Quantification of sources and removal mechanisms of atmospheric aerosol particles

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Abstract
The focus of this work has been to quantify important processes for climatically relevant aerosols, and to improve our understanding of, and ability to accurately model, aerosols in the atmosphere on a large scale. This thesis contains five papers focused on different parts of the life cycle of atmospheric aerosol particles. Two papers describe the physical process of emission of primary marine aerosols. The large uncertainties in these processes are demonstrated by examining the diversity of existing parameterizations for emissions. Building from laboratory experiments to validation of model results with observations, new parameterizations are suggested. These take into account also effects of water temperature on primary marine aerosol production. In the third paper the main focus was to develop a new aerosol wet removal scheme in the Lagrangian transport and dispersion model FLEXPART. Removal timescales and atmospheric concentrations are found to be close to observation based estimates. The final two papers focus on atmospheric black carbon aerosols at high latitudes. As an example of increased human activities in the Arctic, local emissions from cruise ships visiting the research base in Ny Ålesund had demonstrable effects on the level of pollutants measured there. In contrast, inland Antarctic air was shown to be clean compared to the Arctic, due to the extremely long transport time from any major aerosol sources. The work done in this thesis has addressed critical uncertainties regarding the aerosol lifecycle, by better constraining aerosol emissions and atmospheric lifetimes. The development of the new wet removal scheme has improved FLEXPART model accuracy, which will be beneficial in future applications of the model.

Keywords: aerosol, aerosol removal, aerosol emission, aerosol sources, FLEXPART, Arctic aerosol.

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Sammanfatning

List of Papers

The doctoral thesis consists of a summary and the five papers listed below:

**PAPER I**

**PAPER II**

**PAPER III**

**PAPER IV**

**PAPER V**
Contribution of Grythe, H.:

**PAPER I**: Performed the model simulations, data analysis and wrote the manuscript.

**PAPER II**: Performed FLEXPART model simulations, did most of the data analysis and wrote sections on FLEXPART model description and FLEXPART results.

**PAPER III**: Conceptualized and implemented the removal scheme. Performed most model simulations and data analysis. Wrote the manuscript.

**PAPER IV**: Procured the shipping data and took part in the data analysis.

**PAPER V**: Performed the model simulations, did most of the data analysis and wrote the manuscript.
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Abbreviations

ACCMIP Atmospheric Chemistry and Climate Model Intercomparison Project
AEROCE Aerosol Oceanic Chemistry Experiment
AeroCom Aerosol Comparison between Observations and Models project
AIS Automatic Identification System
BC black carbon
CC total surface cloud cover
CCN cloud condensation nuclei
CP convective precipitation
CTBTO The Comprehensive Nuclear-Test-Ban Treaty Organization
CTWC cloud total water content
d diameter
dW perturbation by a stochastic Wiener-process
DMPS Differential Mobility Particle Sizer
DMS dimethyl sulphide
ECLIPSE Evaluating the CLimate and Air Quality ImPacts of ShortlivEd Pollutants
ECMWF European Centre for Medium range Weather Forecasts
GFED The Global Fire Emissions Database
IFS Integrated Forecast System
IN ice nuclei
LSM land sea mask
LSP large scale precipitation
m mass
MAC mass absorption coefficient
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>MODIS</td>
<td>Moderate resolution Imaging Spectroradiometer</td>
</tr>
<tr>
<td>NOAA</td>
<td>US National Oceanic and Atmospheric Administration</td>
</tr>
<tr>
<td>NorESM</td>
<td>The Norwegian Earth System Model</td>
</tr>
<tr>
<td>NWP</td>
<td>numerical weather prediction</td>
</tr>
<tr>
<td>OPC</td>
<td>Optical Particle Counter</td>
</tr>
<tr>
<td>PA</td>
<td>Polar Amplification</td>
</tr>
<tr>
<td>PSAP</td>
<td>Particle Soot Absorption Photometer</td>
</tr>
<tr>
<td>RH</td>
<td>relative humidity</td>
</tr>
<tr>
<td>SEAREX</td>
<td>Asian dust network in the North Pacific</td>
</tr>
<tr>
<td>SOA</td>
<td>secondary organic matter</td>
</tr>
<tr>
<td>SSA</td>
<td>sea spray aerosol</td>
</tr>
<tr>
<td>SST</td>
<td>sea surface temperature</td>
</tr>
<tr>
<td>T</td>
<td>temperature</td>
</tr>
<tr>
<td>t</td>
<td>time</td>
</tr>
<tr>
<td>U_{10}</td>
<td>10 metre wind speed</td>
</tr>
<tr>
<td>v</td>
<td>wind vector</td>
</tr>
<tr>
<td>VOC</td>
<td>volatile organic compounds</td>
</tr>
<tr>
<td>X</td>
<td>position vector</td>
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</tbody>
</table>
1 Introduction

Aerosol is derived from the Greek word "aero" -air and "sol" -solution, and is a term applied to solid or liquid materials suspended in the air. Aerosol particles have varied physiochemical properties and span several orders of magnitude in size, but are limited to particles with atmospheric residence time above some hours. With this limitation, aerosols are considered to range approximately from 2 nm to 20 μm in diameter. Quantification of aerosols particles in a given volume of air can be done by e.g. their number density, surface area, mass, size, source, chemical or optical properties.

Primary aerosols are particles entering the atmosphere as particles, secondary aerosols are formed in the atmosphere through condensation of gases. In the troposphere both types of particles thus ultimately have the Earth’s surface as a source, either directly or by emissions of aerosol precursors (Fig. 1). While secondary particles dominate in number, primary particles mainly contribute to the mass and aerosol surface area. Aerosol sources can also be divided into being of natural or anthropogenic origin. Natural sources significantly contribute to both the primary and the secondary aerosols burden. Mineral dust and primary marine aerosols have predominantly natural origin and they also represent the majority of the aerosol mass found in Earth´s troposphere. Biomass fires emit black carbon (BC) particles (primary aerosols), ash debris, and gases that subsequently can form secondary aerosols. Human induced changes to natural aerosol levels, via altering of the environment, can also be considered anthropogenic.

From their various sources, aerosols and precursors are spread around the atmosphere by winds and turbulence (Fig. 1). Though highly heterogeneous in time and space, aerosols are present from pole to pole and ubiquitous in the atmosphere and not limited to the regions near their sources. Atmospheric residence times can range from hours up to several weeks in the troposphere, and much longer than this above the tropopause.

Aerosols are intimately linked to clouds. On one hand, aerosols acting as cloud condensation nuclei (CCN) impact cloud formation and cloud microphysical properties, on the other hand, precipitation from clouds has a large impact on the atmospheric aerosol concentrations. Precipitation is the main removal mechanism for most aerosol particles. In particular for particles in the accumulation mode size range (d=0.1-0.6 μm), it is the only efficient way by which
they are removed from the atmosphere. Smaller, Aitken mode particles (d = 0.01-0.1 μm) can efficiently be removed by dry deposition near the Earth’s surface. Large particles will fall through the air towards the surface and gravitationally settle on the surface. Only in the lowest layer of the atmosphere can aerosols attach to the surface and be removed by dry deposition. The removal rate determines the atmospheric lifetime that together with the strength of the emission controls the concentration of an aerosol species. Particles can also be removed by losing their identity as individual particles. Very small particles are prone to coagulate with other particles, which make them disappear as individual particles, but the material they were made of remains suspended in the air.

Although aerosol particles are tiny and often invisible to the human eye, they do interact with the Earth’s atmospheric radiation budget. With a projected increase in human activities, it is very important, and urgent, to study processes that control the fate of both natural and anthropogenic aerosol. The environment of the Polar Regions where the impacts are larger than the global average is the specific focus of this thesis.

Figure 1: The aerosol lifecycle. Aerosols have their origin near the surface and their physical and chemical characteristics dependent on the source. Urban air pollution is a concern for health but also feeds the troposphere with pollution that can influence weather and climate. From oceans come sea spray, deserts produce mineral dust, and forests emit gases that later form particles. Given sufficiently long enough residence time, particles can reach the free troposphere and influence cloud formation and the radiative budget far from their sources. The main removal mechanism for particles traveling far will mainly be through precipitation scavenging.
1. 1 Aerosols in the climate system

Climate is long term average weather. Aerosols are an integral part of the atmospheric system and influence climate. Climate is impacted by aerosols either directly by affecting the radiative budget, or indirectly by influencing radiation through acting as cloud condensation nuclei (CCN). Aerosols have even been shown to directly influence winds in large scale weather systems (Chakraborty et al., 2015). The radiative effects of anthropogenic aerosols remain the largest uncertainty in climate simulations, mainly due to shortcomings in our scientific understanding about aerosol interactions with clouds (Chalmers et al., 2012; Boucher et al., 2013; Myhre et al., 2013; Hartmann et al., 2013).

In what is known as the direct aerosol effect, aerosols scatter and absorb both longwave and shortwave radiation and thus change the energy balance of the atmosphere (McCormick and Lundwig 1967; Joseph et al., 1973; Srivastava and Satheesh 2016). The efficiency of an individual particle to absorb or scatter radiation depends on the particle refractive indices, determined by the radiation wavelength and the radius and chemical composition of the aerosol (e.g. Dubovik et al., 2000; Haywood and Shine 2000; Levy et al., 2013).

Aerosol scattering occurs mainly for shortwave solar radiation (e.g. Wang et al., 1974; Ångström 1962), and has a strong net cooling effect on the climate (Lohmann and Feichter 2001). Important aerosols that predominantly scatter shortwave radiation, and therefore have a strong cooling effect, are sea spray aerosols (SSA) and sulphate aerosols. For this reason, several geoengineering schemes propose injection of these aerosols to the atmosphere to temporarily counteract global warming (Feichter and Leisner 2009). The direct radiative effect of naturally occurring SSA alone is estimated to contribute to a global average climate forcing of -0.6 to -4.0 Wm⁻², which is comparable in size to the total forcing of a doubling in CO₂ (Satheesh & Moorthy 2005).

The most important aerosol that has a large proportion of light absorption is soot, or BC (Haywood and Boucher 2000). Both scattering and absorption lower the irradiance at the surface. However, absorption of incoming radiation by aerosols can, over highly reflective surfaces, have a net warming effect. Although this warming effect is known to be smaller than the cooling provided by the lowered irradiance, the location and composition of the aerosol particles and partition between absorption and scattering determine the net magnitude of the cooling by the direct aerosol effect (e.g. Haywood and Shine 1995;
Lohmann and Feichter 2001. The total climatic impact of an aerosol is a function of its intensive properties and lifetime in the atmosphere. Hence, aerosol lifetime is of principal importance to the Earth’s energy budget.

The *aerosol indirect effects* is a term composed of the ways in which aerosol particles influence clouds and alter the energy balance. The indirect effect of aerosols can easily be seen from space by satellites in the bright lines caused by ship emissions in low level clouds (Schreier et al., 2007). Clouds have on their own both strong heating and cooling effects on the energy budget. Overall, clouds cool the Earth’s climate significantly, but a single cloud may be either warming or cooling depending on location, solar elevation and microphysical properties of the cloud (e.g. Lohmann and Feichter 2005; Garrett and Zhao 2006). The individual radiative effects of a cloud are so variable and large that even small changes to the Earth’s cloud population can have a large impact on the radiative budget. Most cloud microphysical aspects are linked to the availability of CCN and ice nuclei (IN) on which cloud droplets and ice crystals may form. This abundance may also influence the extent, radiative properties and formation of precipitation from clouds in a family of indirect aerosol effects, which are poorly quantified.

Hence, to lower the uncertainty of aerosol radiative effects is a key aspect of predicting climate change (Myhre et al., 2013). Anthropogenic aerosols introduced into the atmosphere add to the aerosol concentration from natural sources. A first step in understanding the human influence is to understand the natural emissions to the atmosphere. An accurate representation of natural aerosol levels is therefore an as important part of reducing uncertainties in anthropogenic aerosol climate forcing, as understanding anthropogenic emission themselves (Carslaw et al., 2012).

The observed climate change is not uniform all over the globe and in the Arctic region this warming is larger than the global average. This enhanced warming rate is known as *polar amplification* (PA). PA is described as the relative change in temperature of Polar Regions to the change in global temperature for a given forcing (Manabe and Stouffer 1980). The factor of PA has been estimated to be 2-3 for the Arctic and 1.2 for the Antarctic (Taylor et al., 2013). This means that a 2 degree global average temperature increase would correspond to a 4-6 degrees change in the Arctic which may therefore be more susceptible to anthropogenic perturbations than the rest of the globe. This warming is coupled to multiple effects, e.g. more ocean surface being exposed to the atmosphere as the sea ice retreats earlier and forms later in the year.

The existence of PA-effects due to the lowering of albedo by the melting of snow and ice surfaces has long been an accepted part of the climate system
(Holland et al., 2003; Screen et al., 2010), but also other mechanisms generating PA have been identified and quantified (Taylor et al., 2013). The main difference between the two poles is that whilst the Arctic consists of an ocean surrounded by continents, Antarctica is a continent surrounded by ocean. In contrast to the Arctic, where increased cloudiness will amplify temperature changes, Antarctic cloudiness does not increase to the same extent and so the southern warming is assumed overall smaller than its northern counterpart (Taylor et al., 2013). Also, in the Arctic, vast land areas may be affected by the longer snow free seasons, which is not a large effect in Antarctica. Besides the direct climatic impact less snow covered land has, altering the reflectivity of the Earth’s surface, it also subsequently affects many other processes due to the thawing of the permafrost and the availability of liquid water.

Studies of atmospheric thermal energy transport show that the exchange of polar airmasses with lower latitude air will change as a consequence of climate change (Shepherd 2014). Transport of air is also transports aerosols. Changes in this transport will cause changes to the composition of aerosols in the Polar atmosphere by bringing in more aerosols originating outside the Polar Regions (Paasonen et al., 2013). Thus, in addition to changes in internal Arctic aerosol sources, changes in transport patterns can be expected to influence the overall aerosol composition at high latitudes.
2 The aerosol lifecycle

To advance our knowledge about aerosol emission sources, transport and transformations, and ultimately the removal of aerosols from the atmosphere, there is a need to use a variety of different tools. For instance, we need observations of the ambient properties to characterize our environment, but it is not possible to observe everything. The world is a very complex system and often details are obscured by the many interactions that take place at the same time. Laboratory studies can help simplify the system and detailed processes such as the production of SSA from the oceans can be better understood. However, the conditions in the laboratory are not necessarily representative for the real world conditions. Numerical models are mathematical descriptions of our current knowledge about processes and the system they build. Hence, the models will be par with our level of process understanding. In furthering our understanding, models and observations complement each other through validation of models and interpretation of observations. However, to predict the climate in the future, numerical models are our main tools.

The rate of change in mass ($m$) of particles inside a given volume can be described as the difference between sources and sinks:

$$\frac{dm}{dt} = \sum sources - \sum sinks$$  \hspace{1cm} (1)

where sources are emissions, atmospheric production (secondary production) and transport into the volume. The sinks are transport out of the volume and deposition to the surface, either by precipitation, by dry deposition or by gravitation. For individual particles also atmospheric transformation can be considered as a loss processes. Together these processes make up the aerosol lifecycle.

At each stage of a particle life cycle, different processes play more or less important roles. As particles are introduced into the atmosphere, either emitted as primary particles or formed as a result of condensing gases, their start may look very different. In particular, processes related to gas-to-particle conversion are highly non-linear. Over many regions of the globe natural and anthropogenic sources of aerosol pre-cursor gases (e.g. organic vapours, sulphate and nitrate, and ammonia) interact with solar radiation to form new particles
in the atmosphere and also add material to already existing particles. This pathway of forming new particles and growing existing particles is a highly non-linear multi-component physiochemical process. Actually, most particles formed this way will lose their identity as an individual particle very quickly and add to the growth of larger particles. Nevertheless, some will grow all the way to become cloud nuclei. Hence, chemical processes in the early stage of particle formation as well as inside cloud droplets, are very important aspects of the aerosol life cycle. This research field is very large and great efforts are made, to try to implement these non-linear processes in climate models (e.g. Kulmala et al., 2007). Because of the limitations in the Lagrangian model tool, which do not directly handle non-linear processes, this work did not treat the gas-to-particle conversion pathway in the aerosol life cycle.

While aerosol particles are also relevant to urban air pollution, it is in their large scale climatic effect that the aerosols investigated in this thesis (both natural and anthropogenic) have their main importance. The main objective has been to improve our description of the processes of emission, transport and wet removal in numerical models.

2.1 Emissions

The atmosphere and the entire surface of the Earth exchange material in the form of gases or particles. From this stems the complexity of aerosols, which are as diverse as their sources. Aerosol particles and semi-volatile gases that can form aerosols in the atmosphere are emitted from oceans, deserts, roads, forests and biota, fires and volcanoes as well as from combustion (e.g. Seinfeld and Pandis 2006; Tegen 1997).

Quantifying aerosol emissions and improving relevant numerical formulations are two of the key steps towards improving our understanding about the role of aerosols in the climate system. This can be done by starting with the physical process of emission directly by scaling up local or experimentally determined emissions to global scale (bottom-up approach), or by indirectly estimating emissions by taking known atmospheric concentrations (often derived from indirect satellite observations) and scaling them back to emission (top-down approach). Both methods rely on understanding the physical processes involved. Both approaches have strengths, weaknesses and uncertainties and a combination of both is often preferred (Pechony et al., 2013).
Emission estimates from Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) for mass emitted annually for main components of aerosols are shown in Table 1. The oceans and exposed dry land is the source of the two largest constituents of aerosol particles, SSA and mineral dust. The emitted mass of SSA and dust are roughly two orders of magnitude higher than that of other aerosol particles. Most of this mass is made up of coarse particles (d >1μm). These estimates are based on bottom-up emission inventories relying on physical parameterizations (Dentener et al., 2006; Ginoux et al., 2001, 2003; Gong et al., 2000). Sulphur is emitted from a large variety of sources. Dimethyl sulphide (DMS) is primarily from oceanic algae (Boucher et al., 2003) and secondary organic aerosol (SOA) formed from natural volatile organic compounds (VOC) (Kanakidou et al., 2005). The uncertainty factor given in Table 1 is based on the range of existing emission inventories and variability and uncertainty factors in these (Dentener et al., 2006). Both emissions of SSA and mineral dust are wind driven. Winds do not only transport the air containing aerosol particles, but it is also the direct cause of emissions of almost 95% of the aerosol mass released to the atmosphere (Table 1).

Table 1: The largest groups of aerosols and their main sources. Emitted mass is taken from Textor et al., (2007) and Dentener et al., (2006) and are emissions used for Aerosol Comparison between Observations and Models (AeroCom). Where given the typical range of aerosol diameter is given. The range of the estimates is given from the variability in existing emission inventories.

<table>
<thead>
<tr>
<th>Type of Aerosol</th>
<th>Main Source</th>
<th>Size</th>
<th>Tg yr⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>SSA</td>
<td>Oceans</td>
<td>0.01 -10μm</td>
<td>8200±80%</td>
</tr>
<tr>
<td>Mineral dust</td>
<td>Deserts</td>
<td>0.01 -10μm</td>
<td>1640 ±50%</td>
</tr>
<tr>
<td>Sulphur</td>
<td>Ocean, volcanoes, anthropogenic.</td>
<td>0.01 -0.3μm</td>
<td>183 ±30%</td>
</tr>
<tr>
<td>BC</td>
<td>Anthropogenic &amp; natural</td>
<td>0.01-0.3μm</td>
<td>7.8 ± 80%</td>
</tr>
<tr>
<td>DMS</td>
<td>Oceans</td>
<td></td>
<td>26±60%</td>
</tr>
<tr>
<td>SOA</td>
<td>Biogenic</td>
<td></td>
<td>19±80%</td>
</tr>
</tbody>
</table>
2.1.1 Sea spray aerosol (SSA)

Over oceans, winds cause surface stress, which in turn generate ocean surface waves. The main mechanism by which SSA is produced is well known. When a wave grows too steep and the crest of the wave reaches a critical point, the wave brakes from the water surface. Bubbles are entrained into the water and will burst when they resurface. Water droplets with dissolved matter is released into the atmosphere. When the droplets evaporate, most of the matter left is composed from NaCl mixed with other trace elements and highly variable contribution of organic matter. While open water and wind are required to form breaking waves and bubbles that can burst, there are several poorly quantified factors that may influence both the wave breaking process and the bubble bursting process (e.g. Lewis and Schwartz 2004). Organic matter produced by ocean biological activity produce surfactants, and ocean temperatures are thought to influence both the amount and chemical composition of SSA produced (O’Dowd et al., 2004).

Emission estimates for SSA span more than one order of magnitude. Emissions from parameterizations or prescribed inventories are used in climate models, and the uncertainty factor of the emission term is carried onward through predictions made by the models. Being wind-driven, emissions of SSA are most commonly bottom up parameterizations of physical processes, which depend on atmospheric input. High accuracy of the emission term is therefore a fundamental part of understanding the processes SSA undergo in the atmosphere (Leeuw et al., 2011).

While SSA is relatively inert in the atmosphere the particles span several orders of magnitude in diameter covering virtually the whole size range of aerosol particles from 0.01 to 20 μm. The aerosol size distributions also vary between different estimates. Consistent SSA emission estimates are therefore needed for the whole size distribution on a level of size dependent factors as a function of wind speed, temperature and other influencing factors (Leeuw et al., 2011; Jaegle et al., 2011). In addition to the large uncertainties in the dependence of SSA emissions on wind speed, several studies have shown a strong effect of the SSA emissions on changes in sea water temperature (Mårtensson et al., 2003; Jaegle et al., 2011; Zábori et al., 2013; Struthers et al., 2013). It has also been suggested that variations in salinity can influence the emissions from the ocean (Mårtensson et al., 2003; Sofiev et al., 2011; De Leeuw et al., 2011).
2.1.2 Black carbon (BC)

BC is an important type of aerosol particle. It is emitted as a result of incomplete combustion and can occur both naturally and from anthropogenic sources. Forest fires are a large and natural source of BC. Agricultural burning and burning of fossil fuels are large anthropogenic sources. While the emitted BC mass (see Table 1) 7.8 (2-29) Tg yr\(^{-1}\) (Bond et al., 2013) is small compared to SSA, it is a climatologically relevant aerosol due to its optical properties, causing a net warming effect on the Globe (Myhre et al., 2013). Globally, natural emissions make up a third of emitted BC (Evangeliou et al., 2016).

The Global Fire Emissions Database (GFED) is a satellite information based fire activity map. Monthly gridded burned area and emissions from fires are included in the product (http://www.globalfiredata.org). Emitted BC is calculated based on emission factors, which depend on the type of vegetation that is burning. Satellites give snapshots collected to give pseudo global coverage and not continuous coverage. GFED v3 is based on the area burned, which is derived by coupling moderate resolution imaging spectroradiometer (MODIS) fire pixel counts with surface reflectance images (Giglio et al., 2009; Giglio et al., 2010). This widely used emission inventory has uncertainties that arrive both from the emission factors and also from the amount of burnt material. A comparison of this bottom-up inventory with top-down inventories found large regional differences, and top-down estimates were about 30% higher (Bond et al 2013).

The ECLIPSE (Evaluating the CLimate and Air Quality ImPacts of ShortlivEd Pollutants) v4 inventory, is based on fuel consumption inventories. Regional and fuel type specific emission factors are used (for details see Klimont et al., 2016). Excluding agricultural waste burning, all anthropogenic emissions were taken from this inventory, which results in annual BC emissions of, 5.4-6.2 Tg yr\(^{-1}\).

2.2 Transport Modelling

Nearly all aerosol sources are at or close to the surface. Thus, vertical lifting of aerosol particles depends on turbulent vertical motions. Within the surface boundary layer aerosols are mixed efficiently when turbulence is present. As there is less turbulence in the free troposphere, further vertical mixing by turbulence requires more time spent in the atmosphere. Correct estimates of atmospheric concentrations are thus not only dependent on emissions, but also
require knowledge of the atmospheric residence time and how aerosols are moved through the atmosphere and ultimately removed.

Vilhelm Bjerknes suggested first in 1904 that, by solving a set of nonlinear equations, it would be possible to predict weather. Initially driven primarily by military interests, predictions of winds, clouds and fogs became a sought after commodity. The first attempts to numerically predict weather were made by Richardson in 1922 (Richardson 2007), using pen and paper. The early developers of computers recognized the problem of weather forecasting was a well suited application for their machines. In 1950, as one of the first applications of computers, the first weather forecasts were made from numerical models. Since then, the development of atmospheric models has been a continuous incremental process, encompassing ever more aspects of atmospheric processes. Numerical models have now been developed with increasing sophistication and details over the past decades to not only predict weather, but also climate and transport of pollutants.

One of the largest constrain has always been computational capacity. Our ability to accurately find solutions are dependent on simplifications or parameterizations of processes we cannot resolve (Knutti 2008). Furthermore, the growing number of tasks performed by atmospheric numerical models requires different aspects to be resolved with high precision. Examples of this are highly resolved regional and local weather forecast, chemical transport models. In addition a hierarchy of models exist to improve our understanding of underlying processes, dealing with aerosol processes, cloud microphysics, and small scale turbulence.

One such category of models is transport and dispersion models. Originally their primary ability was to trace conserved quantities through the atmosphere. However, the late 1970’s saw the emergence of pollution transport modelling, e.g. linking remote sulphur dioxide emissions in central Europe to acidic rain in Scandinavia (LRTAP 1979). This problem not only dealt with emission and transport, but also dispersion, chemical transformation and removal from the atmosphere. This spurred transport models to evolve further to be able to deal with new challenges. The more recent link found between aerosols and climate poses additional challenges for the quality and accuracy at which transport models can trace aerosols through their lifecycle.
2.3 Removal

Near the surface aerosols may attach directly to the surface and thus be removed from the atmosphere by a process called dry deposition (e.g. Sehmel 1980; Hinds 2012). Since only very large aerosol (d >4-5 μm) have gravitation strong enough to fall through the atmosphere (Maxey 1987) most aerosol particles are advected by winds and primarily removed from the atmosphere by precipitation.

The process of wet deposition describes how aerosol particles are removed by precipitation. Wet deposition is a term used collectively for both impaction scavenging and nucleation scavenging by both liquid and solid phase water. Extensive theories have been developed to describe both processes (e.g. Köhler 1925; Greenfield 1957). Once in the free troposphere the only removal outside the surface layer is with precipitating water. If not for precipitation, small aerosols would remain in the atmosphere for months and the amount of these atmospheric aerosols would be increased by more than one order of magnitude (Grythe et al., 2016). Atmospheric aerosol concentrations are thus closely linked to the atmospheric water cycle.

The mechanism of how aerosol particles attach to falling precipitation (impaction) has been shown to be primarily dependent on the precipitating droplet- and aerosol particle- size (Giorgi and Chameides 1986; Andronache 2003; Laakso et al., 2003). For rain, it was first described by Marshall and Palmer (1948) that the raindrop size distribution can be well described by the precipitation intensity. For snow the relationship between aerosol removal and precipitation intensity is less direct, as snow crystal shape has a large impact.

However, especially for submicron aerosol particles there are large differences between observed removal rates and those predicted by theory. This discrepancy primarily comes from a strong sensitivity to both aerosol particle size and falling raindrop size, but may also be related to variable atmospheric properties such as turbulence and temperature (Laakso et al., 2003). Small measurement inaccuracies can therefore lead to differences of one order of magnitude or more (Volken and Schumann 1993). Therefore, in models empirical relationships of impaction scavenging are generally preferred over first principle calculations (Andronache 2003).

Though the process of nucleation is hard to measure or simulate in controlled experiments, it is of critical importance to understand, not only for the removal
of aerosols but also to estimate aerosol indirect effects. When cloud droplets grow to precipitation, falling to the surface, only the activated CCN are removed. In liquid clouds, an aerosol particle’s ability to act as CCN inside clouds has been shown to be determined by an estimated 90% aerosol particle size rather than the chemical properties of the aerosol (Dusek 2006). Though recent progress has been made in also understanding the role of chemistry (e.g. Riipinen et al., 2015), a large part of the variability comes from aerosol concentration and updraft velocity.

It is not only temperature that determines the cloud water phase, but also the availability of IN. The water phase of a cloud is important for climate feedbacks, among these is the formation of precipitation. The abundance of IN at a given temperature is a major source of uncertainty, primarily because it is poorly understood which aerosols act efficiently as IN (Joly et al., 2014). Overall a much lower fraction of aerosols have been found in ice than in liquid (9% and 92% respectively, Verheggen et al., 2007). Strategies to implement removal of aerosols in global models therefore must consider substantial uncertainties related to what aerosols are immersed in clouds. Overall, removal is also dependent on the assumptions made about cloud phase dependence in the conceptual framework used. To identify key uncertainties it is therefore critical to validate new strategies against observations.
3 Research objectives

The overarching aim of this thesis was to reduce uncertainties in aerosol emission source strengths, transport pathways, and their atmospheric removal processes and fate. The principle tool used in this thesis is the FLEXPART Lagrangian particle transport and dispersion model. In order to address the research objectives, FLEXPART was improved with respect to the representation of climatically relevant aerosol populations in the model. This required the development of tools and techniques to improve the comparison between the model and global observational data sets, and led to new parameterizations of the removal of particles from the atmosphere. The accuracy of the improved version was assessed using global data sets, and specifically tested against observations in the Arctic where the uncertainty in the output of the model is expected to be the greatest. Simply stated, in the instances where modelled and observed disagree, we can be rather sure that we lack in understanding of the controlling processes.

Generally, this thesis addresses several fundamental processes controlling the abundance of an aerosol species in the atmosphere; namely, emission strengths and removal processes. For this work, focus was given to two very important, but different types of aerosols, SSA and BC. The former is white and dominated by natural processes over the oceans. The latter is black and is dominated by anthropogenic activities over land.

The specific research objectives of this thesis are to:

1) Evaluate the relative importance of the different physical parameters in existing parameterizations that are considered important for emission of SSA.

2) Develop an aerosol property specific wet removal scheme for FLEXPART and assess model accuracy in simulated long range transport of pollutants.

3) Assess how aerosol concentrations in the Arctic and Antarctic are different, with respect to sources and transport paths. In particular, characterise the optical properties of aerosols in the Antarctic interior.
4 Methodology

Elaborate numerical computer models are used to make predictions of future climates. On large computers, these models numerically solve sets of discrete equations. These equations can either be derived from first principles or be parameterized (Knutti 2008). Even with rapidly increasing computational power, atmospheric models that treat aerosol do not resolve the microphysical properties of individual aerosols and mainly rely on parameterizations based on empirical relationships. Improving parameterized aerosol behaviour is therefore one of many important tasks needed to obtain higher accuracy in climate predictions.

The model used in this study uses historical forecast weather input fields that have been assimilated with observations. This means that the state of the atmosphere at each point in time is reproduced to the best of our knowledge. Large scale weather patterns are therefore well reproduced, whereas smaller scale variations are too detailed and must be represented by model parameterizations.

4.1 Modelling Tools

4.1.1 FLEXPART Lagrangian model

For most of the work in this thesis a Lagrangian transport and dispersion model has been applied. The equations of motion for the atmosphere can be solved from two perspectives. In the Eulerian perspective, air flows through a fixed point while the Lagrangian perspective follows a parcel of air through the motions. The “FLEXible PARTicle dispersion model” (FLEXPART) is an off-line Lagrangian model which solves the trajectory differential equation:

\[
\frac{dx}{dt} = \nu[X(t)]
\]  

(2)
Here, the position vector $\mathbf{x}$ rate of change over time ($t$) is solved by a first
order Taylor expansion (Stohl et al., 1998). The wind vector $\mathbf{v}$ is composed of
three scales of motion, grid- ($v_{\text{nwp}}$), meso- ($v_{\text{meso}}$) and turbulence- ($v_{\text{turb}}$) scale
motion:

$$ \mathbf{v} = v_{\text{nwp}} + v_{\text{meso}} + v_{\text{turb}} \quad (3) $$

FLEXPART is a linear model and solves Eq. (2) by a first order Taylor
approximation which is accurate to the first order. The standard version of
FLEXPART (Stohl et al., 2005) uses input meteorology from European Centre
for Medium range Weather Forecasts (ECMWF) for $v_{\text{nwp}}$. Trajectories then
follow the mean flow as given by the input 3D fields. Additionally, the NWP
fields include surface 2D fields of cloud cover (CC), large scale (LSP) and
convective (CP) precipitation and 3D fields of temperature (T) and specific
humidity (and more recently cloud water (CTWC)). For small enough time
steps, the solution of Eq. (2) by the iterative Petterssen (1940) scheme is nor-
mally sufficiently accurate to the level of the interpolation of wind fields de-
scribing $v_{\text{nwp}}$.

The large scale wind fields do not include mesoscale motions $v_{\text{meso}}$ which is
included by a parameterization, solving the Langevin equations (Thomson
1987), using the variance of the wind in surrounding grids (Stohl et al., 1998).
Additionally, turbulence ($v_{\text{turb}}$) for an individual trajectory is parameterized
using the Langevin equations:

$$ dv_{\text{turb}} = a_i(x,v_{\text{turb}},t)dt + b_i(x,v_{\text{turb}},t)dW_j \quad (4) $$

In this form, $a$ is the drift term of the turbulence, $b$ is the diffusive term and
d$W$ represent a random incremental perturbation (Legg and Raupach 1982).
In FLEXPART, $dW$ is from a Gaussian distribution, independent in time and
space. As trajectories follow the mean flow, the stochastic turbulence is su-
perimposed to emulate diffusion in the atmosphere. There is no numerical dif-
fusion (as is the case for Eulerian models) when the trajectory equation is
solved. Numerical diffusion in Eulerian models come from the limitation of
resolving small scales numerically. However, in a Lagrangian model such as
FLEXPART the diffusion term arrives only from the turbulence imposed on
the trajectory. This more physical way of treating diffusion can be an ad-
vantage when compared to Eulerain models.
In all applications in this thesis the input meteorology was taken from the ECMWF global reanalysis data sets. This data set is assimilated historical numerical Integrated Forecast System (IFS) model fields (Forbes et al., 2011; Forbes and Tompkins 2011) with observational data, to form our best guess on how the atmosphere was at a given time in the past. Although FLEXPART also can be in forecast meteorology mode this option is not applied in this thesis.

An advantage of using a Lagrangian framework is the compatibility between simulating transport both forward and backward in time. Often a few observation points are relied upon for monitoring pollutants. Sources are typically diverse and generally spread out. It is therefore more computationally effective to do backward mode simulations if the number of emission sources exceeds the number of observations. Additionally, it is advantageous that each measurement can efficiently be linked to an individual emission at a given time and place, also when there are large numbers of emission points. This feature of Lagrangian models was utilised in this thesis when investigating emission and removal uncertainties, as FLEXPART was, for the most part run in backwards mode from observation points.

In forward mode, FLEXPART simulates trajectories of emissions. The results are given on a user-specified 3D output grid at given times after the emission. The mass of species emitted represents the actual mass, and mass concentrations are calculated by combining the masses of each individual trajectory inside each grid cell. In backward mode trajectories start at the receptor site (e.g. measurement site) and the concentration is calculated diagnostically after each simulation step by multiplying the emission fields by the output emission sensitivity fields as post processing. The emission sensitivity fields produced by the backward simulation have an additional advantage when investigating source term uncertainty. Only one backward FLEXPART simulation per observation is needed as the emission sensitivity field is independent of source strength. Therefore only the post processing needs to be applied to derive atmospheric concentrations from different source terms.

FLEXPART v9 was used in PAPER I & II. It consists of subroutines written in FORTRAN 95, and is run on Linux machines. A beta version of FLEXPART V10 (Kristiansen et al., 2016 in prep.) was used in consecutive papers. This version contains several updates, among them the new cloud and wet removal scheme described in PAPER II. The FLEXPART v10 code and previous versions are freely available from www.flexpart.eu. The FLEXPART model description of version 10 is still in preparation form, but some details of the cloud definitions scheme are described there and not in PAPER II.
4.1.2 **FLEXPART Aerosol removal**

FLEXPART has had removal of particles included in the model since early versions. The general outline is that for each time step for each individual trajectory, deposition is calculated based on its proximity to the surface and where it is in relation to any cloud (Fig. 2 left). It is only if the particle is below a reference height (30 m) above the surface, that dry deposition is considered. Wet deposition is considered if there is surface precipitation in the grid of the particle. For precipitating grids both convective and large scale precipitation are considered. In FLEXPART versions preceding v10, the trajectory is considered in a cloud if the relative humidity (RH, see Fig. 2 left) exceeds 80%. Below this value the trajectory is considered to be in cloud free air, which could be either above or below a cloud if any cloud is diagnosed in the model column. From FLEXPART v10, the ability to read the additional 3D cloud fields replaces the RH criteria for defining clouds, and the calculated cloud water content from the numerical weather prediction model is used to define where clouds are.

The ECMWF wind fields, clouds and precipitation used as input are averages over a grid box, which is typically larger than the scale of the pertinent processes. Therefore, a sub-grid parameterisation scheme is used. This scheme is a development of the existing (v6 of FLEXPART) removal scheme (Hertel et al., 1994).

Sub-grid parameterization in FLEXPART considers the surface cloud cover and precipitation type, to scale total precipitation intensity (CP+LSP) by the precipitating fraction of the grid cell (F) in order to obtain a sub grid precipitation intensity as in Stohl et al., (2005). The cloud water is similarly scaled, to only take into account the cloud water fraction of the part of the clouds that is actually precipitating.

The actual removal of aerosol mass, described in PAPER III, subtracts a fraction of the aerosol mass from each trajectory for each time interval ($\Delta t$) according to:

$$m(t+\Delta t) = m(t) - m(t) F (1 - \exp(-\Lambda \Delta t)) \quad (4)$$
Figure 2: Left: Schema of the locations of removal in a grid cell with precipitation in FLEXPART. Right: Determination process of which type of removal is applied in FLEXPART v10. The green box indicates the determination process of the removal scheme in earlier versions of FLEXPART.

Where $m$ is the mass of the aerosol population the trajectory describes, and $\Lambda$ is the scavenging coefficient (s$^{-1}$) for the aerosol in that time step. $\Lambda$ is calculated independently and sequentially for dry and wet deposition. The wet deposition $\Lambda$ is dependent on whether the trajectory is within or below clouds, type of clouds and precipitation type (Fig.2 right).

4.1.3 Sea spray module

An off-line tool (FLEX-SSA) was developed specifically to simulate SSA with FLEXPART. It is written in FORTRAN 95 code and has a library of 53 different sea spray functions available from the literature, including the relations suggested in PAPER I and II. It uses inputs from the ECMWF model. These inputs are the wind speed at 10 m above the surface ($U_{10}$) and the sea surface temperature (SST). Optional inputs are land sea mask (LSM) and sea ice fraction. The FLEX-SSA was also tested with optional inputs using ocean salinity based or satellite derived chlorophyll-a maps used as a proxy of biological activity (Werdell & Bailey, 2005). Global input variables are by default read from ERA-interim with 3 hr temporal resolution, on a 1x1 degree grid (although any data source or resolution would be possible).

Sea spray source functions in the literature are normally given as number size distributions per unit time which in FLEX-SSA are converted to mass size distributions in four size classes up to 10 $\mu$m diameter. Emission fluxes for each size class are then coupled to surface emission sensitivity fields from
FLEXPART backward simulations to estimate concentrations. Alternatively, it can be used to first produce annual emission flux 2D fields, which in turn can be used directly for annual emission flux estimates or as input files for FLEXPART forward simulations.

The SSA module was first implemented in FLEXPART (v9.0), which was not a parallelized model. One major obstacle running forward simulations was the vast number of emission points required for SSA calculations, which resulted in very time consuming simulations. One of the upgrades in v10.0 was the parallelization of the trajectories in FLEXPART so that forward simulation times became less of a problem.

4.2 Measurements

The different aerosol species (SSA, BC, sulfate (radio nuclei) and mineral dust) used in this thesis have different roles in the atmosphere and are best quantified by different properties. The SSA data set used to compare with the model was based on atmospheric filter samples. The analysis of these filters provides information about the chemical composition of the aerosol. For BC aerosol light absorption measurements were used. To measure the blackness of a filter is an indirect method to derive mass concentrations in the atmosphere. To estimate sulphate lifetime in the atmosphere, radionuclides were used. The Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) network of stations provides information about the levels of $^{137}$Cs. The locations of the stations used are presented in Figure 3.
4.2.1 Filter sampling

SSA concentrations are monitored worldwide by various techniques and platforms. Depending on the technique and field of interest, different properties of the aerosols are being measured and comparison between different techniques and experiments is often a difficult task. Global coverage is obtained from space, but has large uncertainties for SSA (Anguelova and Webster 2006). As a choice of measurements to compare model concentration with, filter samples from marine observation sites were used. These are the most straightforward to use as the observations are mass concentrations, just as for the model output. The set of stations provided a good distribution globally.

![Figure 3 Map showing locations of measurement stations used in this thesis. Blue circles show sites of Na+ measurements used to analyse SSA. Black circles show location of measurements of aerosol absorption (σAP) and the larger circles the general area of ship measurement campaigns (Na'). Red circles show the location of the CTBTO measurement network measuring 137Cs.](image)

The different sites use similar high volume filter sampling setups. Air is pulled through an inlet and onto a filter where particles are collected, which are then analysed by ion chromatography. Within individual networks there are standardized sampling protocols, but this does not necessarily exist between different networks. There is no truly global long-term network for these kinds of observations and this makes intercomparison with models on a global scale challenging. The different networks used in this thesis, both to analyse SSA and mineral dust were: Asian dust network in the North Pacific (SEAREX),
Aerosol Oceanic Chemistry Experiment (AEROCE) and American department of energy measurement network. In addition, US National Oceanic and Atmospheric Administration (NOAA) ship campaigns carried out on several oceans, where Na⁺ measured by filter samples were used.

**Na⁺ from ion chromatography**
The Na⁺ measurements used to compare model results are all derived from an ion chromatographical analysis of high volume filter samples. Solvents are extracted from filters using purified water, and then molecules are separated based on their respective charge group. This is a widespread technique to measure inorganic ion concentrations done by several laboratories. SSA concentrations were calculated based on the sodium (Na⁺) concentration of each filter. Mineral dust can be based on assumed aluminium content of the dust or by the weight of insoluble matter remaining after the filter was burnt at 500 degree C. SSA aerosols follow the ocean sodium concentration and consist of about 37% Na⁺ which is consistent over aerosol size distribution with only small variations (Salter et al 2015; Prospero et al 1988).

**Radionuclide based aerosol removal estimates**
CTBTO measures radionuclides at 80 stations, of which 40 also measure noble gases (see ctbto.org for details). Operations of this network started in 1996 to monitor the ban on nuclear test explosions in the atmosphere. Although it was not the original intention, the network did document radioactive debris from accidents in nuclear power plants. Filters samples are collected daily using a high-volume sampler. The daily sampled filters are then analysed for radioactive particles. The gamma spectrum is further analysed to enable the distinction between different radioactive compounds such as $^{137}$Cs.

### 4.2.2 Optical measurements

BC is an inherently complex material which is difficult to measure and typically it is operationally defined depending on the technique used. Therefore, there is no conformity in the terminology which makes it even harder to compare one observation to another (Andreae et al., 2006; Wiedensohler et al., 2016; Petzold et al., 2013). Intercomparison of different optical instrument types shows more than 30 % difference under idealized conditions so a need for uniformity is evident (Müller et al., 2011). In addition, over time BC may transform and the mass specific absorption by particles may be very different away from sources compared to freshly emitted particles.

To reduce the intra technique uncertainty, only stations operating Particle Soot Absorption Photometer (PSAP) instruments were selected. A constant mass
absorption coefficient (MAC) of 10 m²g⁻¹ was used although intercomparisons of European stations show that this value can vary between stations, as well as for individual stations over the year by as much as 70% (Zanatta et al., 2016). For stations where explicit calculations of the stations MAC were available, that coefficient was used and replaced the default value for the MAC. Emphasis was also put on choosing stations at different distances from major sources of BC and in order to get samples that well describe different stages of BC in the atmosphere.

4.2.3 Aerosol size distribution measurements

Both Differential Mobility Particle Sizer (DMPS) and Optical Particle Counters (OPC) were used for measurements utilized in this thesis. The DMPS charges and classifies particles according to their electrical mobility. A near mono-disperse aerosol is selected by the given geometry of the instrument, flow, and voltage of the electrical field (Hinds 2012). The selected particles are counted using a condensation particle counter. By changing the voltage different sizes can be selected, which are then inverted to size distributions. DMPS systems are practical for particles up to about 1 μm in diameter. For particles larger than that OPC’s are more tractable (Hinds 2012). In these instruments the intensity of light scattered by single particles is inverted to a spherical equivalent size based on assumed refractive index of the particle material.
5 Results

5.1 Summary of PAPER I

In PAPER I, a total of 19 different SSA emission parameterizations were tested against the assembled network of observations of atmospheric concentrations. To do this, an offline add-on tool (FLEX-SSA see sect 4.1) for FLEXPART was developed in order to apply generic SSA source functions in FLEXPART. The source functions were selected from a total of more than 50 parameterizations found in the literature. The main goal of the study was to assess differences in the total annual SSA mass produced under varying conditions and locations. As a result from the comparison between observations and different parameterizations, a set of source functions were derived. The new parameterizations tend to yield greater SSA emissions compared to most of the tested functions. The most important reason for making a new source function was for use in future SSA studies using FLEXPART. Another reason was that most previous formulations were based on a single experiment or measurement campaign and not verified on a global scale as is the new source function.

A main objective was to derive new constraints on SSA mass production. Different wind and temperature dependences were evaluated by comparison of model simulations to a set of global coastal measurements. Results obtained using existing source functions were compared to each other, to obtain a reference basis for the variability in estimates of the SSA mass emitted globally. Since the filter measurements used have limited information on aerosol size distribution, an estimate for this was obtained by a fit to existing source functions.

The main findings were:

- More than 80% of the SSA produced globally are at wind speeds between 5-14 ms\(^{-1}\).
- Emission strength of individual source functions commonly used in models have emissions that range between \(\sim\)2-20 Pg yr\(^{-1}\).
• Best fit to observations were found using a source function with annual mass emitted (9 Pg yr\(^{-1}\)) and a wind speed dependence of U_{10}^{3.5}.
• Data also suggested that SSA mass production increased with increasing SST temperature.

### 5.2 Summary of PAPER II

A controlled laboratory experiment was performed using a tank with artificial sea water to under carefully controlled conditions, to study the effect on SSA production as a function of water temperature. A plunging jet was used to produce bubbles in a tank. The particle size distribution produced by the bursting bubbles was measured using two different particle counters.

A fit was made to the observed aerosol size distribution produced in the tank. This resulted in a new parameterization that also takes into account that the aerosol size distribution is influenced by temperature. FLEX-SSA was then used to assess concentrations from the newly derived source function against observations within the same framework as in PAPER I. The new source function was also implemented in the earth system model (NorESM) to assess the effect of the new source term on the global distribution of SSA.

The main findings were:

• The new source function produces 5.9 Pg yr\(^{-1}\) of SSA, with about the same prediction skill compared to observations as the source function in PAPER I. The 40% lower mass results in an approximate similar underestimation of mass (of 40%) when compared to surface stations.
• Increasing water temperature from a minimum of 2°C to 30°C lead to a shift to larger sizes in the aerosol size distribution.
• Excepting high latitudes, a lower SSA column burden was found than that of the previous parameterization.
5.3 Summary of PAPER III

This paper describes the implementation of an aerosol wet removal scheme in FLEXPART. The new option to use information about cloud water content from the ECMWF data to define clouds in FLEXPART is presented. In both PAPER I & II, it was found that FLEXPART had a somewhat lower prediction skill when compared to observations at high latitudes. Earlier studies with BC aerosols in FLEXPART had displayed a similar tendency by under predicting BC concentrations in the Arctic. Together with the addition of important high latitude sources, the role of BC removal was investigated. Also atmospheric e-folding lifetimes and concentrations of mineral dust and sulfate aerosols were evaluated so that the new aerosol removal scheme was tested for a wide range of aerosol types.

The main features of this scheme were:

- Prescribed clouds from ECMWF, IFS cloud water data that place clouds realistically to precipitation.
- Below cloud scavenging considers both precipitation type and particle size. The in cloud removal considers the cloud water phase.
- Different types of aerosol qualities such as size and solubility were accommodated.
- Atmospheric e-folding lifetimes depend on particle size and type, but range typically from a few hours (particles d >6μm), up to 32 days for the longest lived accumulation mode particles.

5.4 Summary of PAPER IV

In PAPER IV, long term measurements of aerosol light absorption from Dome C, Antarctica are presented. Together with statistical characteristics of the measurements a detailed analysis of the origin and pathways of aerosols are presented.

The main findings of this study were:

- Overall, the levels of surface measurement of aerosol absorption and scattering from the Antarctic inland are extremely low compared to other surface measurements.
A strong seasonality with increased austral summertime aerosol absorption.

Periodic strong covariation to South Pole measurements indicating a large-scale phenomenon causing variations on a scale of weeks.

Direct transport from other continents is very rare and the aerosol transported through the stratosphere seems to provide the most direct link.

5.5 Summary of PAPER V

Emissions from ships are only a small fraction of the global anthropogenic emissions, they can however be significant because collectively, ships are a diffuse source of pollutants, often operating and emitting pollutants in otherwise pristine areas. The effect of increased cruise ship traffic was investigated near the remote research station Ny-Ålesund on Svalbard in PAPER V.

The main findings of this study were:

- Local ship traffic periodically influences the measurements at Ny-Ålesund significantly.
- With the predicted future increase in ship traffic in the Arctic, measures to reduce ship emissions should be considered.
6 Main conclusions

This thesis is comprised of five papers dealing with several of the important aerosol processes that determine the atmospheric aerosol life cycle and distribution. In succession the papers have dealt with aerosol sources, removal, lifetime and concentrations. In this chapter, broader conclusions based on the results of the individual papers are drawn. The conclusions are synthesized and organized along the main objectives listed early in the thesis in Chapter 3.

1) Evaluate the relative importance of the different physical parameters in existing parameterizations that are considered important for emission of SSA.

Of special interest to this thesis is the SSA source term and potential sensitivity to changing environmental conditions. PAPERS I & II research these topics. Both papers consider solely the emission of SSA inorganic mass. The findings in PAPER I showed that the relative contribution to the source term uncertainty (with respect to mass) from oceanic salinity differences are negligible compared to even small changes in the wind speed. A significant contribution of PAPER I is an evaluation of the frequently used SSA source function for modelling purposes. There are vast differences between source terms, source functions ranging from 1.8 to 600 Pg yr⁻¹. The sea spray module developed represents a novel approach and is a promising tool and reference for future development of new and even more accurate SSA source functions. Throughout PAPER II several discoveries were made that highlighted the advantage of fully scaling up individual laboratory results for implementation in models. The derived SSA source function in PAPER II is to our knowledge the most detailed inorganic SSA function implemented in a fully coupled Earth System Model (NorESM). It includes a temperature dependent and particle size resolved emissions. It is highlighted that the SSA source term is important in determining both aerosol direct and indirect effects.

2) Develop an aerosol property specific wet removal scheme for FLEXPART and assess model accuracy in simulated long range transport of pollutants.

The research with this objective is presented in PAPER III. Here the implementation of the new aerosol removal scheme is described. The properties of the new aerosol removal scheme were thoroughly tested for aerosol particles.
both solid and liquid soluble ranging from accumulation mode to 20 μm. As expected, the difference between the new and old scheme was seen most strongly on the e-folding lifetimes of accumulation mode aerosols. The new scheme resulted in an overall longer model residence times for particles in the atmosphere.

For accumulation mode aerosol particles in the free troposphere there is no efficient removal other than through nucleation of cloud droplets, and therefore the lifetime of these particles are the most sensitive to in-cloud removal. A clear illustration of this is accumulation mode dust aerosols which were found to have very long e-folding times, above 32 days in FLEXPART, due to their inefficiency to act as CCN. Somewhat lower e-folding lifetimes were found for BC (16 days) and especially sulphate (10 days). These e-folding timescales are representative of lifetime after adequate mixing in the atmosphere. A result of the increased lifetime is substantial increased concentrations in remote regions for all of the three aerosol types examined.

The new wet-removal scheme will however have little influence on the SSA results obtained in PAPERS I and II. The main focus in these papers was on the SSA source strength and only coastal stations were used for comparison. Therefore, removal processes had little time to influence the observed SSA concentrations. The new scheme may have larger impact on receptor areas further away from source regions, but this was not specifically tested for SSA.

The wet removal module for FLEXPART presented in this thesis is state-of-the-art in the field of transport modelling. It fulfils all requirements to accurately represent wet removal for the diversity of aerosol particles where FLEXPART is currently applied. This is demonstrated by thorough validation and testing against observations. Another achievement of this work is the increased level of accuracy in clouds obtained by using cloud water content from prescribed clouds. This improves the definition and description of clouds in FLEXPART, and will benefit all future applications of the model.

3) Assess how aerosol concentrations in the Arctic and Antarctic are different, with respect to sources and transport paths. In particular, characterise the optical properties of aerosols in the Antarctic interior.

The work in PAPER IV is an important addition to our understanding how anthropogenic aerosol sources in the Arctic and Antarctic differ. In addition, the temperature dependence in the SSA source functions also contributes towards increasing our understanding of natural aerosol sources. However, a significant fraction of the aerosols in the Arctic atmosphere is the result of
long range transport. In PAPER III it was demonstrated that exaggerated removal rates of aerosol particles contributed to FLEXPART underestimating aerosol concentrations at high latitudes. In PAPER V a unique dataset from the Antarctic interior was analysed. Analysis of atmospheric aerosol optical properties showed strong seasonal cycle in single scattering albedo, similar to the one found in the Arctic. This has, to our knowledge, not been reported for inland Antarctica previously. Air mass transport patterns analysis showed that both transport time and aerosol removal processes are likely responsible for the observed seasonality in aerosol properties of the Antarctic. Combined the findings of PAPER IV and V highlight the important aspect of emission and removal to high latitude aerosol populations.

This thesis covers aerosols from their largest sources with high concentrations to remote locations, such as Antarctica, with the lowest concentrations. BC concentrations in the Antarctic inland were found to be near the detection limit of instruments. The long distance between sources of BC and Antarctica means that transport and removal processes are even more important for this region than for the Arctic. FLEXPART simulations show that the BC aerosols observed in the Antarctic inland often have travelled more than 30 days from the sources. Near sources, atmospheric concentrations of aerosol particles are to a large degree controlled by the source strengths. As the particles travel further away from the sources, details in dispersion patterns and removal processes become increasingly important. Although, each phase of the aerosol life cycle is associated with its own uncertainties, the processes are difficult to isolate from the complex complete system. Nonetheless, this thesis has addressed key processes involving sources, transport, and ultimately removal of atmospheric aerosols. A large range of scales had to be considered, from laboratory tank experiments, to estimates of global emissions, as well as transport patterns covering both Polar Regions.
7 Outlook

Several findings in this thesis suggest directions for further research and development. All simulations of SSA were done in backwards mode. To further reduce the source term uncertainty there is more untapped information available that can be coupled to the emission sensitivity grid from these simulations. Future improvements can include more observational details, such as accurate SSA size distribution from field stations or ocean biological activity. Both could provide more evidence of other yet unknown processes. Ocean wave properties and atmospheric stability in the source region are also important parameters and should be included in future studies.

Another approach is to increase the spatial and temporal resolution by applying an inversion algorithm on a fine model grid to obtain a more accurate emission estimate for SSA. An improved resolution of the model domain may be more appropriate for laboratory derived source functions, which are very detailed compared to those derived from bulk observations. The possibility to include greater details may help bridge the gap between laboratory based source function and those derived with the use of the FLEXPART model. This is of particular interest with respect to the effect on emissions from variation in sea surface temperature.

To be able to address the research objectives, the most demanding task of this thesis was the development of a new removal scheme for aerosols in FLEXPART. While there are limitations on the complexity of the processes that can be described in a Lagrangian model, the current state-of-the-art knowledge clearly shows that more aspects of aerosol-cloud interactions should be incorporated. Specifically, aerosol accumulation by entering clouds that do not necessarily precipitate is a feasible feature. Further improvement can possibly also be achieved by coupling the removal scheme to the scheme for convective motion so that particles moving up in the stochastic scheme of vertical motion by the convective scheme are linked to the removal.

Since 2012 an Automatic Identification System (AIS) satellite has gathered detailed information on global ship movements, with 6 digit accurate information on ship position, and vessel engine information. Data is available for the entire global merchant shipping fleet required to use AIS transmitters. Combined with detailed information about ship and engine characteristics, a
bottom up emission inventory has been created with unprecedented detail and accuracy for all ship emissions. Having access to this constricted data set has already presented exciting opportunities and resulted in several other publications (Kivekäs et al., 2014; Buixade et al., 2014). In addition, work on lake sediment sources (Ruppel et al., 2013) and mineral dust emissions (Groot Zwaaftink et al., 2016), provide a broad base of understanding aerosol emissions. These publications has already provided insights to the effect of global ship emissions on BC, sulphate, mineral dust and gases in near and remote areas and their consequences, and the knowhow of all these emission data will hopefully present interesting perspectives in the future.
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9 References


