, P. *et al.* (2023) 'Black carbon scavenging by low-level Arctic clouds', *Nature Communications*, 14(1). Available at: https://doi.org/10.1038/s41467-023-41221-w.

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