Lifecycle of Black Carbon in the Arctic

Roxana S. Cremer

I have my head in the clouds, literally. I always liked the shapes and the atmosphere they create for landscape photography and since my bachelor in meteorology I love to study them in detail. How physics drives the formation, development and break up of clouds is fascinating and how clouds influence the global climate is still a piece of the big puzzle that doesn’t fit properly at each corner.

I study an aerosol particle called black carbon and its interactions with clouds up in the Arctic on Svalbard. The Arctic became one of my favourite places in the world, a second home. The calm, the only sounds are the wind and the ice, the thousands of shades of blue that fill the ocean, the ice and the sky. My data are coming form an observatory on a mountain called Zeppelin. Human influence are kept to a minimum which means that we can track the particles life with models and figure out where the particles were emitted in the atmosphere. Some aerosol particles travel for weeks around half of the globe inside of clouds to get caught by our instruments just 200km away from the North Pole.

It makes you reflect on the impact an action can have far away from the originating source.
Lifecycle of Black Carbon in the Arctic

Roxana S. Cremer

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Abstract

This PhD thesis investigates the atmospheric life cycle of Black Carbon (BC) in the Arctic. The Arctic region has been rapidly changing in the last decades and the role of BC aerosols in this is still uncertain. BC aerosols are mainly produced by incomplete combustion of biomass burning and fossil fuel and stand out from other aerosol species due to their strong ability to absorb solar radiation. The impact of BC on the Earth’s radiation budget is estimated to be overall warming. While the indirect effect, interactions with clouds, is estimated to be negative, the direct radiation effect is positive because of the absorption ability of the BC. These estimates are uncertain, especially for aerosol-cloud interactions. To estimate the role of BC in the Arctic, it is necessary to know the size distribution of BC, the transport pattern and the loss processes that affect the BC concentration. In this thesis, in-situ observations from the Zeppelin observatory in the Arctic, as well as global modelling tools, are used to answer the following research questions: 1. What kind of new insights about BC size distributions can be gained from simultaneous long-term measurements of absorption and aerosol number size distributions? 2. How do source regions impact BC size distributions measured at Zeppelin? 3. How are observations of biomass burning tracers at Zeppelin connected to transport from source regions with active fires? 4. How do emissions, as well as, wet and dry removal pathways drive the diversity of the BC life cycle in General Circulation Models (GCMs)?

A statistical method to derive BC size distributions from filter-based absorption measurements was developed and applied to long-term data from the Arctic measurement station Zeppelin on Svalbard. Promising results were obtained for inferring BC number size distributions from absorption and size distribution data, except for the most polluted conditions with the air masses arriving from Northern Eurasia and Russia - as identified from an analysis using back trajectories. Trajectory analysis was also used to link events with elevated biomass burning tracers and BC at Zeppelin to fire activity measured by satellites on the continents around the Arctic. To investigate the interplay of emissions and removal processes of BC in models and to understand the diversity in model representation of BC in the Arctic, a detailed analysis of processes in four GCMs was performed. The BC concentrations in the Arctic were compared and their response to removal processes during long-range transport to Zeppelin. The results underline the importance of BC sources and processing far away from the Arctic.

The knowledge gained about the BC life cycle will facilitate a better assessment of the large-scale influence of BC on the Arctic climate and environment.

Keywords: Black Carbon, Aerosols, Arctic, lifecycle, absorption.
LIFECYCLE OF BLACK CARBON IN THE ARCTIC

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"Det som sker i Arktis påverkar allt liv på vår vackra planet. Vi delar jorden med så mycket annat liv, så mycket skönhet. Allt vi göra har betydelse och påverkar inte bara oss själva."

from Bortom Isbjörnens Rike
by Melissa Schäfer & Fredrik Granath
Abstract

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The knowledge gained about the BC life cycle will facilitate a better assessment of the large-scale influence of BC on the Arctic climate and environment.
Sammanfattning


Den ökade kunskapen om BC:s livscykel kommer att underlätta en bättre bedömning av hur BC i en större skala påverkar det arktiska klimatet och miljön.
List of Papers

The following papers, referred to in the text by their Roman numerals, are included in this thesis.

Using correlations between observed equivalent black carbon and aerosol size distribution to derive size resolved BC mass concentration: a method applied on long-term observations performed at Zeppelin station, Ny-Alesund, Svalbard
*Tellus B: Chemical and Physical Meteorology*, **73.1**: 1–17.
DOI: 10.1080/16000889.2021.1933775

Airmass Analysis of Size-Resolved Black Carbon Particles Observed in the Arctic Based on Cluster Analysis
*MDPI: Atmosphere*, **13(5)**: 648.
DOI: 10.3390/atmos13050648

Impact of biomass burning on Arctic aerosol composition
*under review in ACS Earth and Space Chemistry*

Investigating the role of air mass history on the diversity of GCM estimates of atmospheric black carbon in the Arctic
*Manuscript*

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Author’s contribution

Paper I:
I participated in the setup of the laboratory experiment that was used to test the method for acquiring BC size distribution under controlled conditions. I collected the laboratory data together with Johan Ström and conducted all the calculations and corrections required for the related data analysis (including e.g. converting the measured intensities to absorption coefficient, correcting the absorption of the PSAP, data interpolation, wavelength correction of the MAAP data, as well as calculation of the MAC values). I produced the related Figures in the Paper (Figs. 1-6). I discussed the interpretation of the findings with Johan Ström and Peter Tunved and contributed feedback on the manuscript during the writing and review process. I was responsible for the communication with Tellus during the review process with help from Johan Ström and Peter Tunved.

Paper II:
I am the lead author of the paper which is based on the results from Paper I. I computed and analysed the trajectories and the related data, and wrote the manuscript. The analysis of the absorption signal was done together with Peter Tunved. Throughout the process, the progress and the results were continuously discussed with Peter Tunved and Johan Ström.

Paper III:
In this paper, I contributed to the air mass analysis which was done to investigate connections of biomass burning tracers observed at Zeppelin with active fire activity on the continent. I computed the trajectories, cleaned and analysed the satellite data for the fire events and produced the corresponding Figures (Figs. 8, S3). I contributed to the discussion on the links between air mass transport and observed biomass burning tracers at Zeppelin. I wrote the part of the methods describing my contribution (Sec 2.3), discussed the trajectory analysis with the lead author and provided feedback on the manuscript.

Paper IV:
I developed the research plan together with Dan Partridge based on the Experiment GCMTraj of the AeroCOM Phase III Project. The model and trajectory data were provided by the co-authors’ working groups. I collocated the trajectories with the GCM data, analysed all data, made all the Figures and wrote the manuscript. I am the lead author of the manuscript.
Contents

Abstract i
Sammanfattning ii
List of Papers iii
Author’s contribution v

1 Introduction 1

2 The Arctic climate system 5
  2.1 Arctic amplification, aerosols and clouds . . . . . . . . . . . . . . 5
  2.2 Transport to the Arctic . . . . . . . . . . . . . . . . . . . . . . . . . 6

3 Life cycle of Black Carbon 9
  3.1 Sources of Black Carbon . . . . . . . . . . . . . . . . . . . . . . . . . 9
  3.2 Black Carbon and clouds . . . . . . . . . . . . . . . . . . . . . . . . 10
  3.3 Impact on the radiation budget and climate . . . . . . . . . . . . . . 11

4 Data and methods 13
  4.1 Experimental . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 13
  4.1.1 Measurement techniques . . . . . . . . . . . . . . . . . . . . . . 13
  4.1.2 Laboratory study . . . . . . . . . . . . . . . . . . . . . . . . . . 14
  4.1.3 Measurements at Zeppelin . . . . . . . . . . . . . . . . . . . . . 15
  4.2 Numerical modelling . . . . . . . . . . . . . . . . . . . . . . . . . . . 15
  4.2.1 Trajectory models . . . . . . . . . . . . . . . . . . . . . . . . . . 15
  4.2.2 General Circulation Models . . . . . . . . . . . . . . . . . . . . . 16
  4.2.3 AeroCom . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 18

5 Results 19
  5.1 Derivation of size-resolved BC mass distribution . . . . . . . . . . 19
  5.2 Biomass burning events at Zeppelin . . . . . . . . . . . . . . . . . . 22
  5.3 Representation of BC in GCMs . . . . . . . . . . . . . . . . . . . . . 23

6 Conclusion and outlook 25

References xxix
Acknowledgements xxxvii
1. Introduction

Aerosols are small particles which are suspended in the Earth’s atmosphere. These solid or liquid particles can alter the radiation budget of the Earth through scattering and/or absorbing incoming solar radiation, interacting with clouds and hence changing the planetary albedo. Most aerosol species are estimated to have a negative radiative effect (see Fig. 1.1), meaning they cool the overall system by increasing the outgoing energy flux. Black Carbon (BC) particles are unique among the various atmospheric aerosol types, due to their strong light absorbing ability (Samset et al., 2018). Atmospheric BC can therefore have both cooling as well as warming impact on climate. The sign of the climate effects of BC aerosol during the industrial period is yet to be determined, with the current range of the effective radiative forcing since 1850 estimated to be between $-0.2$ and $+0.42$ Wm$^{-2}$ (Szopa et al., 2021).

BC particles originate from incomplete combustion of e.g. fossil fuels and biomass (Petzold et al., 2013). Natural sources of BC include forest fires and volcanic eruptions, while the largest anthropogenic sources are industry and traffic. Pure BC particles are classified as primary aerosol, meaning that it is emitted directly in the

![Figure 1.1: The multi-model mean effective radiative forcings (ERFs) due to changes in aerosols from pre-industrial times (1850) to the recent past (1995-2014). No lines indicate regions where the signal is robust across the models and 80 % of models agree on the sign of the ERF. Lines indicate no change or no robust signal and crossed lines indicate regions where the models disagree on the sign of the change. For more information see Szopa et al. (2021), Figure 6.10a.](image-url)
condensed phase rather than being formed in the atmosphere. BC is highly hydrophobic upon release, but upon atmospheric ageing, the BC-containing particles coagulate and collect vapours and other particulate species, generally making them more hydrophilic with time. With increased size and hygroscopicity, BC-containing particles can act as cloud condensation nuclei (CCN) and thus form cloud droplets. Inside a cloud, additional changes may occur to the morphology and composition of BC, influencing also cloud microphysics and their impact on the Earth’s radiation budget. The effect BC has on the Earth’s radiative budget is thought to be dominated by aerosol-radiation interaction, i.e. the absorption of incoming solar radiation which leads to the warming of the atmospheric layer surrounding the BC. This can lead to a local cooling at the surface but on larger scales, it leads to overall warming at the surface due to the mixing of the atmospheric layers. This warming is dampened by changes in clouds, water vapour and the thermal structure of the atmosphere (Szopa et al., 2021) – so-called ‘rapid adjustments’ – meaning that the overall effect of BC is smaller than the immediate radiative forcing. The aerosol-cloud interactions of BC have a cooling effect.

The Arctic is a sensitive environment which is currently undergoing rapid changes due to e.g. Arctic amplification. Disruptions in local factors, such as clouds, surface albedo and temperature can lead to irreversible changes in the environment (Goosse et al., 2018, Pithan and Mauritsen, 2014, Previdi et al., 2020, Rantanen et al., 2022). Fundamental research and e.g. defining a baseline for comparison with an altered environment in the future is necessary to estimate the consequences of changes in Arctic aerosol and their impact on the climate (Schmale et al., 2021). Many state-of-the-art General Circulation Models (GCMs) generally tend to overestimate absorption and BC concentration in the Arctic (Leaitch et al., 2020, Samset et al., 2014, Sand et al., 2021). GCMs differ in their representation of BC, including size distribution of aerosol particles, particle ageing, CCN activation, in-cloud processes related to ice formation and the removal of the particles by dry and wet deposition. Accurately describing the BC size distributions in models will help to reduce the uncertainties in the GCMs due to aerosol-cloud interactions and radiative forcing in general (Seinfeld et al., 2016). Many of these uncertainties have been proposed to be linked to e.g. wet deposition descriptions of BC (Croft et al., 2016, Ding et al., 2019) along with the relevant emission inventories.

Field observations are crucial for improving our understanding of aerosol-cloud interactions and the performance of GCMs (Schmale et al., 2021). In particular, having an accurate account of BC in the Arctic is becoming increasingly important as natural emissions of BC around the Arctic increase due to forest fires and other biomass burning (McCarty et al., 2021). It is known that long-range transport drives the BC concentration in the Arctic (Backman et al., 2021, Hirdman et al., 2010, Korhonen et al., 2008, Stohl, 2006, Stohl et al., 2007, Tunved et al., 2013). The relevant source regions and their contributions however differ between studies, pointing to anthropogenic emissions and forest fires in Russia, agricultural fires in East Europe and the possible influence of Asian BC on the Arctic. Knowing the size distribution of BC in the Arctic is necessary to understand the ability of BC to act as CCN and interact with radiation. Currently, size-dependent BC measurements in the Arctic are sparse and limited to a few short-time campaigns, mostly taking place during the haze in springtime (Abbatt et al., 2019, Ohata et al., 2019, Zanatta et al., 2018). Studying the seasonality of BC concentrations and size distributions together with the transport patterns into the Arctic will help understand how the airmasses from various
source regions are influenced by microphysical processes and precipitation along the transport.

The aim of this thesis is to increase the understanding of atmospheric processes governing concentrations and sizes of BC aerosols in the Arctic. The main focus is on the size distribution, removal processes and transport into the Arctic, utilising observational data from the research station Zeppelin Observatory on Svalbard as well as numerical modelling. The following research questions are answered in the papers compiled in this thesis:

1. What kind of new insights about BC size distributions can be gained from simultaneous long-term measurements of absorption and aerosol number size distributions?
2. How do source regions impact BC size distributions measured at Zeppelin?
3. How are observations of biomass burning tracers at Zeppelin connected to transport from various source regions with active fires?
4. How do emissions, as well as wet and dry removal pathways drive the diversity of the BC life cycle in GCMs?

The thesis is structured as follows: An overview of the Arctic climate system is given in Chapter 2 and the life cycle of BC is described in Chapter 3. Chapter 4 gives an overview of the data and methods used in the thesis. The results of the Papers are discussed in Chapter 5, before giving a summary and an outlook for future research in Chapter 6.
2. The Arctic climate system

The Arctic plays an important role in the Earth’s climate system. It is influenced by processes outside of its regional boundaries and sensitive to small changes which can result in large impacts. The changes we can observe over the last decades are an indicator of the fragility of our global climate. In this Chapter, the fundamentals of the Arctic climate system are explained.

2.1 Arctic amplification, aerosols and clouds

The concept of Arctic amplification was first introduced in the late nineteenth hundred by scientist Arrhenius, who hypothesised that an increase in carbonic acid emissions will lead to an increase in surface temperature, especially in the polar regions (Arrhenius, 1896). The increase in surface temperature is closely linked to the sea-ice loss that is observed in the Arctic, through the sea ice-albedo feedback (Curry et al., 1995). The increase in temperature leads to an increase in melting, and with the disappearing bright surfaces more incoming radiation is absorbed, triggering a further temperature increase. The negative trend of the Arctic sea ice extent is present throughout the year, being the largest during the melting season in September. The ice sheets are also thinning (Serreze and Meier, 2019, Stroeve et al., 2012). Over the last decades a number of studies, e.g. Cohen et al. (2014), Rantanen et al. (2022), Serreze and Barry (2011), Shindell (2007), Stuecker et al. (2018), Wendisch et al. (2022), have been published showing the extreme increase in the surface temperature in the Arctic – thus proving the hypothesis of Arrhenius. Rantanen et al. (2022) stated the increase in temperature to be nearly four times faster than the global average since 1979. In addition to the sea ice-albedo feedback, several mechanisms play a role in Arctic amplification, including water vapour feedback and lapse rate feedback, which influence the further continuation of Arctic amplification (Pithan and Mauritzen, 2014, Stuecker et al., 2018).

Aerosols have been suggested as a factor influencing Arctic amplification (Li et al., 2022, Sand et al., 2016, 2017, Schacht et al., 2019, Schmale et al., 2021). As explained above, aerosols interact directly with incoming solar radiation and depending on their characteristics, directly scatter and/or absorb radiation. Additionally, aerosols influence cloud formation by acting as CCN. Depending on the aerosol particle properties and concentrations, this can affect the radiative properties and lifetime of clouds. Aerosol particles are scarce in the Arctic because most natural and major anthropogenic emissions sources are located outside of the Arctic, which is commonly defined as North of 60°.

Clouds in the Arctic can both cool and warm the surface. In the summer, the reflection of incoming solar radiation by the clouds dominates, resulting in an overall cooling effect. During the polar night with no incoming sunlight, however, the absorption of outgoing thermal heat from the Earth in the lower atmospheric layers dominates, resulting in an overall warming effect. The global net effect of clouds
Figure 2.1: Temperature Change on Svalbard for the years 1898 until 2022 in relation to the average temperature between 1971-2000. Figure adapted from Ed Hawking, ShowYourStripes.info

is uncertain due to the response to climate forcing mechanisms, while estimated to be slightly warming (Boucher et al., 2013). In the polar regions, the capability of clouds to increase the planetary albedo is becoming more important with less snow and ice-covered surface (Goosse et al., 2018).

The environmental changes in the Arctic also change the conditions for aerosols and clouds, with more local sources of aerosols, e.g. more open water leading to more marine aerosol emissions (Gilgen et al., 2018). A recent study by Schmale et al. (2021) concluded that more information on Arctic aerosol emissions, development and transport in the atmosphere needs to be gathered to quantify the effects on cloud microphysics and the radiative forcing by aerosols in the Arctic with the changing conditions in the Arctic.

2.2 Transport to the Arctic

The strong temperature gradient between the Arctic and the mid-latitudes drives the transport into the Arctic region. The temperature difference is a result of the energy imbalance between the polar regions and the equator. Less solar energy reaches the Arctic surface due to the geometry of the Earth and the high albedo in the Arctic. Atmospheric transport from lower latitudes towards the poles brings heat, moisture and other chemical species including aerosols.

When the warm moist air travels northwards, it is lifted above the cold and dry air in the Arctic, forming an Arctic front (AMAP, 2015), or 'polar dome' (Stohl, 2006). Mixing between the warm and cold air masses only occurs with perturbations in the atmosphere, such as turbulence, which is limited in wintertime due to minimal solar radiation. In winter, when the northern land masses experience cool enough temperatures, the Arctic front can travel southwards, closer to the emission sources on the continents and allow low-level transport into the Arctic (Stohl, 2006). The seasonality of the source regions and transport patterns drive the aerosol concentration in the Arctic (Tunved et al., 2013). Long-range transport from sources outside of the Arctic largely determines the aerosol size distribution and composition during the winter months. Eurasia is established as the dominating source region during the winter months (Hirdman et al., 2010, Tunved et al., 2013). In the summer, during the polar
day, NPF is commonly happening in the Arctic (Tunved et al., 2013) with overall lower aerosol number concentrations and a relatively high abundance of Aitken mode particles compared to spring time. The Arctic BC is influenced by biomass burning in the form of forest and agricultural fires. This influence has been investigated through both case studies and long-term measurements. Sharma et al. (2013) appointed a third of the Arctic BC burden to biomass burning over 16 years of data. A case study of a high pollution event in spring 2006 identified small-scale agricultural fires as the main source for the elevated BC concentration (Stohl et al., 2007). Any change in aerosol emissions in the mid-latitudes outside of the Arctic will also affect the Arctic region due to long-range transport (Shindell et al., 2008). Additionally, the warming, the decline of the sea ice cover, and changes in clouds influence the atmospheric transport pathways of energy, moisture and aerosols into the Arctic (Mewes and Jacobi, 2019).
3. Life cycle of Black Carbon

3.1 Sources of Black Carbon

Black Carbon (BC) is the byproduct of incomplete combustion of fossil fuels and biomass (Petzold et al., 2013), where there is not sufficient oxygen present to convert all matter to carbon dioxide and water. BC can be emitted by natural and anthropogenic sources, and over different scales. For example, anthropogenic BC is emitted inside houses, i.e. by cooking, candlelight, and heating, on the streets by vehicles, by other means of transport like planes or ship traffic, as well as by energy production and factories. Fossil fuel combustion and open burning are large emission sources (Ramanathan and Carmichael, 2008). Natural sources of BC are biomass burning, such as forest fires (Diehl et al., 2012) and volcanic eruptions. Only a few BC emissions sources are located in the Arctic. Stohl et al. (2013) attributed some BC in the Arctic to gas flaring and residential combustion. With a sea-ice-free Arctic in the near future, it is expected however that emissions from ship traffic will increase (Gilgen et al., 2018). Biomass burning emissions may increase in the future due to the increasing number and intensity of forest fires with climate change. In the last decades, fire events have become more frequent and severe, and this trend is predicted to continue in the future (Jones et al., 2022, Van Der Werf et al., 2017). Clear atmospheric influence from regional and seasonal biomass burning events such as agricultural burning in Eastern Europe has been observed in the Arctic (Stohl et al., 2007). The long-range transport of aerosol particles into the Arctic is the subject of several studies, and these show various dominating source regions for different measurement sites in the Arctic (Willis et al., 2018).

For anthropogenic aerosol and aerosol precursor gases, global models have to rely on emission data sets, while for aerosols of natural origin (e.g. sea salt, biogenic emissions and dust) emission parametrisations that depend on e.g. wind speed, temperature and other environmental factors are commonly used. BC emission inventories are typically produced by combining information from several different sources. Large-scale biomass burning and burned area can be derived by using satellite products, like the active fire count from the MODIS instrument or the change in vegetation detected using satellite images. For emissions from industries or agricultural burning, BC data are generally provided by various state agencies and then scaled (Bond et al., 2004, Klimont et al., 2017). Emission inventories are recognised as a major source of model uncertainty, especially for BC. The uncertainty in BC emissions has been estimated to lie in between a factor of 2 and 3, depending on the emission source (Bond et al., 2013, Klimont et al., 2017) and region (Ramanathan and Carmichael, 2008).
3.2 Black Carbon and clouds

Many freshly emitted BC particles are hydrophobic, non-spherical aggregates with low particle density. Shortly after emission, the BC particles transform by collapsing to dense smaller clusters and start to grow in the Aitken mode (size range 10-70 nm) through coagulation with co-emitted particles and condensation of gas phase species (AMAP, 2015, Gustafsson and Ramanathan, 2016). As the particles grow to larger sizes they become generally more hydrophilic due to the condensation of hygroscopic compounds like sulphuric acid, which also influences the optical properties of the particles making them more spherical. Once the BC-containing particles have collected enough soluble material, they act as cloud condensation nuclei (CCN) and contribute to the formation of a cloud droplet if high enough water vapour supersaturations are reached. In cold temperatures, BC-containing particles might even act as ice nucleating particles (INPs), facilitating ice crystal formation in clouds. The role of BC-containing particles as potential INPs is however very uncertain (Hendricks et al., 2011, Vergara-Temprado et al., 2018).

Although freshly emitted BC is considered a poor CCN, the BC transported into the Arctic is more likely to act as CCN due to the aging processes taking place in the atmosphere (AMAP, 2015). The collection of hydrophilic material by condensation and coagulation alters the hygroscopicity and composition of the particle. In the Arctic, the CCN are in general scarce and several studies suggest that aerosol particles even within the Aitken mode can contribute to Arctic CCN (Croft et al., 2016, Heintzenberg et al., 2015, Korhonen et al., 2008, Willis et al., 2018) - as opposed to higher activation diameters required at more polluted regions. This was shown even for particles with low hygroscopicity in the case of limited accumulation mode particles (Bulatovic et al., 2021).

BC particles can decrease the precipitation rate of the cloud (AMAP, 2015). Smaller cloud droplets associated with BC can result in a prolonged cloud lifetime, which can reduce the availability of water vapour for precipitation formation, ultimately leading

![Figure 3.1: Schematic of the life cycle of BC, indicating the sources on the bottom and processes in the atmosphere with the radiative implications related to BC particles in the atmosphere.](image-url)
to lower precipitation rates. BC particles can also change the temperature structure in the cloud by increasing the absorption in the cloud and thus contribute to evaporation. With increasing size, BC particles generally are more susceptible to wet deposition. Wet deposition is the main removal mechanism for BC, commonly divided into below-cloud and in-cloud scavenging. Below-cloud scavenging describes the process where precipitation removes the BC particles below the cloud by collision and collection. The other process is in-cloud scavenging when BC particles act as CCN, become a cloud droplet or ice crystal, and ultimately precipitate out (Croft et al., 2010). In mixed-phase clouds ice-growing processes, like the Wegener-Bergeron-Findeisen process play a role. The Wegener-Bergeron-Findeisen process describes the ice crystal growth at the expense of evaporating cloud droplets. When the cloud droplets evaporate the aerosol particles inside, like BC, are redispersed into the atmosphere.

### 3.3 Impact on the radiation budget and climate

BC influences the Earth’s radiation budget through aerosol-radiation interactions (direct effect) and aerosol-cloud-interactions (indirect effect). Overall the effective radiative forcing, i.e. the net effect compared to pre-industrial conditions, from these interactions is calculated to be positive (Fig 3.2, Szopa et al. (2021)), but the uncertainties are large so that the overall forcing could also be slightly cooling. Aerosol-radiation interaction describes the direct effect aerosols can have on the radiative budget at the top of the atmosphere (TOA). As BC particles are highly absorbing, they absorb incoming solar radiation and emit long-wave radiation heating the surrounding atmosphere. Additionally, a layer of BC particles lowers the planetary albedo. Changes in the temperature profiles weaken the radiative-convective coupling between the atmosphere and surface, and with lower temperatures at the surface evaporation and rainfall are inhibited (Ramanathan and Carmichael, 2008).

Clouds can strongly impact surface temperatures, and their effects are highly variable between cloud types and regions. A common cloud type in the Arctic is low-level mixed-phase clouds. These clouds exist in the planetary boundary layer with both liquid water and ice particles present. They form in temperatures below 0°C and may exist until about -38°C (Szopa et al., 2021), at which point cloud droplets will homogeneously freeze. The role of BC in mixed-phase clouds is not fully understood yet. Schmale et al. (2021) suggest that observations on the effects of INPs and improvements in the description of ice-containing clouds in GCMs could close some of these knowledge gaps.

After the removal from the atmosphere, BC can influence the surface albedo, especially if it is deposited on bright surfaces such as on the Arctic ice-covered ocean. BC darkens the surface and less sunlight is reflected. Due to this, the melting rate increases, slowly diminishing the snow and ice surfaces. Combining the direct and indirect effects, the globally averaged positive effective radiative forcing (ERF) of BC is about 0.1 Wm$^{-2}$ (Szopa et al., 2021). However as mentioned earlier, the uncertainties are large, and it cannot be concluded that the overall effect of BC on the Earth’s radiation budget is indeed warming. Locally, the impact can be important, especially close to the sources of BC and in regions, where BC influences precipitation patterns (Liu et al., 2018, Ramanathan and Carmichael, 2008). In addition, changes in global BC evoke large responses in the Arctic, showing the importance of rapid adjustments in the remote radiative forcing (Sand et al., 2013, 2016, Stjern et al., 2017).

In comparison to greenhouse gases, such as methane and CO$_2$, the effects of aerosols
Figure 3.2: Contribution to effective radiative forcing (ERF) from component emissions between 1750 to 2019 based, Figure adapted from Szopa et al. (2021), based on data by Thornhill et al. (2021).

The effect of mitigating aerosol emissions would therefore be expected to be observed faster than for greenhouse gases. Szopa et al. (2021) state that it is likely that BC mitigation helps offset warming effects by emission reductions of sulphur dioxide. Regionally, BC mitigation could have an overall cooling effect on the surface but the interplay of the aerosol-cloud interactions is complex and the impact varies and requires more studies. Reducing BC emissions would limit their direct warming effect (AMAP, 2015), but would likely be offset by rapid atmospheric adjustments, resulting in a relatively weak mitigation impact overall (Takemura and Suzuki, 2019). Decreasing BC pollution on local scales, however, creates more liveable and healthy cities and helps decrease the number of premature deaths due to particulate matter (Kühn et al., 2020, WHO, 2021).
4. Data and methods

The life cycle of BC is influenced by processes on various spatial scales, from the nano-scale microphysical processes to transformation over days while transported in the atmosphere. Therefore, to answer the research questions of this thesis, different data and analysis tools were used.

4.1 Experimental

4.1.1 Measurement techniques

Black Carbon

BC can be measured by optical, thermal, incandescence, acoustic and microscopic methods (AMAP, 2015, Petzold et al., 2013). In this thesis, the focus was on optical measurements, which are inexpensive methods for measuring BC aerosol continuously, utilising its strong light-absorbing characteristics. The two instruments used are the Particle Soot Absorption Photometer (PSAP) and the Multi-Angle Absorption Photometer (MAAP). Both are filter-based instruments which measure the decrease of light intensity with time on the filter collecting absorbing aerosol.

The PSAP works with a single angle attenuation of light and the comparison of two simultaneous filter measurements to determine their optical transmission. An increase in attenuation of light over a particle-loaded filter surface provides, in comparison with a reference filter, the absorption coefficient ($\sigma_{\text{abs}}$). Several corrections have to be applied to account for e.g. scattering enhancement as described by Bond et al. (1999). The MAAP works on the same principle as the PSAP, but uses four photodetectors to account for multiple scattering. The photodetectors are set up at different angles to measure light attenuation, multiple scattering and absorption enhancement caused by reflection.

The BC mass concentration is calculated by multiplying the absorption coefficient with the mass absorption cross-section (MAC). The MAC value is a quantity that describes how effective the absorption signal is per area of BC. MAC values vary greatly between regions and studies due to aerosol age. Several studies are published showing the variation of the MAC values with instrumentation and for different Arctic measurement sites (Ohata et al., 2021, Zanatta et al., 2018). The MAC value can be calculated from the absorption coefficient and BC mass data:

$$\text{MAC} = \frac{\sigma_{\text{abs}}}{\text{BCmass}} \text{[m}^2\text{g}^{-1}]$$

Size distributions

Aerosol size distributions (see Fig. 4.1) are, in this thesis, measured with a Differential Mobility Particle Sizer (DMPS). A DMPS couples a Differential Mobility Analyser (DMA) with Condensation Particle Counter (CPC) to produce aerosol size
Figure 4.1: Schematic of the size distributions of aerosol particles, with the nucleation mode with diameters below about 10 nm, the Aitken mode with a diameter between about 10 and 100 nm, the accumulation mode from about 100 to 1000 nm and the coarse mode with particle diameters above 1 \( \mu m \).

Distributions. A DMA measures the size of particles by their electric mobility \( Z_p \), which can be derived from Stokes’ law:

\[
Z_p = \frac{n \cdot e \cdot C_c}{3 \pi \cdot \eta \cdot D_p} \tag{4.2}
\]

The electric mobility depends on the number of charges on each particle (\( n \)), the elementary charge (\( e \)), the Cunningham correction factor (\( C_c \)), the viscosity of air (\( \eta \)) and the particle diameter (\( D_p \)). The Cunningham correction factor accounts for the deviation of fluid flow behaviour at very small length scales and is especially important for small particles (Seinfeld and Pandis, 2016).

4.1.2 Laboratory study

For Paper I, a laboratory study was designed to link measured light absorption to particle size, using the setup shown in Fig. 4.2. One PSAP (PSAP1 in Fig. 4.2) measured the ambient aerosol absorption of background Stockholm air. In parallel a second PSAP (PSAP2 in Fig. 4.2) was connected after a DMPS to measure the absorption coefficients of small pre-set increments of the aerosol size distribution in the range of 11 – 540 nm. The data recorded by PSAP2 were then integrated over all size bins and compared to the total absorption measured by PSAP1. The hypothesis was that the correlation between the aerosol number concentration and the total absorption coefficient can give information about the particle size distribution of BC. Therefore the Pearson correlation coefficient (\( r^2 \)) between the absorption coefficient with the number concentration of each size bin of the DMPS was calculated and the conversion described in Hull (1927) was applied to derive the scaling factor \( f_s \):

\[
f_s = 1 - \sqrt{1 - r^2} \tag{4.3}
\]

Multiplying the scaling factor \( f_s \) with the median absorption signal for each size bin and dividing by the MAC value results in a BC mass size distribution.
4.1.3 Measurements at Zeppelin

The Zeppelin Measurement Station is situated on the Arctic archipelago Svalbard at a latitude of 78.9°N and longitude of 11.9°E. The station is located above the town Ny-Ålesund, on a mountain, at 474 m asl. The station was established in 1993 and collects data on long-term trends of aerosols and their physical characteristics and chemical composition (Platt et al., 2022). The collected data are important for fundamental and applied aerosol research, as they are one of the longest corresponding time series and provide valuable insights into background aerosol concentrations. Zeppelin is considered a background station because it is located on a mountain, far away from major industrial and natural sources. In this thesis, the absorption coefficient and size distribution data from 2002 until 2010 are used for Papers I and II, from years 2009 - 2013 for Paper IV and year 2020 for Paper III. For Papers I, II, IV absorption coefficient data from the PSAP was used. For Paper III data from different instruments employed for the 1-year long Ny-Ålesund Aerosol Cloud Experiment (NASCENT) campaign (Pasquier et al., 2022). The aerosol chemical composition data in Paper III are based on the analysis of measurements by a FIGAERO-CIMS. The FIGAERO-CIMS is a high-resolution time-of-light mass spectrometer which measures gases and aerosol via a filter inlet. The BC data are from a MAAP and additionally, particulate matter data were taken from an optical particle size spectrometer FIDAS.

4.2 Numerical modelling

4.2.1 Trajectory models

Trajectory models are well-known tools for studying potential source regions for transport to a receptor site. The Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT; Stein et al. (2015)) developed by NOAA was used in this thesis to investigate source regions of Arctic BC over eight years (Paper II), specifically for biomass burning aerosol (Paper III) and to collocate model data along the trajectories to study BC’s transport into the Arctic in GCMs (Paper IV). The model
uses meteorological fields from reanalysis or model data sets as the input conditions to compute the trajectory movement with time. Backwards trajectories, following an air mass from a measurement location back in time, is a common method of identifying potential source regions. In all studies, the backward trajectories were calculated for ten days to span over the lifetime of BC in the atmosphere, as described in AMAP (2015), Boucher et al. (2013), Stohl (2006). Also, the model was run in ensemble mode for Papers II and III meaning that for each starting time, not one trajectory was calculated, but 27, with a small perturbation at the starting location. This increased the statistics for the studies while all trajectories are representative of the starting conditions. Several different meteorological input data were used for the studies. In Paper II the reanalysis data were taken from FNL (Final Analysis of the National Centers for Environmental Prediction) and GDAS (Global Data Assimilation System) on a 1° × 1° grid for both data sets. In Paper III GDAS data were used as well and for Paper IV trajectories were calculated based on ERA-Interim and the meteorological fields from the models used in the study.

4.2.2 General Circulation Models

GCMs describe the interactions between the atmosphere, ocean and biosphere on a global scale. They can provide valuable information on our complex Earth system and understand the connections and impacts of different processes with each other. They are a beneficial tool to model experiments on future climate. Sub-grid scale processes in GCMs are typically described with parameterizations, i.e. simplified mathematical expressions instead of the physical calculation of the process in question. In Paper IV the global modelling focus is mainly on process understanding studying the BC life cycle and how different models describe processes related to aerosol transport and removal. The models used for Paper IV are the UK earth system model (UKESM) and three configurations of ECHAM, a general circulation model developed by the Max Planck Institute (see Tab 4.1 for more information).

Aerosol size distributions

Size distributions of aerosol particles in GCMs are commonly described by several log-normal modes covering nucleation, Aitken, accumulation and coarse mode, divided into soluble and insoluble fractions (Mann et al., 2010, Stier et al., 2005, Tegen et al., 2019). Another option is to describe the aerosol size distribution with a sectional aerosol scheme dividing the size distribution into finite size bins (Kokkola et al., 2018). In the modal approach, more assumptions about the shape of the size distribution are integrated making it less precise, but less computationally heavy compared to a sectional aerosol scheme. In the latter, the larger number of size bins compared to modes are calculated for each time integration of the model.

The specific limits to the size ranges, either modal or sectional, vary slightly between the GCMs. In the modal schemes of the GCMs used in Paper IV, BC is represented in the Aitken, accumulation and coarse modes with the mean radii being between 5 nm and 1 µm (Mann et al., 2010, Tegen et al., 2019). In the sectional aerosol scheme in ECHAM-SALSA BC is assumed to have a geometric mean radius between 40 nm and 4 µm (Bergman et al., 2012, Holopainen et al., 2020, Kokkola et al., 2018). In all the GCMs, BC is emitted into the insoluble Aitken mode, but over time moved to the soluble modes due to coagulation and coating with soluble species (Croft et al., 2010, Holopainen et al., 2020, Mann et al., 2010, Stier et al., 2005).
Table 4.1: Models used in Paper IV including relevant additional information: The resolution is given in latitude and longitude in degree and number of vertical levels in the model. The aerosol scheme used in the model set up, nudging settings to Era-Interim and relevant references.

<table>
<thead>
<tr>
<th>Model</th>
<th>Resolution</th>
<th>Aerosol emissions dataset</th>
<th>Aerosol scheme</th>
<th>Nudging</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>UKESM1</td>
<td>1.25x1.875x85</td>
<td>CMIP6+CMIP6 Biomass Burning (GFED4) + tuning over biomass burning areas</td>
<td>Open</td>
<td>GLOMAP, modal</td>
<td>ERA-Interim 6hr horizontal wind</td>
</tr>
<tr>
<td>ECHAM6.3-HAM2.3</td>
<td>1.875x1.875x47</td>
<td>CMIP6+CMIP6 Biomass Burning (GFED4)</td>
<td>Open</td>
<td>HAM-M7, modal</td>
<td>ERA-Interim 24hr surface pressure, 6hr vorticity, 48hr divergence</td>
</tr>
<tr>
<td>ECHAM6.3-HAM2.3-P3</td>
<td>1.875x1.875x47</td>
<td>CMIP6+CMIP6 Biomass Burning (GFED4)</td>
<td>Open</td>
<td>HAM-M7, modal</td>
<td>ERA-Interim 24hr surface pressure, 6hr vorticity, 48hr divergence</td>
</tr>
<tr>
<td>ECHAM6.3-SALSA2</td>
<td>1.875x1.875x47</td>
<td>CMIP6+CMIP6 Biomass Burning (GFED4)</td>
<td>Open</td>
<td>SALSA, sectional</td>
<td>ERA-Interim 24hr surface pressure, 6hr vorticity, 48hr divergence</td>
</tr>
</tbody>
</table>
an example, this process takes place within a day so that nearly 90% of the insoluble BC mass becomes soluble before the removal from the atmosphere (Stier et al., 2005).

BC emissions

BC emissions in the analysed GCMs are prescribed by emission inventories and contain volcanic emissions, biomass burning and anthropogenic emissions. A common inventory used in GCM experiments is the Global Emission Fire Dataset (GFED), which is developed from in-situ data and satellite observations. Differences in the models originate from tuning the original data or in the injection height into the model. An example of tuning the BC emissions is UKESM, where the emissions over biomass burning areas are intensified to be closer to the observed aerosol optical depth (Johnson et al., 2016). The injection height for biomass burning emissions is often split up over the atmospheric column, e.g. following (Val Martin et al., 2010), where three-fourths are evenly distributed within the planetary boundary layer, 17% in the level closest to the surface and lastly above the boundary layer.

Removal

Removal processes in models are split into dry and wet deposition. Dry deposition occurs via gravitational sedimentation and the removal of aerosol particles in the lowest model layer by turbulence, which depends on the surface roughness in the model (Mann et al., 2010, Stier et al., 2005, Tegen et al., 2019, Zhang et al., 2012). Wet deposition happens via cloud processing and precipitation. Models have parametrisations implemented for several different processes, i.e. below cloud scavenging, in-cloud nucleation scavenging and convective scavenging. Kipling et al. (2013) introduced a specific parametrisation for the removal of aerosols by precipitating convective clouds, this is called plume scavenging and is implemented into UKESM.

The removal processes can be quantified by calculating the loss rate coefficient (LR, days$^{-1}$):

$$ LR = \frac{\text{deposition rate}}{\text{column burden}} \quad (4.4) $$

LR can be calculated for all removal processes or each process individually for each aerosol species and is a measure of how effective the removal process acts on the aerosol in the GCM.

4.2.3 AeroCom

The AeroCom-project is an international framework to further the understanding and impact of aerosol particles on the Earth’s climate. Therefore models and observations from different sources (satellites and surface observations) are combined to analyse and evaluate the current state of aerosol modelling in GCMs. The project is organised in phases, where under each phase, several studies take place simultaneously, with a common experiment protocol established to make a multi-model intercomparison and evaluation possible. The project is now in its third phase. Paper IV utilises the data of the Aerosol GCM Trajectory (GCMTraj) experiment of AeroCom Phase III. This experiment studies aerosol processes in GCM in a Lagrangian framework. Data fields from the GCM are collocated along trajectories, and calculated based on the meteorological fields of the GCMs. The goal of the experiment is to improve the understanding of sources and sinks in GCMs, the performance of their parametrisations and the spread between the contributing models.
5. Results

5.1 Derivation of size-resolved BC mass distribution

In Paper I, the relation between absorption and size distribution was analysed using a combination of a laboratory study and measurements at the Zeppelin Observatory.

In the first part of the laboratory study, the two PSAPs were compared before the experiment illustrated in Fig. 4.2 was conducted. The two-time series compare well to each other over time, but the integrated absorption is about a factor of two higher than the total absorption (see Fig. 4, (Tunved et al., 2021)). The two possible explanations for this are that either light-absorbing particles have e.g. a different charge probability distribution from non-absorbing particles and therefore need to be corrected differently in the DMPS, or the filter measurements behind the DMPS result in a different optical response due to the limited size range (<540 nm) or the mixing state of the particles as compared to the total absorption. Comparison of the scaling factor $f_s$ with the size-resolved absorption coefficient measured by PSAP2 reveals that the two approaches result in maximum absorption around the same particle size but more particles end up in the Aitken mode when the scaling factor $f_s$ is used as suggested by the PSAP2 measurement (Fig. 5.1).

![Figure 5.1](image-url)

**Figure 5.1**: Comparison of the truncated and normalised Pearson correlation scaling factor $f_s$ (red) and the normalised median $\sigma_{\text{abs}}$ distribution based on DMPS and PSAP2 measurements (blue). The dotted lines are log-normal fitted distributions and the y-axis is normalised to 1. Figure taken from (Tunved et al., 2021).
The approach using the scaling factor $f_s$ was applied to size distribution and absorption measurements at Zeppelin. For the period of 2002 until 2010 consecutive parallel measurements of aerosol size distributions and filter-based absorption measurements were used to explore the statistical relationship between the two data sets and the derivation of size-resolved BC mass distribution. The aerosol size distributions were clustered and grouped into four categories to simplify the analysis (see Fig. 5.2). The most common size distribution group is denoted 'Washout' with nearly half of all size distributions belonging to this category. The least frequent groups denoted 'Nucleation' and 'Intermediate', are connected to the seasonal sunlight cycle in the Arctic. The last group with about a third of the data was called 'Polluted' and shows influences of long-range transport of pollutants. For each group, the scaling factor $f_s$ was derived and a corresponding BC mass distribution was calculated. A comparison of the derived BC mass distribution with SP2 measurements from Svalbard, even though these are limited, shows that they agree reasonably well with each other (see Fig. 10. in Tunved et al. (2021)). Next, the BC mass distributions were converted to number distributions with the assumption that BC is externally mixed and using the MAC value of $9.4 \, \text{m}^2 \, \text{g}^{-1}$ from the laboratory study. Reasonable BC number size distributions were derived for most of the data at Zeppelin station, except for the 'Polluted' cases (see red curves in Fig. 5.2). For the 'Washout', 'Nucleation'

![Figure 5.2: Comparison between observed particle size distributions and inferred number size distribution of BC, $dN_{BC}/d\log D_p$. The solid black lines represent medians and the error bars show the upper and lower quartiles for the observed size distribution. The solid red line is based on the median absorption coefficient for each category of clusters and the shaded area is based on the upper and lower quartiles (Tunved et al., 2021).](image-url)
and 'Intermediate' groups, the predicted BC number concentration was generally in the range of the total aerosol number concentration and follows the overall shape of the distribution. For the 'Polluted' group, however, the calculation produced a second peak in the Aitken mode hence overestimating the absorption around 50 nm, exceeding the total aerosol number in this mode. This method of deriving the number size distribution is directly linked to the MAC values used in the conversion. With a greater MAC value, the number of BC particles per size bin would decrease.

The overall particle size distributions for the 'Polluted' group display a high number concentration in the Accumulation mode with very few particles in the Aitken mode. Hence the hypothesis is that within this group BC concentrations might correlate with 50 nm particles unrelated to light absorbance. In other words, the cases with high absorbance seem to be connected with simultaneous increases in Aitken mode particle concentrations. Therefore a closer look at this group was taken in Paper II. It was discovered that the scaling factor $f_s$, derived for the whole group does not display the characteristics of the whole group very well and leads to an under-/overestimation of the absorption coefficient. Therefore, the polluted group was split into two subgroups, which show similarities in the chemical footprint and size distribution but are different in regard to absorption.

For these two subgroups, the BC mass concentration was recalculated with individual scaling factors $f_s$. With this new scaling factor $f_s$ for Subgroup 1, a reasonable bimodal BC number concentration is displayed while in Subgroup 2 the statistical approach breaks down and shows a peak in BC number concentration around 50 nm with twice the amplitude of the total aerosol number concentration peak in the Accumulation mode (see Fig. 2 in Paper II). The origin of the air mass connected to these two subgroups was analysed. Figure 5.3 shows the effective potential source contribution for the two subgroups, which indicates the probability of the location contributing to the BC signal measured at Zeppelin. The BC in Subgroup 1 most likely originated from Western Europe and the North Atlantic. These regions are linked to precipitation events and cloud processing, which is in line with the predicted bimodal BC number size distribution. The source region for Subgroup 2 lies in Northern Eurasia and Russia. The transport in this group was less affected by

![Figure 5.3: Normalised effective potential source contribution of absorption for the 'Polluted' group split into subgroup 1 (a, representing lower absorption) and subgroup 2 (b, representing higher absorption). The red star marks the location of Zeppelin Observatory. Figure is taken from (Cremer et al., 2022).](image_url)
precipitation events and shows a higher concentration of BC. Also, the effective potential source contribution highlights the region east of the Ural Mountains across to the Kamchatka Peninsula, a region that in other studies is not associated as a main origin for air masses measured at Zeppelin. In the air masses arriving from these regions there hence seems to be an interrelationship between Aitken mode particles and BC, showing a strong correlation while the particles in this size range do not necessarily contain any BC.

5.2 Biomass burning events at Zeppelin

Paper III looks into the chemical signature of biomass burning aerosol at Zeppelin and investigates its potential source regions with back trajectory analysis. Seven distinct peaks in BC concentration and the biomass burning tracer levoglucosan occurring during the year-long study were used as case studies. A specific period in time was classified as a biomass burning event if levoglucosan and BC mass both exceeded the 97th percentile of the monthly measurements. Particle number size distributions generally display a peak in the accumulation mode for all biomass burning events (see Fig. 5 in Paper III). Most of the events show a higher O:C ratio (oxygen to carbon ratios) compared to non-event times (see Fig. 3 in Paper III). In the chemical composition for the events, a notable increase in BC and organics together with a significant relative reduction in sulphate could be observed in comparison to non-events.

To link the observations at Zeppelin to biomass burning on the landmasses surrounding the Arctic, an analysis of the source regions with trajectories and the fire activity detected by satellites was performed (see Fig. 5.4). Not all biomass burning events at Zeppelin could be neatly tied to fire activity along the back trajectories. For some

**Figure 5.4:** Integrated fire radiative power with the transport probability of 10-day back trajectories to Zeppelin during biomass burning events in 2020. The figure is taken from Gramlich et al. (2023). The events in July (Event 4) and November (Event 7) were less certain events, in which the increase in levoglucosan and BC was small as compared with the other events.
cases (Event 5) the air mass analysis showed nearly no contact with land or close proximity to the fire activity from MODIS. For other events, like Event 6, the connection between the transport pattern and fire activity was clear. For Event 6, the biomass burning recorded at Zeppelin originates from agricultural fires close to the Black Sea. This analysis highlights the importance of transport from the Eurasian region and agricultural fires in determining Arctic aerosol properties, especially during the haze season. The analysis of the precipitation along the trajectory paths for the biomass burning events show similarly mixed results as for the transport analysis. Events with transport affected by precipitation can still show strong signals of biomass burning indicators at Zeppelin Station, while others with little to no precipitation show lower concentrations of BC and levoglucosan. In conclusion, linking the measurements at Zeppelin to fire activity with a transport analysis shows ambiguous results for most events and needs further investigation of the effect of precipitation and e.g. fire altitude (see also Figs. S3 and S4 in Paper III).

5.3 Representation of BC in GCMs

**Paper IV** investigates the BC aerosol evolution during transport in a Lagrangian framework for four GCMs, to improve the understanding of the role of sources and sinks on BC concentration at Zeppelin. The study looks into five years of data (2009 – 2013) for the GCMs UKESM and ECHAM, the latter in three configurations (HAM, P3, and SALSA). Using three versions of ECHAM allows for investigating specifically the role of the aerosol scheme and the removal parametrisations.

First, an Eulerian overview was made to investigate the main differences in the average Arctic BC column burden between the models. ECHAM-P3 showed twice as much BC in the Arctic in comparison to the other models. Large differences were shown in the loss rate coefficient, because of the different descriptions of dry and wet removal in the models. UKESM and ECHAM-SALSA showed high wet effective sink rate coefficients. The dry effective sink rate coefficient in UKESM was at least a factor of three larger compared to the other models.

The analysis in the Lagrangian framework confirms these results. 10-day back trajectories were calculated, clustered and grouped into transport sectors: the Arctic Ocean (Fig. 5.5a), Eurasian (Fig. 5.5b) and Greenland Sea (Fig. 5.5c) transport. In each of the transport groups, ECHAM-P3 shows a considerably larger BC column burden during the back time of the trajectories. On the opposite side of the scale, UKESM and ECHAM-SALSA are very close to each other and follow the same evolution, especially in the Greenland Sea and Arctic Ocean transport. As expected, the Arctic Ocean transport results in the lowest and Eurasian transport the highest BC burdens. The importance of dry deposition is highest in the Eurasian transport case, where the trajectories cover more landmass during their travel to Zeppelin, as the dry deposition in the models depends on the surface roughness and BC concentration. ECHAM-P3 is the least efficient in the removal of BC, which could be linked to the change in the description of the ice processes in this model. The model-to-model differences in BC column burden cannot be fully explained by the loss coefficients during the trajectory back time. The results therefore indicate that ten days are not enough to study the key drivers of the BC transport, and a longer lifetime of BC in the Arctic. The BC concentration at Zeppelin is driven by long-range transport and the key changes and interactions of emissions and removal processes take place outside of the Arctic. This underlines the need to mitigate emissions in mid-latitudes to minimize the negative consequences of climate change on the Arctic.

23
Figure 5.5: Overview of the three transport groups and the related diagnostics co-located along the trajectories. The first column shows the Greenland Sea Transport, the second the Eurasian Transport and the third column the Arctic Ocean Transport. The transport frequency (a,b,c), the BC column burden (d,e,f), the dry loss coefficient (g,h,i) and the wet loss coefficient (j,k,l) for the four included models: ECHAM-HAM, ECHAM-P3, ECHAM-SALSA and UKESM.

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6. Conclusion and outlook

In this thesis, experimental and theoretical tools were used to characterise BC aerosols and the relevant processes in the Arctic. In particular, aerosol microphysical properties, origin and pathway to the measurement station Zeppelin on Svalbard were investigated.

In **Paper I** long-term data from Zeppelin were statistically analysed to answer the question: *What kind of new insights about BC size distributions can be gained from simultaneous long-term measurements of absorption and aerosol number size distributions?* We find that our method of deriving BC mass distribution compared well with a reference method based on measurements with the SP2 instrument. This shows that a statistical relationship based on the correlation of the total measured absorption with the size bins of the measures aerosol number distribution has potential to be used to investigate the size distribution of BC particles, and that the developed method can be a valuable tool for assigning a size distribution to BC from long-term observations. The mass of BC was found to be mostly located in the accumulation mode between 180–250 nm, while in the BC number size distribution most particles are in the Aitken mode with a maximum located around 40–50 nm. Converting the BC mass distribution to a number distribution with the derived MAC of 9.4 m$^2$g$^{-1}$ produces results aligning with total aerosol number size distribution, except for the most polluted cases with high levels of both absorption and aerosol particle loadings in general (see also **Paper II**).

**Paper II** aimed to study the air mass transport into the Arctic for derived BC distributions of **Paper I** and the seasonality as studied by Tunved et al. (2013) to understand in particular *How do source regions impact BC size distributions measured at Zeppelin?*. The focus of the study was on the 'Polluted' size distributions because of the overprediction of BC particles in the Aitken mode.

To investigate possible reasons for the overestimation of BC Aitken mode particles the 'Polluted' group was split into a low- and high-absorbing subgroup. For both subgroups, a separate scaling factor $f_s$ was then derived which led to a reasonable BC number size distribution in the first subgroup, but a continued overestimation of the Aitken mode particles for the second, more absorbing, subgroup. This overestimation could not be fully explained, several hypotheses were listed as potential explanations. One is in the assumptions for our calculations, assuming a fully internal mixing state, or the assumed density over the sizes and the MAC value, which could, in reality, be size-dependent. Also, we could not include particles larger than 630 nm due to the size limitations of the DMPS. Lastly, the statistical relationship could be sensitive to small particles which covary with BC. The trajectory analysis for the size distributions classified as 'Polluted’ show transport from Eurasia and a large fraction of the data is related to the haze season. The group is associated with high aerosol numbers and elevated particulate matter. Both subgroups are associated with transport from...
Eurasia. The less absorbing subgroup shows BC transport from Western Europe and the North Atlantic and the more absorbing subgroup shows transport from the East of Russia with a higher BC signal. Further characteristics of the particles in the second subgroup will be an interesting topic for future study.

**Paper III** focuses on biomass burning cases studied at Zeppelin Observatory to answer the following question: *How are observations of biomass burning tracers at Zeppelin connected to transport from various source regions with active fires?*. The case studies were defined by high levels of levoglucosan and BC measurements during the NASCENT campaign in 2020. Finding a relation between biomass burning events and fire activity on the landmasses surrounding the Arctic was investigated with a trajectory analysis. While for all events fire activity was observed from satellite data and in most cases in the proximity of the transport calculated with the back trajectory model HYSPLIT, a common pattern for all events could not be established. A closer look into the precipitation during the transport, the transport height and also the characteristics of the fires are needed to provide further insight explaining the subtleties of the biomass burning signal at Zeppelin.

**Paper IV** concentrates on the representation of BC in GCMs and focuses on the question *How do emissions, as well as wet and dry removal pathways drive the diversity of the BC life cycle in the GCMs?* The comparison between the models shows major differences in the BC column burden in the Arctic. Wet deposition is the main removal pathway and is dependent on the parametrisations in the models. UKESM has the most efficient wet removal. In ECHAM-P3 the wet removal is reduced as a result of the different description in the ice processes, causing a BC column burden twice as large as for the base version ECHAM-HAM. Dry deposition plays a minor role during the transport into the Arctic and becomes more important close to Zeppelin Station. The influence of emission during the back trajectory time can only be observed when the airmasses arrive from the Eurasian place. Long-range transport older than ten days is therefore the main driver of BC concentrations in the Arctic, and the main contributors to the BC column burden are located further South, outside of our study area. None of the models reproduces the haze season at Zeppelin. While UKESM and ECHAM-SALSA are close in magnitude, ECHAM-HAM and ECHAM-P3 overestimate the BC concentration in the haze season as well as in the winter. Studying the removal processes and emissions of BC in a Lagrangian framework gives useful information about the efficiency and interplay between the different removal processes. Such studies can help guide the further improvements of the GCMs. It is necessary for the future to continue the efforts made in recent years to measure BC and related processes directly in the Arctic as well as to analyse the transport to the Arctic.

Using statistical tools, like those introduced **Paper I**, can hopefully help analyse the existing data in the Arctic to be able to quantify the current changes in more detail together with trend analyses of absorption, BC concentrations and changes that might appear in the air mass transport to the Arctic. A valuable next step to improve the method and test its robustness would be to run an experimental setup with the laboratory setup used in **Paper I** in parallel with an SP2 to directly compare the measurements under the same ambient conditions. This could also tell us the importance of small BC particles below 80 nm, which are not measured by the SP2 measurement but seem to appear when the statistical method is applied. Trajectory analyses like those utilised in **Paper II-IV** are a commonly used tool to
investigate possible sources and origin regions for pollutants in the Arctic. While for case studies trajectories do not always give the full transport history of the measured aerosol properties, they can give valuable information on the influence of long-range transport. By looking in more detail into trajectories over longer periods, transport patterns and seasonality can be investigated. The likelihood of specific source contributions can be studied and related to aerosol properties, as done in Paper II. This can help investigate changes in transport and influences of source regions on aerosol properties with the ongoing changes in the Arctic.

For Paper IV specifically, further information on the interactions and processes along the trajectory could be analysed by adding precipitation and cloud data to the analysis. Comparing the precipitation effect on the BC aerosol concentration at the station in the GCMs with the relation shown in Tunved et al. (2013) for Zeppelin and in Isokääntä et al. (2022) in the boreal forest region will help evaluate the performance of the GCMs and how well they describe aerosol-precipitation interactions. Additionally, observational data from ERA-Interim could be added to compare the models with observations. This would in particular involve looking into cloud cover and how much time the air mass spends in clouds in the GCMs and the reanalysis data to estimate the influence of the cloud processes on the BC burden in GCMs and how accurately the GCMs reproduce the observations. Furthermore, calculating how much time the air mass spends over land or ocean could give an estimate of the travel speed of the air mass to Zeppelin. Continuing to understand the processes will benefit the modelling community to better describe BC concentrations in the Arctic and reduce uncertainties in aerosol-cloud interactions.

This PhD thesis studied the life cycle of BC in the Arctic with in-situ measurements and modelling tools. Information was gathered about BC size distributions at Zeppelin Observatory, transport patterns into the Arctic on a seasonal and case-study basis and the representation of BC in GCMs. The gathered results and new methods will help future investigations in the Arctic research community to describe the BC concentration and its impact on the environment.

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