AEROSOL PROCESSES IN POLAR REGIONS — FROM THE FORMATION TO CLIMATIC IMPLICATIONS

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Academic dissertation
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Abstract

Atmospheric aerosols affect our health, air quality, visibility and climate. They can impact the climate through their ability to interact with radiation and to alter cloud properties by acting as cloud condensation nuclei (CCN) or ice nuclei (IN). Globally, aerosols cool the climate, but locally their effect may be opposite. Their climatic effects are determined by their concentration, size distribution and chemical composition as well as their vertical and spatial distribution and the underlying surface type. Currently, the largest uncertainties in estimating our future climate are related to atmospheric aerosols and their interactions with climate.

Polar regions are experiencing faster warming than the Earth on average. This enhanced warming leads to many dramatic changes in the cryosphere, including rapid shrinkage of Arctic summer sea ice. Arctic amplification also decreases the temperature gradient between the Arctic and polar air masses. Both of these changes feed back to the atmospheric dynamics and thus the transport of pollutants into the Arctic. The rapid climate change alters also the sources — both natural and anthropogenic — and sinks of secondary aerosols in polar regions. Therefore, it is crucial to understand the formation and growth mechanisms of atmospheric aerosols in these areas in order to assess their climatic effects. High latitudes also offer a great natural laboratory to study the aerosol dynamics and timescales for reaching climatically relevant sizes or obtaining a balance between sources and sinks, with very little anthropogenic influence. Moreover, as the precipitation amount and patterns will change in the future, the removal of aerosols is also subject to change. Quantifying this requires parameterization for climate models.

This thesis adds to the understanding of all of these aforementioned parts in the aerosol processes and their climatic effects in polar regions. It offers the first observations of Antarctic new particle formation (NPF) from continental biogenic precursors and shows that areas with melt water ponds over glaciers and continental ice sheets are important regions for the formation of secondary aerosols and the organics evaporating from such ponds have the potential to grow the particles up to climatically relevant sizes even in timescales of only few hours. In this thesis, it is also shown that in areas with low background aerosol concentrations, large sources of anthropogenic sulphur have a substantial impact to the trends in NPF and potential CCN in the scale of few hundreds of kilometers. This thesis also introduces a new way to study the evolution of aerosol number size distribution during air mass transport and shows that the aerosol condensational growth is markable even in the absence of evident NPF. Finally, this thesis offers the first parameterization of snow scavenging in a way that is easily applicable to climate models.

Keywords: new particle formation, aerosol growth, aerosol dynamics, aerosol-cloud-climate interactions, aerosol removal, Arctic, Antarctic
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# Nomenclature

Some of the _italicized_ terms that appear in this thesis explained in short:

<table>
<thead>
<tr>
<th>Term</th>
<th>Explanation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Activation-type nucleation</td>
<td>Nucleation of an aerosol particle through activation of a cluster by a vapour molecule, typically participating vapour is sulphuric acid</td>
</tr>
<tr>
<td>Arctic amplification</td>
<td>Enhanced Arctic warming compared to lower latitudes</td>
</tr>
<tr>
<td>Cloud processing</td>
<td>Transformation (in the physical and microphysical characteristics) of aerosol population through acting as seeds for clouds</td>
</tr>
<tr>
<td>Dry deposition</td>
<td>Scavenging of aerosols from the atmosphere without the involvement of water</td>
</tr>
<tr>
<td>Nucleation rate</td>
<td>The rate at which new particles are formed by nucleation, unit $[\text{cm}^{-3}\text{s}^{-1}]$</td>
</tr>
<tr>
<td>Greenfield gap</td>
<td>A minimum in the below-cloud scavenging efficiency, typically for around 100 nm — 1 $\mu$m sized particles, due to less efficient removal mechanisms in this size region</td>
</tr>
<tr>
<td>Growth rate</td>
<td>The rate at which aerosol particles grow from one size to another, unit $[\text{nmh}^{-1}]$</td>
</tr>
<tr>
<td>Kinetic-type nucleation</td>
<td>Nucleation of an aerosol particle through two molecules colliding with each other, typically participating vapour is sulphuric acid</td>
</tr>
<tr>
<td>Primary (aerosol) particles</td>
<td>Aerosol particles that are introduced into the atmosphere in the particulate phase</td>
</tr>
<tr>
<td>Secondary (aerosol) particles</td>
<td>Aerosol particles that are formed in the atmosphere through new particle formation</td>
</tr>
<tr>
<td>Stratospheric fault</td>
<td>Air descending from the stratosphere down to the (lower parts of) troposphere</td>
</tr>
<tr>
<td>Trajectory</td>
<td>The path along which an airmass travels, can be calculated backward or forward in time</td>
</tr>
<tr>
<td>Wet deposition</td>
<td>Scavenging of aerosols from the atmosphere by (liquid or solid) water: clouds, fog or rain</td>
</tr>
</tbody>
</table>
List of publications

This thesis consists of an introductory review, followed by 4 research articles. In the introductory part, these papers are cited according to their roman numerals. Papers I-III are reproduced under the Creative Commons Licence. Paper IV is reproduced with the permission granted by the Boreal Environment Research Publishing Board.


1 Introduction

Aerosol particles are very small, liquid or solid particles that are suspended in a gas, in our case the atmosphere. Their sizes range several orders of magnitude – from nanometers \(10^{-9}\) m up to some hundreds of micrometers \(1 \mu\text{m} = 10^{-6}\) m. Thus, a big fraction of them are so small that they are not affected almost at all by gravitational settling. Their concentration in the atmosphere also ranges widely. In very clean, winter Antarctic atmosphere the concentration of submicron aerosols is usually of the order of \(1 - 10\) cm\(^{-3}\) (Järvinen et al., 2013), whereas in heavily polluted cities their concentration can reach hundreds of thousands particles in cm\(^3\) (Mönkkönen et al., 2005). Aerosol scientists are often interested especially on the aerosol concentration as well as their number, surface or volume size distributions since these can tell a lot about the effects the aerosols may have. Typically, very small particles, less than 100 nm in diameter, dominate the number size distribution whereas most of the aerosol mass is in the upper size range (particles larger than 100 – 500 nm).

Aerosol particles have adverse effects on our health. It is estimated that by 2020, even up to 250000 lives are lost annually within EU due to anthropogenic particulate matter (Holland et al., 2005). In addition to their health effects, aerosols can also reduce visibility. In Antarctica where the aerosol number concentration is very low, the visibility can reach several hundreds of kilometers (Figure 1a). An example of greatly reduced visibility due to additional anthropogenic aerosols is the well-known London smog or the hovering cloud of pollution over the present-day megacities. The same phenomena is shown in Figure 1b above a Swiss town surrounded by the Alps during a temperature inversion which prevents mixing of the air and blocks the aerosol close to the ground.

The least understood part of the aerosol effects are their ability to interact with the climate trough various ways. They can either directly scatter or absorb solar radiation or then affect the climate trough changing cloud properties. The latter part is less known and more difficult to assess. Despite their negative effects on health and visibility, aerosol net effect on climate is cooling (IPCC, 2013). The radiative forcing (RF, a measure of the imbalance in the Earth’s energy budget) caused by different aerosol species interacting with radiation is estimated to be \(-0.27\) Wm\(^{-2}\), lower and upper estimate being \(-0.77\) Wm\(^{-2}\) and \(0.23\) Wm\(^{-2}\), respectively. The RF from cloud adjustments due to aerosols is even more negative, and is estimated to be \(-0.55\) Wm\(^{-2}\) \((-1.33\) Wm\(^{-2}\) to \(-0.06\) Wm\(^{-2}\)) (IPCC, 2013). The uncertainties in the estimations are, however, very large, yet the values are markable compared to the total anthropogenic RF relative to 1750 which lies somewhere between \(+1.13\ldots + 3.33\) Wm\(^{-2}\), with the best estimate being \(+2.29\) Wm\(^{-2}\) (IPCC, 2013). The ability of aerosols to cool the climate but on the other hand their harmful nature poses a difficult ethical and political dilemma (Arnett et al., 2009): How much should we, as a humankind, reduce pollution either in the form of aerosols or their precursors, to find a balance between the casualties caused by...
Figure 1: Examples of aerosol effects on visibility and climate.
the pollution and on the other hand, by the natural disasters and extreme phenomena caused by increasing global temperature?

He effects that aerosols have on our health, visibility and climate depend on their concentration and number size distribution as well as their chemical composition. These in turn are mostly determined by the origin and age of aerosol particles. Aerosol particles can be divided into primary and secondary aerosols (Seinfeld and Pandis, 2006). Primary particles enter the atmosphere in a particulate phase, whereas secondary aerosols are formed from precursor gases in the atmosphere through new particle formation. Sources of primary and secondary aerosols are usually very different. The majority of primary particles are anthropogenic, originating e.g. from combustion processes or when car tires are grinding the road. Some of the primary particles are also natural, like desert dust, sea salt, pollen or bacteria. Primary particles are usually rather large, and settle down quickly. However e.g. desert dust can travel thousands of kilometers reducing visibility at the surface and scattering sunlight back to the space. From the climatic perspective, secondary aerosols are more important since they are more likely to affect the climate but their formation mechanisms are still not completely known. However, after their formation they are still too small to affect the climate but they need to grow to larger sizes. The precursor gases that initiate the new particle formation are typically anthropogenic, most important being sulphuric acid ($H_2SO_4$), and abundant in the atmosphere. However, also natural biogenic emissions take part in the formation and growth of secondary aerosols and increasing global temperature enhances those emissions (Kulmala et al., 2004a; Paasonen et al., 2013).

The ability of aerosol particles to act as cloud condensation nuclei (CCN) contribute the largest uncertainty compared with other anthropogenic forcers to estimating the future climate. On average, clouds are cooling the atmosphere by reflecting solar radiation back to space. An increase in the aerosol number concentration has the potential to increase the cloud reflectance and lifetime trough increased cloud droplet number concentration, and to decrease the precipitation intensity by decreasing the size of cloud droplets inside the cloud. The feedback mechanisms related to aerosols and clouds are, however, complicated and may under certain circumstances even cause a warming effect on climate. For example, contrails (Fig. 1c) can, if the upper atmosphere is humid enough, be spread out and form Cirrus or Cirrostratus clouds. Such clouds are typically very thin and the solar radiation penetrates trough them, whereas Earth’s thermal radiation is reflected back to the troposphere, thus warming the atmosphere. This effect is globally almost insignificant (IPCC, 2013), but locally it may have a large radiative forcing (Burkhardt and Kärcher, 2011). Also, when clouds cover already reflective surfaces, such as glaciers or sea ice, or when they are over areas with little or no solar radiation e.g. in the Arctic winter, their cooling effect may be dampened or they may even warm the surface (Mauritsen et al., 2011; Morrison et al., 2012; IPCC, 2013). Overall, net effect of clouds on the atmosphere is cooling (IPCC, 2013) even if they warm
Why is it important to do aerosol research particularly in polar regions when even getting there is already a challenge, let alone conducting measurements in those environments? Research in polar regions requires thorough and long-term planning and experienced people who are prepared for all kinds of unexpected incidents that they might face when doing measurements in harsh conditions. The fierce weather conditions of the Arctic and Antarctic pose a challenge for both instruments and expedition members themselves. Bringing instrumentation and people to these conditions also costs more than measuring in easier conditions due to challenging logistics. All these things in mind, it is not surprising that we know much less about the aerosol sources and processes in the polar areas than elsewhere. Yet, there are several reasons why more research should be conducted in these inhospitable areas of our planet.

Firstly, high latitudes are experiencing more rapid climate change than other regions in our planet. In terms of atmospheric temperature, Arctic is warming twice the rate of the global average (Shindell and Faluvegi, 2009; IPCC, 2013). This enhanced warming compared to the lower latitudes is often referred to as the Arctic amplification. Also parts of Antarctica are warming faster than the global average (Steig et al., 2009; Schneider et al., 2012). The increased warming leads to many dramatic changes in the cryosphere, such as melting of glaciers and ice sheets, ice shelf collapses, shorter snow cover duration, shorter ice-cover duration in lakes and rivers and permafrost melt (Quale et al., 2002; Rignot et al., 2004; Lawrence et al., 2008; Chen et al., 2009; Pritchard et al., 2009; Screen and Simmonds, 2010; AMAP, 2012; Jacob et al., 2012). One of the most important changes with respect to the responses in atmospheric dynamics and thus atmospheric transport to and from Arctic is the fast disappearance (Riihelä et al., 2013) of the white ice cap of the Arctic — namely the ice cover of the Arctic Ocean which has been decreasing even faster than the models have predicted (Stroeve et al., 2007; Boé et al., 2009; Wang and Overland, 2009; Stroeve et al., 2012). Recent studies have consistently shown that the shrinking Arctic summer sea ice cover is linked with the recent cold and snowy winters in Europe (Balmaseda et al., 2010; Cohen et al., 2010; L’Heureux et al., 2010; Outten and Esau, 2012; Screen et al., 2012; Yang and Christensen, 2012; Tang et al., 2013). This is primarily due to increased heat and water vapour fluxes from the open ocean, which in turn results in earlier autumn snow cover in Siberia (Allen and Zender, 2010; Cohen et al., 2012; Inoue et al, 2012). The earlier snow cover then intensifies the high pressure over Siberia and thus brings cold continental Arctic air mass to Europe. The intensified heat and water vapour fluxes can also alter the polar vortex by either overturning it or slowing it down, and thus allowing the cold Arctic air mass to spread southwards (Thompson et al., 2002; Overland and Wang, 2010; Screen et al., 2012).

Arctic amplification also decreases the temperature gradient between Arctic and polar air
Since this temperature gradient ($\nabla T$) is essentially the driving force for polar jet stream and thus cyclonic activity, decreasing $\nabla T$ weakens the jet stream and leads it to meander more in north-south direction (Francis and Vavrus, 2012; Screen and Simmonds, 2013), thus increasing the probability for atmospheric blockings. Since low-pressure systems are the most efficient atmospheric transport method from mid-latitudes to the poles, changes in the jet stream positioning will alter the atmospheric transport routes for pollutants. Also, in case of atmospheric blockings, cyclones need to pass the blocking high pressures either from north or south. For North Atlantic and Europe, this means intensified atmospheric transport into the Arctic (Inoue et al, 2012). Both Arctic and Antarctic are very pristine areas and thus very sensitive to anthropogenic pollution. Thus, changes in the atmospheric transport routes will also likely affect the fragile ecosystems if more pollution reaches those regions.

The rapid climate change in the Arctic and Antarctica presumably changes the sources and sinks of primary and secondary aerosols also within the polar regions. Primary aerosol concentration is prone to increase in polar regions due to changes in sea spray concentration (Struthers et al., 2011) and biogenic feedback mechanisms related to increasing temperature (Paasonen et al., 2013). Possible changes in biogenic sources for secondary aerosols are discussed in Paper I, whereas Paper II focuses on long-term changes in the anthropogenic sources of secondary aerosols. These studies show that in the changing climate, it is expected that both natural and anthropogenic sources for secondary aerosols will increase in polar regions as larger areas become exposed and more meltwater forms, and also maritime activities increase in the Arctic Ocean. A more thorough evaluation of the relevance of the studies in this thesis in climatic perspective is given in section 4.5.

Finally, due to very limited anthropogenic influence especially in continental Antarctica, polar regions offer a great natural laboratory to study the actual physical processes that take place in atmospheric new particle formation and growth. It is possible to investigate e.g. the timescales that are required for aerosols to grow into climatically relevant sizes in the absence of large anthropogenic pollution or even major vegetation, such as boreal forests, that emit vast amounts of condensable vapours. Also, comparing the observations with those measured at rural vegetated areas or in urban areas can give more detailed information of the importance of vegetation or anthropogenic factors to NPF mechanisms.

Broadly, the objectives of this thesis were to understand the aerosol phenomenology in polar regions, including new particle formation (NPF) in different environments, aerosol growth, aerosol-cloud-climate interactions and aerosol removal. We wanted to gain a better understanding of the NPF in polar regions as well as under which circumstances and how rapidly they can grow to sizes where they can act as cloud condensation nuclei (CCN). All this was done in order to establish a better knowledge of how the sources and sinks for climatically relevant particles will change in the changing polar climate. More elaboratively,
the objectives of this thesis can be divided into three categories, all from polar perspective:

i  to identify the possible changes in biogenic and anthropogenic aerosol precursors in the changing climate and their cloud forming potential (Paper I & Paper II),

ii to understand the aerosol dynamical processes that evolve the aerosol size distribution during transport over land, and how long it takes to establish a balance between source and sink processes (Paper III), and

iii to quantify the aerosol removal by snow and make a simple parameterization that can be used for modelling purposes (Paper IV).
2 Aerosol processes

Figure 2: An example of a new particle formation event, observed at Aboa on 17th-18th January, 2010, and a schematic figure illustrating the aerosol processes from the formation to the climatic effects. Aerosols are formed (region II) from constantly existing pool of clusters (region I). In region III, constant competition between growth by condensation and losses by coagulation (and deposition in the upper end of this region) takes place. Finally, when particles have reached the region IV, they can affect the climate trough acting as seeds for clouds and after growing up to around 400 nm, by interacting with solar radiation. Regions I, II and partly III are adapted from Kulmala et al. (2013). The colour scale represents $dN/d\log D_p$.

There are wide range of aerosol processes which form, age and modify the aerosol population. These include the nucleation of secondary aerosols, which is often referred to as the new particle formation (NPF), particle growth by condensation, coagulation acts mainly as a sink, and finally, the removal of aerosols from the atmosphere by deposition. The aerosol phenomenology also includes aerosol-cloud-climate interactions which include a wide range of different types of processes, many of which are still poorly understood. From NPF point of view, we can distinguish four different regions of interest from the aerosol number size distribution, all of which have their characteristic processes taking place in that size range (Fig. 2):
Region I: This region consists of small clusters and molecules that are present in the atmosphere all the time and do not have direct connection to NPF. In this region, clusters are formed by vapour uptake and lost by evaporation continuously and the growth is very slow (Kulmala et al., 2013). The upper limit of this region is $1.1 - 1.3 \text{nm}$. 

Region II: In this region new particles are formed, when the clusters grow slowly by sulphuric acid condensation and are stabilized by e.g. amines, ammonia or organic vapours. New particle formation is explained in Chapter 2.1 in more detail.

Region III: This region covers particle sizes from $1.3 - 1.9 \text{nm}$ up to around $50 - 100 \text{nm}$. Particles larger than $1.5 - 1.9 \text{nm}$ grow fast and sulphuric acid condensation can no longer explain the observed growth. Majority of the growth in the lower size range of this region is most likely caused by oxidized organic vapour uptake. Essentially, the lower end of this region is existing only during active aerosol formation. Chapter 2.2 deals with the processes related to this region. The relationship between growth and loss through coagulation determines the fraction of particles reaching the region IV.

Region IV: Finally, after growth, particles may reach climatically relevant sizes. In this region the source and sink processes are balanced and the particles do not grow significantly anymore. Rather, they can be scavenged from the atmosphere e.g. by acting as cloud condensation nuclei (CCN). Processes characteristic to this region are discussed in Chapters 2.3 and 2.4.

More traditionally, the size distribution of secondary aerosols can be divided into different modes that are a result of the formation, growth and removal processes: cluster mode, nucleation mode, Aitken mode and accumulation mode. Cluster mode consists of atmospheric clusters less than $2 \text{nm}$ in size (Kulmala et al., 2013). Typically nucleation mode is defined as aerosols with diameter $d_p$ below $20 \text{nm}$, Aitken mode as particles between $20 - 90 \text{nm}$ and accumulation mode ranging from $90 \text{nm}$ to $1 \mu\text{m}$ (Kulmala et al., 2004b; Seinfeld and Pandis, 2006). A fifth mode, coarse mode (particles larger than $1 \mu\text{m}$ in diameter, Seinfeld and Pandis, 2006), consists only of primary particles (Hinds, 1999).

2.1 New particle formation

In the atmosphere, small clusters are formed by molecular collisions and chemical reactions and lost by evaporation constantly (Kulmala et al., 2013). These clusters are very small, around $1 \text{nm}$ in size, and form a pool which is present all the time in the atmosphere. Their sizes, however, cannot be determined unequivocally since they are composed of only few molecules. The existence of these clusters was predicted already in the beginning of the
21st century (Kulmala et al., 2000) and the first observations were made some years later (Kulmala et al., 2007).

Under certain conditions, these clusters can start to grow. This step is called atmospheric nucleation, or new particle formation. NPF is a frequent phenomena and has been observed almost everywhere in the world (Kulmala et al., 2004b; Vana et al., 2008; Boulon et al., 2010; Manninen et al., 2010; Vakkari et al., 2011; Papers I, II, III). In the recent years, the nucleation research has taken giant leaps forward (Kulmala et al., 2013, 2014). Although there are still open questions, the mechanisms and chemistry behind the very initial steps of nucleation have started to dawn on aerosol scientists. The single most important atmospheric constituent with respect to NPF has been known already for a while to be sulphuric acid (\(H_2SO_4\)) (Weber et al., 1995; Kulmala et al., 2006; Sipilä et al., 2010), yet it cannot continue growing all by itself (Kurtén et al., 2008). Recently, with the help of new instrumentation, also other compounds, such as ammonia, amines or low-volatile organics, have been identified by laboratory and field experiments to have a major role in the nucleation process in both stabilizing and activating the clusters and thus allowing their growth (Kirkby et al., 2011; Petäjä et al., 2011; Jokinen et al., 2012; Almeida et al., 2013; Schobesberger et al., 2013; Kulmala et al., 2013, 2014). Altogether, common for all of the compounds taking part in the formation of secondary aerosols, is that either the volatility of these vapours needs to be low or they need to be able to facilitate the formation of low-volatile complexes (Kulmala et al., 2014). E.g. amines and ammonia are both volatile as themselves but together with acids they can form low-volatile acid-base complexes. Atmospheric oxidants are important in this respect, since they produce both \(H_2SO_4\) trough various pathways and on the other hand, precursors for low-volatile organic compounds. Generally, all of these reactions need light to be initiated.

Several nucleation mechanisms have been proposed. Since the ratio between the formation rate of newly formed particles is usually to the power of \(1 - 2\) of the sulphuric acid concentration (Kulmala et al., 2006; Sihto et al., 2006; Riihimäki et al., 2007; Paasonen et al., 2010), the most investigated and commonly assumed nucleation mechanisms are the kinetic (Eq. 1) (McMurry and Friedlander, 1979) and activation-type (Eq. 2) (Kulmala et al., 2006) nucleation involving \(H_2SO_4\):

\[
J = k[H_2SO_4]^2
\]  
\[J = A[H_2SO_4]\]

Here \(J\) is the nucleation rate, \(k\) is kinetic coefficient including both the collision frequency and the probability of formation of stable particles after the collision and \(A\) is the activation
coefficient. In areas with high sulphur pollution, the power in the relation between \( J \) and \( H_2SO_4 \) concentration is usually closer to 2 (Kuang et al., 2008), suggesting that in these environments, kinetic nucleation mechanism is more dominant. Other nucleation mechanisms have been proposed, including those affected by organic vapours (Paasonen et al., 2010) and following \( e.g. \) the relation

\[
J = k_1[H_2SO_4]^2 + k_2[H_2SO_4] \cdot [\text{Org}]
\]  

where \( \text{Org} \) is the concentration of first order products from monoterpene oxidation, has been shown to correlate well with various environments (Paasonen et al., 2010).

Ions are very abundant in the atmosphere and they also contribute to NPF by increasing nucleation and formation rates (Duplissy et al., 2010; Kirkby et al., 2011) under favourable conditions. The effect of solar activity to global climate via this pathway has been studied to a great extent. Ion-induced and ion-mediated nucleation are especially thought to be important in the upper and mid-troposphere (Yu and Luo, 2010). Measurements \( e.g. \) in Antarctica suggest that ions may contribute significantly to aerosol formation rates in some lower tropospheric environments as well (Asmi et al., 2010).

Regardless of the nucleation mechanism, after certain size (usually around 1.7 ± 0.2 nm), the availability of sulphuric acid cannot explain the cluster growth anymore but the particles grow by multicomponent condensational growth (Kulmala et al., 2014). From this on, the growth rate \( (GR) \) of the nanoparticles tends to increase with increasing particle size (Kulmala et al., 2013). This increased \( GR \) due to decreasing Kelvin effect is called the Nano-Köhler theory (Kulmala et al., 2004c).

"Bananas" and "Apples": The visual classification of NPF events

There exist several — more and less detailed — classification systems for NPF events in literature, nearly all of which however are based on the visual shape of the surface plot depicting the time-evolution of the aerosol size distribution during the aerosol formation. Due to the Eulerian way of measuring the air mass that is moving, the shape of the NPF event is a signature of the temporal and spatial scales and intensity of the event, combined with the state of the air mass (Fig. 3).

The basic classification into non-event, undefined and event days (Class I or Class II) was first introduced by Dal Maso et al. (2005). This classification was later expanded when new instruments capable of measuring ions down to \( \sim 1 - 2 \) nm were introduced (Hirsikko et al., 2007), enabling NPF with small temporal and spatial scales to be visible in the size
spectrum. A more detailed criteria for classifying the event days is shown e.g. in Yli-Juuti et al. (2009) and Manninen et al. (2010). Recently, Kulmala et al. (2012) proposed a protocol for identifying NPF events, combining all the steps from the adequate measurements to detailed analysis of NPF event days.

In general, days can be classified as event or non-event days depending whether new particle formation is observable or not. More specifically, on an event day, a new growing sub-25 nm particle mode, lasting for at least an hour, is observed. If the formation of new particles is ambiguous, that is, a new mode below 25 nm appears but does not grow or if a mode above 25 nm is seen to grow, the day can be classified as an undefined day. To classify the event days further, one can divide them into Class I or Class II depending on whether it is possible to determine both formation and growth rates of freshly nucleated particles from the aerosol number size distribution (Class I) or not (Class II). Sometimes the Class I events can be further divided into classes Ia and Ib, depending on the smoothness of the event: compared to Class Ib, Class Ia events can be used for modeling case studies due to their clear, distinguished shape.

The traditional ”banana” events are observed when NPF takes place over large areas and last for long time (from several hours to days) in a relatively homogenous air mass. The event appears in the size distribution as a wide band starting from the lower limit of the size distribution and then curving smoothly towards the larger sizes (Fig. 3). Such an event is shown in Fig. 2. These events may be called as regional events. If, on the other hand, the formation of new particles takes place locally, it appears as a round- or obscure-shaped short-time increase in the particle concentration in the lowest particle sizes, and can be called — depending on the shape — as ”apple” or ”hump” (sometimes referred to as ”hump”). Events can also be ”wind-induced” or ”rain-induced”. Common for all local events is that it is not possible to determine growth rate of particles due to the shape of the event. Ion bursts observed with air-ion spectrometers, that cannot be explained by wind or rain, are called ”mixed-type” events.

2.2 Reaching to clouds — competition between source and sink processes

After the formation, aerosol particles need to grow by condensation up to 50 – 100 nm in size (Kerminen et al., 2012), in order to affect the climate. The rate with which the particles grow from size $d_{p1}$ to $d_{p2}$ (growth rate, $GR$) is determined as

$$GR = \frac{dd_p}{dt} = \frac{\Delta d_p}{\Delta t} = \frac{d_{p2} - d_{p1}}{t_2 - t_1}.$$
Essentially, there are two methods to determine $GR$ from the size distribution data: *maximum-concentration* and *mode-fitting method* (Yli-Juuti et al., 2011; Kulmala et al., 2012). With the maximum-concentration method, the $GR$ is calculated by a linear fitting to the maximum concentration of each size bin. In the mode-fitting method, a log-normal distribution function is fitted to each timestep in the size distribution and the $GR$ is then obtained with a linear fitting to the mode peak diameter time evolution. Usually the most of the particle growth is contributed by condensation of organic vapours (Riipinen et al., 2012). As a result, atmospheric $GR$s usually exhibit a strong maximum during the summer in the boreal region (Yli-Juuti et al., 2011). However, in areas with high sulphur pollution, sulphuric acid condensation can explain a major fraction of the growth. *E.g.* in Värriö, eastern Lapland, the growth rates have another maximum in winter (*Paper II*) due to high amount of sulphur pollution.

Aerosols cannot grow endlessly, but a constant competition takes place in the atmosphere between processes that grow or scavenge particles. After some time, sinks balance the effect of NPF and the aerosol number concentration is not increasing with time anymore. Over a boreal forest, an aerosol population reaches its equilibrium between the sources and the sinks after few tens of hours of overland time (*Paper III*), depending on the initial size distribution.

The ratio between the sinks and sources determines essentially the fraction of formed particles that can reach a certain size ($d_p$). The aerosol losses can be characterized by *coagulation sink* ($CoagS$). It is a measure of how fast aerosol particles are removed from the atmosphere.
through coagulation scavenging (collision between two particles) and is a function of both
the size of the scavenged particle and the size distribution of the other particles (Lehtinen
et al., 2007). Condensation sink (CS), which is typically used in aerosol studies to represent
the particle losses in the atmosphere instead of CoagS, is a measure of how fast vapour
molecules will condense onto pre-existing aerosol and is dependent on the effective surface
area of the particles and pre-existing aerosol size distribution. Both of these sink terms
are calculated from the aerosol size distributions (Kulmala et al., 2001; Dal Maso et al.,
2002; Kulmala et al., 2012). E.g. if the accumulation mode concentration is high, most of
the vapours will rather condense on those particles than take part in NPF or subsequent
growth. On the other hand, if the background aerosol consists mainly of large particles,
the freshly-formed particles will be quickly scavenged by coagulation. For very small, newly
formed particles, condensation and coagulation are essentially the same phenomenon as the
particles are close to the size of molecules. According to the Kerminen-Kulmala equation
(Kerminen and Kulmala, 2002; Kulmala et al., 2006), the formation rate of particles at size
d_p (J(d_p)) is exponentially dependent on the ratio between CS and GR:

\[
J(d_p) = J^* \exp \left\{ 0.23 \left( \frac{1}{d_p} - \frac{1}{d^*} \right) \frac{CS'}{GR} \right\}
\]

where \( J^* \) is the nucleation rate and \( d^* \) the initial cluster size (typically chosen to be 1 nm).
\( CS' \), having unit m\(^{-2}\), is directly proportional to \( CS \). According to Eq. 5, \( \frac{J(d_p)}{J^*} \) decreases
rapidly if the aerosol losses are high. For polar areas, \( CS \) is usually very low but during
summer \( GR \) may reach reasonably high values (e.g. Paper I). This means that the fraction
of newly-formed particles that reach certain size \( d_p \) can potentially be high, but there are only
few particles to begin with, since typically the values of \( J^* \) are low in polar areas (Kulmala
et al., 2004b), Yu et al. (2008), Paper I).

Planetary boundary layer (PBL) development plays also a major role in the daily cycle of
the aerosol concentration. When the PBL height increases during the day, the pre-existing
aerosol is diluted (given that the aerosol number concentration above the PBL is lower than
inside of it) and the aerosol total surface area per unit volume decreases. This process
may decrease the \( CS \) and CoagS sufficiently enough enabling NPF to initiate and particle
condensational growth up to climatically relevant sizes.

### 2.3 Aerosol-cloud-climate interactions

In order to affect the climate, aerosols need to be big enough — either to be able to inter-
act with solar radiation or to be able to act as cloud condensing or ice nuclei (CCN and
IN, respectively). Also the chemical composition and concentration of aerosols determines eventually the strength and sign (positive or negative) of the radiative forcing they yield. Aerosols affect the climate via various pathways. Traditionally these have been divided into direct effects (scattering and absorption), semi-direct effects (absorbing aerosols affect clouds by changing their stability) and 1st and 2nd indirect effects (increased aerosol number concentration changes cloud albedo and lifetime). In the most recent IPCC AR5 report (IPCC, 2013), the aerosol climatic effects are divided into aerosol-radiation and aerosol-cloud interactions. This new division is more physically based and divides also between atmospheric forcings, rapid adjustments and feedbacks. Thus, the “old” semi-direct and cloud lifetime effects are no more directly affecting the radiative forcing (RF), but they are considered as rapid adjustments, while direct and cloud albedo effects are changing directly the radiative forcing. Together, the direct and semi-direct effects contribute to the effective radiative forcing (ERF) caused by aerosol-radiation interactions and the other effects contribute to the ERF caused by aerosol-cloud interactions.

**Aerosol-radiation interactions**

Aerosols change the Earth’s radiative properties by scattering and absorbing the solar radiation. These processes are called aerosol-radiation interactions (ARI) and their radiative effects due to the somewhat straight-forward physical background are mostly well understood. Usually, scattering aerosols tend to cool the climate, whereas absorbing aerosols have a warming effect. In general, particles having smaller diameters than the wavelengths of visible light (around 400 – 750 nm) have very little impact on ARI. *E.g.* in southern central Finland, particles < 100 nm were found to contribute only less than 0.2% to the scattering and almost the entire contribution came from particles < 1 μm (Virkkula et al., 2011). If aerosols form a thick layer, they can decrease the amount of solar radiation reaching the surface. Also, if they are large enough, they can scatter, absorb and emit thermal radiation (Haywood and Boucher, 2000).

The influence of scattering or absorbing aerosols on climate depends not only on the properties of the interacting aerosol (chemical composition, size distribution and morphology as well as horizontal and vertical distribution) but also on the properties of the surface (Mylhe et al., 2009, 2013). Over bright surfaces, the effect of scattering aerosol is smaller than above dark surfaces, since most of the radiation is in any case reflected by the surface. However, if the aerosol is of absorbing kind (mainly black carbon, BC), it may be very effective in warming the atmosphere over reflective surfaces, such as glaciers or sea ice (Law and Stohl, 2007; Ramanathan and Garmichael, 2008; Sand et al., 2013), and enhance the Arctic warming even further.

ARI includes also rapid adjustments that arise from absorbing aerosols below, above or
inside the cloud, causing increased cloud heating and evaporation of cloud droplets and thus
decreasing precipitation (Ackerman et al., 2000; Myhre et al., 2013; IPCC, 2013). The extra
heating also changes the vertical temperature profile close to the cloud. In the literature,
these rapid adjustments are usually referred to as semi-direct effect.

**Aerosol-cloud interactions**

Clouds are an important part in the Earth’s energy balance. Globally, their effect on the
radiation budget is negative (IPCC, 2013), but clouds on different heights and over different
surface types act in various ways, sometimes cooling, sometimes warming the atmosphere.
This is due to their ability to scatter short-wave radiation as well as emit long-wave radia-
tion. Clouds that are in the lower part of troposphere are typically cooling the climate as
they scatter solar radiation and emit almost the same amount of long-wave radiation as the
earth’s surface. High altitude clouds, on the other hand, are usually so thin that the sun’s
radiation penetrates trough but the outgoing long-wave radiation from the surface of the
earth is reflected back to the atmosphere. Altogether, it is also estimated that the effect
of aerosols on clouds cause a negative radiative forcing yet its magnitude is very difficult
to assess and the estimations vary greatly (IPCC, 2013). In fact, aerosol-cloud interactions
(ACI) possess the largest uncertainties when estimating the future climate. ACI encom-
passes several mechanisms, all of which are all related to altering cloud properties due to
anthropogenic aerosols (Rosenfeld et al., 2014).

In the atmosphere, water vapour needs a surface — a seed — to start condensing on. Aerosol
particles may act as CCN (Kerminen et al., 2012) and a small fraction of them also as IN
(Hoose and Möhler, 2012; Murray et al., 2012), thus allowing cloud droplets or ice crystals
to form. The ability of an aerosol particle to act as CCN depends on the size of the particle
and aerosol chemical composition (Kerminen et al., 2012). Typically the critical diameter
$D_{50}$ — the diameter at which 50% of the particles are activated to form cloud droplets — is
somewhere around 80 – 100 nm in boreal and polar environments (Komppula et al., 2005;
Sihto et al., 2011), decreasing with increasing maximum water supersaturation at cloud base
(Kerminen et al., 2012). Thus, if the water supersaturation is high due to low number of
CCN, $D_{50}$ can be as low as 60 nm (Paper I). This kind of situation is typical in polar
regions, and already a small increase in aerosol number may increase cloudiness significantly
(Mauritsen et al., 2011). $D_{50}$ decreases also with aerosol age with decreasing hygroscopicity
(Duplissy et al., 2008). If the growth is sufficient, aerosols may act as CCN already some
hours after their formation (Kerminen et al., 2005; Kerminen et al., 2012; Paper I).

If the cloud droplet number concentration increases due to increasing number of CCN in-
side the cloud, the formed cloud droplets are smaller than in an unpolluted cloud. Con-
sequently, such clouds appear whiter as their unpolluted fellows and scatter more solar
radiation (Twomey, 1977). This effect is often called the Twomey-effect of cloud albedo effect. When a cloud is composed of smaller droplets, it will precipitate less and the time it takes for the precipitation to start is longer. Therefore the cloud can grow in height and its lifetime is also increased (Albrecht, 1989). The decrease in precipitation and increase in cloud height and lifetime are together usually referred to as Albrecht effect or cloud lifetime effect.

There are also effects related to increased amount of IN (DeMott et al., 2010; Murray et al., 2012), and it seems that the increase in their number can be related to the increase in the number of aerosols larger than 0.5 µm (DeMott et al., 2010). However, regarding the changes in precipitation, cloud lifetime and albedo, the effect of increased IN in mixed-phase clouds is opposite to that of increased CCN (Lohmann and Feichter, 2005). While increased CCN decreases the size of the cloud droplets, they also tend to stay in the atmosphere longer and freeze later. This increases the amount of supercooled water inside the cloud and, after it freezes and releases more latent heat, can lead to increase in the cloud height trough increased convection (Rosenfeld et al., 2008). Moreover, as the vapour pressure over ice is lower than over water, once the glaciation process starts, the freezing of the supercooled drops is very rapid (Murray et al., 2012). This means that the formed ice crystals grow rapidly and start precipitating also quickly (Lohmann, 2002). However, clouds are not only sensitive to the number of CCN or IN but also the ratio between CCN and IN determines the effects on e.g. cloud lifetime and precipitation (Murray et al., 2012).

What is needed for clouds to form?

Figure 4: Mechanisms that may lead to cloud formation. LCL is the Lifting Condensation Level — height at which the condensation of water begins.
It is important to note that reaching potential CCN sizes is only one step in the aerosol-
cloud interactions. Firstly, different cloud types — liquid, ice and mixed-phase, stratiform
and convective clouds — behave differently, and have their characteristic supersaturation
(Korolev and Mazin, 2003). Secondly, for clouds to form, special meteorological conditions
are required. The processes which lead to cloud development are many (Fig. 4), and take
place over different temporal and spatial scales (see e.g. Murray et al., 2012), but the base
mechanisms behind all of them can be divided into two: either the air parcel cooles down to
its dewpoint temperature or more humidity is added into the air.

Typical mechanism that cools down the air to its dewpoint temperature in polar and mid-
lattitudes is lifting at the fronts associated with low-pressure systems. The shape of the
forming cloud depends on the scale and speed of the airmass that is lifted. In a warm front,
warm air rises over cold air smoothly over long distance. Clouds that are associated with
warm fronts are typically stratiform in their nature — shallow, but covering large areas —
and the precipitation from these clouds is more or less continuous. In a cold front, the cold
air mass pushes and forces the warm air to rise sharply and rapidly, and the formed clouds
are cumuliform and convective in their nature, precipitating convective showers. Clouds in
occluded fronts are usually a mixture of clouds in warm and cold fronts, depending on the
type of the occlusion. Also convection not associated with fronts, orography or convergence
may lead to cloud formation. Typically the spatial and temporal scale of the clouds formed
by these mechanisms is small. An exception to this are the lenticularis-type clouds that are
formed in to the lee-side of the mountain ranges in the standing gravity waves. This type of
a cloud can stay in one place over somewhat long periods of time, if the air flow direction
and speed remain more or less unaltered.

Another way to produce clouds is to add humidity into the air until the air is saturated with
respect to water or ice. Typical mechanisms leading to water supersaturation are turbulence
or the effect of surface. The first mechanism is responsible for the Stratocumulus-formation
at the PBL top, where air from the free troposphere is mixed with air in the PBL (Crosier
et al., 2011). The latter mechanism is very important e.g. close to the sea ice edge, when air
mass that has been over cold sea ice is transported over open water, becomes supersaturated
with respect to water, and thick Stratus clouds (or fog) are formed. Both of the above
mentioned mechanisms are very common in the Arctic. Especially in the summer, the
Arctic is frequently overcasted by low stratiform clouds (IPCC, 2013).

2.4 Removal of aerosols

Eventually, aerosols are removed from the atmosphere. The processes that remove particles
from the atmosphere and bring them to surfaces (such as ground or needles) can be divided
into dry and wet deposition. The efficiency by which particles reach the ground or other
surfaces depend on a number of factors, such as the size of a particle, temperature and concentration of the particles and their gradient, atmospheric flowfield (including turbulence), relative humidity and the presence of clouds or fog and precipitation (Seinfeld and Pandis, 2006).

![Schematic figure of some of the dry and wet deposition processes and mechanisms.](image)

**Figure 5:** Schematic figure of some of the dry and wet deposition processes and mechanisms.

**Dry deposition**

Dry deposition refers to processes by which aerosols are removed from the atmosphere without solid or liquid water and it includes several mechanisms, some of which are depicted in Fig. 5. It is caused by turbulence (most important near the surface), microphysical and physical properties of the particles (their size, density and shape) as well as the roughness of the surface onto which the particles are deposited (Hinds, 1999; Seinfeld and Pandis, 2006). The most important processes that cause dry deposition are Brownian diffusion, interception, inertial impaction and gravitational sedimentation — all of them depending on the size of the particles.

The process where particle collides with an object due to Brownian diffusion is mostly important for very small particles and the diffusion efficiency decreases with an increasing particle size (Hinds, 1999). The net flux of particles is towards smaller concentrations. The particle transport due to Brownian diffusion is thus a function of particle size and kinematic viscosity. In interception, the particle follows the streamlines of a gas flow but the diameter of the particle is larger than the distance from the streamline to the surface, making the particle to collide with the surface (Hinds, 1999). The collision efficiency in this case depends
on the dimensions of both the particle and the target surface and increases with particle size. Impaction happens if the particle cannot change its direction fast enough due to its inertia, and collides with an object. This process is important for even larger particles (Hinds, 1999). Finally, for very large particle, more than few micrometers in diameter, gravitation starts to be important. Usually this mechanism is, however, less important compared with other dry deposition mechanisms, since it is also dependent on the wind speed: with high wind speeds impaction dominates over sedimentation. With all these processes combined, the dry deposition efficiency is highest for the smallest and largest particles and exhibits a minimum in the accumulation mode.

### Wet deposition

Wet deposition combines a number of processes that decrease the amount of particles in the atmosphere trough liquid or solid water. It can be divided into in-cloud and below-cloud scavenging depending whether the particle capture happens inside the cloud or outside of it (Seinfeld and Pandis, 2006). Sometimes in-cloud scavenging is also called as rainout, while below-cloud scavenging is referred to as washout. Both in-cloud and out-of-cloud (below-cloud) scavenging combine a number of different processes. Nevertheless, in all the processes, particles get into the vicinity of water, are scavenged by the hydrometeors and then deposited onto surfaces.

In-cloud scavenging can occur either when aerosol particles act as CCN or IN (nucleation scavenging) or when they are captured by falling water droplets (Seinfeld and Pandis, 2006). Particles that have acted as CCN (or IN) go trough various processes inside the cloud, trough which their chemical composition and size changes when the cloud evaporates. This transformation of the original particle population is called cloud processing (Seinfeld and Pandis, 2006). Cloud processing increases the particle size and solubility, which in turn change e.g. the activation properties of the aerosols. Evaporation can also take place outside of clouds from precipitating hydrometeors. The part of the aerosol population that stays inside the cloud but is not scavenged by nucleation (or much less important impaction scavenging) is called the interstitial aerosol. This part can be later scavenged by impaction to cloud droplets.

In below-cloud scavenging, the particles are captured by falling hydrometeors and are thus removed from the atmosphere. Below-cloud scavenging can be divided into rain and snow scavenging depending on whether the falling hydrometeors are liquid or solid. In addition to inertial impaction, processes that cause wet deposition during precipitation are Brownian diffusion, phoretic forces (caused by e.g. temperature gradient), turbulence and electrostatic forces. The scavenging efficiency of below-cloud scavenging is difficult to measure and thus not well known. The largest uncertainties are related to the electrostatic forces. However,
if only impaction scavenging is considered, the efficiency of below-cloud scavenging can be described with the scavenging coefficient $\lambda$ (Seinfeld and Pandis, 2006)

$$\frac{dc(d_p)}{dt} = -\lambda c(d_p) \quad (6)$$

where $c$ is the particle concentration, $d_p$ is the particle diameter, $t$ is time and $\lambda$ is the scavenging coefficient. $\lambda$ can be expressed as

$$\lambda(d_p) = \int_0^\infty \left[ \frac{\pi}{4} (D_p - d_p)^2 (U_T - u_T) \right] (D_p, d_p) E(D_p, d_p) N(D_p) dD_p \quad (7)$$

where $D_p$ is the rain droplet diameter, $U_T$ and $u_T$ are the terminal velocities of the falling droplet and scavenged particle, respectively, $E(D_p, d_p)$ is the collision efficiency of between the falling droplet and aerosol particle and $N(D_p) dD_p$ is the concentration of rain drops as a function of droplet diameter (Seinfeld and Pandis, 2006). The volume from which the falling droplet scavenges the aerosols, $\frac{\pi}{4} (D_p - d_p)^2 (U_T - u_T)$ (Fig. 6a) can also be expressed as $[A (U_T - u_T)]$ where $A$ is the area from which the droplet scavenges aerosols. Essentially, $A$ is also a function of time $(A(t))$ since the growing droplet usually grows in size while falling by colliding with other droplets (Fig. 6b). For snow crystals $D_p$ is often replaced by the capacitance of the crystal (a product of surface area factor and equivalent radius) (Miller, 1990; Pruppacher and Klett, 1997). For small particles it is safe to assume that $U_T >> u_T$ (the effect of gravitational settling is negligible for small aerosol particles), thus the first term reduces to $\frac{\pi}{4} (D_p - d_p)^2$.

While rain scavenging is not well known, snow scavenging is even less understood (Zhang et al., 2013) due to the vast amount of different types of solid precipitation. Several models trying to resolve the capture of aerosol particles by snowflakes exist, the most important ones being trajectory and flux models (Pruppacher and Klett, 1997). Trajectory models have suggested that the particles are scavenged at the rim of the ice crystals due to the strong horizontal flow underneath the crystal (Fig. 7a) (Pruppacher and Klett, 1997). The scavenging efficiency displays a maximum for crystals of sizes around 1 mm (Martin et al., 1980). For snow flakes, however, the scavenging efficiency dependent on the filtering trough the flake (Fig. 7b) (Mitra et al., 1990) and is larger than for ice crystals of the same size (Pruppacher and Klett, 1997). Compared to the equivalent water content, the scavenging efficiency of snow crystals can be tens of times larger than that of liquid precipitation (Pruppacher and Klett, 1997).

Theoretical (Martin et al., 1980; Miller and Wang, 1988) and observational (Laakso et al., 2003; Paper IV; Paramonov et al., 2011) studies show that particles of size 100 nm $-1 \mu m$
Figure 6: Volume of air that a falling water droplet of size $D_p$ with terminal velocity $V_T$ wipes while falling ($\left[\frac{\pi}{4} D_p^2 U_t\right]$, black lines) and the volume it scavenges aerosols of size $d_p$ with terminal velocity $v_T$ ($\left[\frac{\pi}{4} (D_p - d_p)^2 (U_T - u_T)\right]$, orange lines). (a) The volume increases with time (or distance) if the falling water droplet grows in size while falling (b).

Figure 7: A falling snow crystal scavenges aerosol particles at the rim of the crystal due to the strong horizontal flow below it, which is created by the falling crystal itself (a), while falling snowflake of the same size scavenges aerosols more efficiently since the air can go through the flake. In this case, the collection efficiency is dependent on the flow through the aggregate (b).

exhibit a strong minimum in the below-cloud scavenging efficiency. This minimum is called the Greenfield gap and is a result of less effective removal mechanisms in that size range. For particles less than 100 nm in size Brownian diffusion dominates, and for particles larger than 1 $\mu$m inertial impaction dominates the capture. On the other hand, the particles in the
size range of Greenfield gap act very efficiently as CCN. Electrostatic forces and turbulence decrease the deepness of the gap and changes in RH also have an effect. In general, higher RH has been shown to increase the scavenging efficiency of snow scavenging (Paramonov et al., 2011).
3 Measurements and data

3.1 Study sites

All of the studies presented in this thesis utilize field data of aerosol size distribution measurements. The study sites are located in different type of environments, ranging from boreal forest zone (Paper IV) and subarctic environments (Papers II-III) to continental ice sheet (Paper I), in both Arctic and Antarctic regions (Fig. 8). Out of the sites in the Northern Hemisphere, three are to the north of Polar Circle. The lengths of the datasets vary from couple of months (Paper I) to 13 years (Paper II). Photographs from all of the measurement sites along their surroundings are shown in Fig. 9.

Paper I was conducted at the Finnish Antarctic Research Station Aboa (73°03′S, 13°25′W) in Dronning Maud Land, Antarctica. The station is a summer station and located at the continental ice sheet, on top of a nunatak Basen, about 500 m a.s.l. and some 130 km from the sea. The nunatak Basen belongs to the Vestfjella mountain range, and the nearest mountain in this range is 30 km away from Basen. The ice sheet rises smoothly toward south and the closest large mountain range with wide range or cliffs is Heimefrontfjella, more than 200 km away (Fig. 1). The aerosol measurements are conducted in a container (Fig. 9) that is located about 200 m from the main station Aboa. The main wind direction is from the north-east making the contamination by the station and vehicles that are used at Aboa, minimal. The inlets used for collecting the air are at approximately 3 m above the ground. During the summer the top of Basen becomes biologically very active when the snow and ice melt into shallow, few tens of centimeters deep ponds and cyanobacteria Nostoc commune (Vaucher) living in the ponds becomes exposed. These ponds are approximately 2 km away from the aerosol measurement site and e.g. during FINNARP 2009-campaign, their total size was some hundreds of m². Aerosol measurements have been conducted at Aboa during several campaigns since 1997 and even year-round aerosol total number concentration and particle number size distribution (0.3–20 µm) measurements have been made from December 2003 until January 2007 (Virkkula et al., 2009). The expeditions and logistics are taken care by the Finnish Antarctic Research Program (FINNARP).

Paper II used long-term aerosol and trace gas measurements from SMEAR I (Station for Measuring Ecosystem—Atmosphere Relations) station (Hari et al., 1994) in eastern Lapland, Finland (67°46′N, 29°37′E). The measurement site was built in late 1991 and is located on top of a hill, 390 m a.s.l., about 1 km north of Värrö Subarctic Research Station and belongs to the Division of Atmospheric Sciences at the University of Helsinki. The station is inside of a 60-yr-old Scots pine (Pinus sylvestris) forest, at the north side of Värrö fell range. The fell range is one of the highest in the eastern Lapland, reaching to 500 – 550 m a.s.l. There are also some fells in the north, highest one being 484 m a.s.l. The area represents typical
Figure 8: Topographical maps of Arctic and Antarctica with the study sites added. Map cartographer Hugo Ahlenius / UNEP/GRID-Arendal
Figure 9: Measurement stations (lower figures) and their surrounding environments (upper figures). A=Aboa, B=Värriö, C=Abisko, D=Pallas, E=Hyytiälä. The measurement location is marked with dashed red circles in cases it is visible in the figure.

Lappish Scots pine forest with very little undergrowth. In addition, there are also gorges and large wetlands in the area. Most of the measurement infrastructure is placed inside a small hut, however the meteorological observations are done from 16-m-high mast and photosynthesis and respiration of the nearby trees are measured with chambers from outside. Also, some instruments have housings outside. In the beginning, the measurements included tree photosynthesis, trace gases, total aerosol number concentration and meteorology. The aerosol number size distribution measurements were started in 1998. The main source of pollution in the area is the smelter industry at Kola Peninsula. In fact, SMEAR I was originally built in order to measure the effect of Kola air pollution to the ecosystem in eastern Lapland. The major industrial cities are located in north (Nikel and Zapolyarnyj) and east (Montchegorsk), about 150 and 200 km from Värriö, respectively. There is also a mining area on the Russian side in Kovdor, 43 km south-east from Värriö and another mine is about to be opened on the Finnish side, in Sokli, only about 12 km north-west from Värriö. The closest small road is 8 km away and the nearest major road 100 km away. There are no local pollution sources.

Aerosol measurements at Pallas–Sodankylä GAW (Global Atmospheric Watch) station (Hatakka et al., 2003) are conducted at Sammaltunturi site (67°58′N, 24°7′E), on top of a fell at 565 m a.s.l., about 300 m above the surrounding area. The station is some 100 m above the timber line of the mixed boreal forest below, in the northernmost limit of the boreal forest zone. The Sammaltunturi fell belongs to the southern part of 50 km long north-south oriented chain of fells, with peaks reaching 600 – 800 m a.s.l. In addition to the forests and fells, there are also hills and wetlands in the area. Sammaltunturi is the largest of the four sites that belong to Pallas station. The closest town is 19 km west of the site.
There are no local or regional pollution sources in the vicinity. Aerosol measurements from Pallas were utilized in Paper III.

Likewise, in the same study we used aerosol data measured at Abisko research station (68°21′N, 19°3′E) in north-western Swedish Lapland (Svenningsson et al., 2008). The measurement site is located 14 km east from the research station, at Stordalen mire, in a wide east-west oriented valley, between lake Torneträsk and the Kiruna-Narvik road and railway lines. The surroundings consist of subarctic mires, birch forests and mountain tundra. Unlike the two other measurement stations in Paper III, Abisko is located close to high mountains, the Scandes, with peaks above 1500 m a.s.l., west from the station. The station itself is located 360 m a.s.l. The Atlantic Ocean is located approximately 100 km west from Abisko.

Paper IV utilizes aerosol number size distribution and detailed precipitation measurements from SMEAR II station in Hyytiälä (61°51′N, 24°17′E), southern central Finland. SMEAR II is currently the most comprehensive station in the world to study the atmosphere–biosphere-interactions. It combines research from various various different disciplines such as atmospheric sciences, forest ecology and soil chemistry. The studied research subjectives include the formation and growth of aerosols and their role in cloud formation, the impact of environment and tree structure on gas exchange, water transport and the growth of trees as well as budgets of a number of substances in the soil. The station includes a main building for trace gas and aerosol research, two huts for mass spectrometry and most of the aerosol instrumentation, 127-m-high mast for meteorological, trace gas as well as flux measurements, few about 20 m high towers for biosphere–atmosphere gas exchange and irradiation measurements and a container area including a new, 37-m-high tower for aerosol and molecular chemistry measurements. SMEAR II has the worlds longest and most complete time series of aerosol number size distribution measurements, measured with a Differential Mobility Particle Sizer (DMPS) from 1996 onwards. In addition to continous long-term measurements, also numerous intensive campaigning are organized at SMEAR II. The station is located 170 m a.s.l. inside a dense and rather homogeneous, approximately 50-year-old Scots pine forest, which extends several kilometers to the north and northeast. Small lakes are within the radius of few kilometers both in north and west. The infrastructure of Hyytiälä forestry station are located close to the nearest lake about 1 km west from SMEAR II. The third largest city in Finland, Tampere, is located about 50 km southwest from the station and another city, Jyväskylä, 100 km northeast from the station. There is also a small sawmill few kilometers south of Hyytiälä.
3.2 Methods

Aerosol and ion size distribution measurements

A Differential Mobility Particle Sizer (DMPS) (Aalto et al., 2001) measures the neutral aerosol particle number size distribution. It consists of a neutralizer, a Hauke-type Differential Mobility Analyzer (DMA) and a Condensation Particle Counter (CPC). First, the aerosol sample is brought to a charge equilibrium with e.g. $^{85}$Kr or $^{14}$C source. Then, the particles are directed into the DMA where they are classified according to their electrical mobility. Particles with selected electrical mobility are then introduced to the CPC which counts their number optically with laser after they are grown into optically visible size by condensing either water or butanol onto them. The sheath flow(s) is maintained with closed-loop arrangement with critical orifice. The size range can vary depending on the size and number of DMAs, detection efficiency of the CPC as well as chosen flowrates. For example, in Hyytiälä the measured range is from 3 nm to 1 $\mu$m but with the DMPS used in Paper I it was from 10 nm to 500 nm since the instrumentation consisted of only one DMA (Table 1). A DMPS system was the core instrumentation used in each of the studies presented in this thesis. Scanning Mobility Particle Sizer (SMPS), which is another instrument that is based on classifying the particles according to their mobility and measures the number size distribution of neutral particles, was also used in Paper III. The total aerosol number concentration is integrated from the measured number size distribution and usually compared with measured concentration with a separate CPC. The measured size ranges at different measurement locations are shown in Table 1. Usually the time resolution of one size distribution is around 10 minutes.

Table 1: Measured size range of aerosols and with DMPS (SMPS) instrumentation and number of DMAs in the DMPS system at different measurement stations. The years refer to the measurement periods used in this thesis.

<table>
<thead>
<tr>
<th>Station</th>
<th>Years</th>
<th>Paper</th>
<th>Size range [nm]</th>
<th>Nbr of DMAs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aboa</td>
<td>12/2009 – 01/2010</td>
<td>I</td>
<td>10 – 500</td>
<td>1</td>
</tr>
<tr>
<td>Värrö</td>
<td>1998 – 2004</td>
<td>II</td>
<td>8 – 850</td>
<td>2</td>
</tr>
<tr>
<td>Abisko (SMPS)</td>
<td>2005 – 2008</td>
<td>III</td>
<td>10 – 500</td>
<td>1</td>
</tr>
<tr>
<td>Pallas</td>
<td>2005 – 2008</td>
<td>III</td>
<td>7 – 500</td>
<td>1</td>
</tr>
<tr>
<td>Hyytiälä</td>
<td>2005 – 2008</td>
<td>IV</td>
<td>3 – 1000</td>
<td>2</td>
</tr>
</tbody>
</table>

Air-Ion Spectrometer (AIS) (Mirme et al., 2007) is a device to measure the number size distribution of charged clusters and aerosols. It is capable of measuring directly the early stages of atmospheric nucleation and subsequent growth and thus offers an effective method to study aerosol particle formation mechanisms. AIS consists of two parallel cylindrical
DMAs with 21 electrically isolated sections, that measure the electric field carried by the ions and produces the mobility distributions of both negative and positive air ions simultaneously between 3.2 and 0.0013 cm²V⁻¹s⁻¹. These values correspond to mobility diameters (Mäkelä et al., 2006) (i.e. Millikan diameters) between 0.8 and 42 nm. From the data provided by AIS it is possible to do detailed visual analysis of the NPF events since sometimes small local events where the particles are not growing much before they are sampled, cannot be observed with DMPS but are measured with AIS. **Paper I** utilizes the data from AIS.

**Atmospheric chemistry measurements**

**Paper I** used quartz filter samples in characterizing the overall chemical composition of the aerosol particles during 2 – 3 days of sampling. The air was sampled to the filters without any cut-off in the inlet and the filters were changed three times a week. Later, the filters were analyzed in Finland with a comprehensive two-dimensional gas chromatograph—time—of—flight mass spectrometer (GCxGC—TOF—MS) in order to elucidate the volatile and semivolatile aerosol organic compounds in the aerosol particles. The method includes extraction of the compounds, derivatization and individual identification (Ruiz-Jimenez et al., 2011a,b). The identification of the organic compounds in this method is based on the comparison of the spectral information obtained from the detector and the retention indexes calculated using authentic standards with the information that is provided by National Institute of Standards and Technology (NIST) and the Golm databases. Further, the identified compounds were classified into groups based on their chemical composition, namely into hy-
drocarbons, halogenated, nitrogen, sulphur, carboxyl, carbonyl, and hydroxyl compounds. During the FINNARP 2009 campaign, we also took samples from one meltwater pond, about 40 m$^2$ in size and 10–20 cm in depth. Two water samples were taken: one containing only pond water and another containing water and pieces of cyanobacteria *Nostoc commune* (Vaucher). Similarly as the quartz filter samples, these samples were brought to Finland in a frozen container and kept frozen until their chemical composition was analyzed with GCxGC—TOF—MS.

Two of the studies (Paper I and Paper II) utilized measurements of different trace gases. In Paper I $O_3$ was used as a tracer for air masses coming from upper troposphere and stratosphere. This information was used in order to see whether the observed NPF was due to aerosols and precursor gases ascending from higher altitudes e.g. during a stratospheric fault. We used a continuous ozone analyzer O342M by Environnement S.A. to monitor the ozone concentration. It uses the ultraviolet absorption in measuring the ozone concentration. The $O_3$ concentration at a given time is detected by the difference in UV absorption between ambient air and ozone-cleaned sample. Sulphur dioxide (used in Paper II) on the other hand, is measured with Thermo Environmental pulsed fluorescence analyzer. The model of the device has been changed several times since 1992 when the measurements were started, however in all of the instruments the operating principle has been based on measuring the emitted fluorescence of $SO_2$ that is produced when the gas absorbs UV radiation.

**Meteorological observations and methods**

In order to calculate the snow scavenging coefficients in Paper IV, we needed to separate between different types of precipitation. For this, we used data from Vaisala FD12P automatic weather sensor. It measures the amount and intensity of precipitation, visibility and distinguishes between fog and different precipitation types (FD12P Manual, 2002). The amount of precipitation is measured with a capacitive precipitation sensor. The weather type is calculated using all measured parameters and interpreted into general weather codes used by WMO (World Meteorological Organization) and NWS (National Weather Service), in e.g. synoptic weather reports.

All of the studies used various meteorological measurements in interpreting the data. The measurement height and parameters used varied in each of the studies. In Värriö, for example, there is a 16-m-high weather mast next to the measurement hut with various meteorological variables measured at different heights, whereas at Aboa, there is a small automatic weather station owned by Finnish Meteorological Institute (FMI). All of the different sites at Pallas-Sodankylä GAW station are equipped with automatic weather stations and Salmaltunturi site has an additional Vaisala *FD12P* present weather sensor. In Hyytiälä, the meteorological measurements are most comprehensive of all of the stations, including the
measurements done from the 127-m-high mast. Measurements of meteorological variables are of utmost importance when analyzing the differences in aerosol properties since they provide background information of the state of the atmosphere and give information of the origin of the airmass.

Three of the studies (Papers I, II and III) used HYSPLIT (HYbrid Single Particle Lagrangian Integrated Trajectory) backward trajectories (Draxler and Hess, 1998) in calculating where the air masses had been at a given time before arriving at the measurement sites. Despite their limitations, back-trajectories are an efficient tool to study the air mass history. In calculating the air mass path HYSPLIT exploits GDAS (Global Data Assimilation System) reanalysis data provided by the NWS (before 2007 the reanalysis is from Final Analyses, FNL, dataset). The uncertainty of the trajectories can be 10 – 30% of the distance the trajectory has travelled (Stohl, 1998), thus after some hundreds of kilometers the uncertainty becomes already quite large. However, to overcome this it is possible to estimate the uncertainties by calculating either matrix or ensemble trajectories. Matrix trajectories are a set of trajectories calculated to a mesh of end-points all close to each other. Ensemble trajectories are also a large set of trajectories but instead of calculating them into different points in space, they are calculated by varying the initial conditions slightly in each single trajectory calculation.
4 Results and discussion

The following chapters summarize the main results of this thesis. For more detailed discussion the reader is encouraged to have a look at Papers I-IV. The first two chapters concentrate on the atmospheric new particle formation from both biogenic and anthropogenic viewpoints. The first chapter focuses on the observation of NPF from biogenic precursors in the continental Antarctica and discusses its importance on a global scale. The second chapter summarizes the effect of sulphur pollution to the trends in atmospheric new particle formation in remote areas. The evolution of aerosol population during air mass transport over land is discussed in chapter 4.3 and the chapter 4.4 concentrates on aerosol removal by snow. The last chapter gathers all the results and looks at them in the context of the changing climate.

4.1 New particle formation in glaciated areas: biogenic influence in the changing climate

Figure 11: Estimated formation areas of particles associated with the observed regional nucleation events, 1.−3.1.2010 (blue markers) and 17.−20.1.2010 (red markers). Approximate regions of meltwater are shaded with light blue. For detailed information about the shape and size of the markers, see Fig. 8 in Paper I.

Previously it was thought that continental Antarctica is completely free of secondary aerosol sources. The known sources for secondary aerosols observed over the continent were marine DMS emissions oxidizing into $H_2SO_4$ (O’Dowd et al., 1997; Davis et al., 1998; Asmi et al.,
2010; Yu and Luo, 2010), long-range transport (Ito, 1993; Fiebig et al., 2009; Hara et al., 2011), an intrusion of air from upper troposphere into the boundary layer (Virkkula et al., 2009) and a very minor contribution from local anthropogenic sources (Park et al., 2004). In contrast to this earlier understanding, Paper I shows that aerosols are also formed from continental biogenic sources and that the found source has importance not only in Antarctica but also globally.

Figure 12: Schematic of the enhancement of meltwater ponds to NPF in Antarctica and other glaciated areas.

During the FINNARP 2009 expedition we observed two intense NPF periods, 1. – 3.1.2010 and 17. – 20.1.2010, including both local and regional particle formation (Fig. 11). Altogether during these periods, 7 local (including 5 apple-type and 2 bump-type) and 5 regional (banana-type) NPF events were observed. During these times the values of $J_{1.6}$ and $J_{10}$ were comparatively high (Table 2) and the particles grew very fast compared to other observations from Antarctica (Table 2). Further, we made an estimation for the formation rate by assuming that ion-induced nucleation would explain 10% of the observed value of $J_{1.6}$. We also assumed that the particles were formed with kinetic nucleation of sulphuric acid (Eq. 1) and used several values for the coefficient $k$. With this, we could then calculate the amount of sulphuric acid that would be needed in order to explain observed particle growth rates. During both periods, the contribution of $H_2SO_4$ to the growth was only few percentages, being most of the time less than one percent (Table 2).
We then estimated the source regions of regional particle formation events using HYSPLIT back trajectories and observed particle formation rates. Particles observed during regional formation events were formed over areas of meltwater (in the large blue ice areas close to the border between sheet and shelf ice and over the mountain ranges, Fig. 11). Also, we roughly estimated the particle growth rates of the local NPF events by making a linear fit from the last observation of no visual particle formation to the first observation of visual particle formation observed in AIS. With this, and local wind speed we could then give an order of magnitude estimation of where the particles had originated. In each case the estimated formation area was over the ponds about 2–3 km away from the station. The formation of aerosols over the ponds was also verified with chemical analyses. We made both quartz filter measurements at the aerosol measurement site and also took samples of the pond water and algae. Both the filters and the biological samples were analyzed using the same technique. We found the same chemical components from the water and algae as from the filters during the intense NPF periods, whereas at other times these components were not observed in the filters or their concentration was very small.

Finally, the second NPF period showed also the potential of the pond emissions to affect cloud droplet number concentration, as the particles that were formed from the precursor vapours in the ponds acted as CCN only few hours after their formation. Without organics growing the aerosols, the growth up to CCN would take days and the particles would likely be scavenged from the atmosphere before reaching those sizes. What was also remarkable that due to very low accumulation mode number concentration, already at 60 nm half of the particles could act as CCN. This episode of CCN activation 17–18.1.2010 showed how meltwater ponds and the algae living inside of them can both be a large source of aerosols during summertime in Antarctica and the organics evaporating from such ponds contribute to the growth of the particles very significantly, allowing them to act as CCN. During this episode, the growth up to CCN took only 9 hours.

Figure 12 summarizes the contribution of melt water ponds and their vegetation (namely Nostoc commune) to the formation and growth of aerosols in Antarctica, even though the same process is likely to take place in other glaciated areas as well. The main source of secondary aerosols in Antarctica is the emission of DMS from the ocean and its oxidation to H$_2$SO$_4$ that then forms clusters. Cyanobacteria Nostoc commune and possibly other biota in the ponds emit also sulphur compounds (DMS and H$_2$S) (Gries et al., 1994) which can take part in this first step of atmospheric new particle formation. However, their major contribution to the atmospheric NPF and subsequent aerosol growth comes from the emissions from organic vapours. Those vapours can stabilize the clusters allowing them to grow further. After this step, the organic vapours also contribute substantially to the growth enabling the particles to act as CCN before they are scavenged from the atmosphere by other means. As Nostoc commune is a common species around the world, and meltwater ponds are found
from all glaciated areas, it is very probable that it takes part into atmospheric NPF and growth of aerosols also elsewhere.

Table 2: Observed values of \( J_{1.6} \), \( J_{10} \) and \( GR \) (calculated from the regional events) as well as calculated percentage that sulphuric acid explains of the observed growth for the two intense NPF periods, 1. – 3.1.2010 (Period I) and 17. – 20.1.2010 (Period II). Also shown are other values obtained from Aboa during a simultaneous observation of a local event in AIS- and a regional event in DMPS (Asmi et al., 2010), comparable to the Period II during FINNARP 2009 expedition, as well as growth and formation rates obtained from elsewhere Antarctica.

<table>
<thead>
<tr>
<th>Period</th>
<th>( J_{1.6} ) [cm(^{-3})s(^{-1})]</th>
<th>( J_{10} ) [cm(^{-3})s(^{-1})]</th>
<th>( GR ) [nmh(^{-1})]</th>
<th>expl % with ( H_2SO_4 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Period I</td>
<td>0.02 – 0.1</td>
<td>0.08 – 0.3</td>
<td>2.1 – 6.5</td>
<td>0.9 – 4.2</td>
</tr>
<tr>
<td>Period II</td>
<td>0.008 – 0.27</td>
<td>0.003 – 0.3</td>
<td>1.8 – 8.8</td>
<td>0.5 – 0.9</td>
</tr>
<tr>
<td>Aboa, 21.1.2007 (Asmi et al., 2010)</td>
<td>0.3 ± 0.1</td>
<td></td>
<td></td>
<td>1 – 3</td>
</tr>
<tr>
<td>Aboa (Koponen et al., 2003) ( J_2 )</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dome C (Järvinen et al., 2013)</td>
<td>0.023</td>
<td></td>
<td>2.5</td>
<td></td>
</tr>
<tr>
<td>Mawson (Gras, 1993)</td>
<td></td>
<td></td>
<td></td>
<td>0.1</td>
</tr>
<tr>
<td>South Pole (Park et al., 2004)</td>
<td></td>
<td></td>
<td></td>
<td>0.13</td>
</tr>
</tbody>
</table>

### 4.2 New particle formation close to large sources of anthropogenic sulphur

Once emitted to the atmosphere, sulphur dioxide (\( SO_2 \)) is oxidized into sulphuric acid (\( H_2SO_4 \)), which is known to be the most important chemical component in the early stages of atmospheric new particle formation. Thus if \( SO_2 \) emissions decrease, also the fraction of NPF days is expected to decrease. But while the probability for NPF decreases, \( CS \) must also decrease since the particles that are formed, form also the pre-existing aerosol (that determines the value for \( CS \)) after atmospheric aging. When more vapours are left in the atmosphere, their probability to take part in NPF, and hence the probability of NPF, increases. These counteracting processes related to decreasing sulphur pollution were studied in Paper II.

Due to the convenient location next to Kola Peninsula and one of the worlds longest time-series of aerosol number size distributions, Värriö is an ideal place to study the effect of decreasing sulphur emissions to the trends in new particle formation. The site experiences high sulphur pollution episodes when air masses arrive at the station over the industrial
areas in Kola Peninsula but the background aerosol concentrations in general at the site are low. We did a throughout survey taking into account a vast number of different NPF-related quantities and parameters for the years 1998 – 2011 to see what kind of trends they exhibited. Lacking direct measurements of $\text{H}_2\text{SO}_4$, we calculated the proxy for the $\text{H}_2\text{SO}_4$ concentration with two already available parameterizations (Equations 8 and 9). The first parameterization is physically well-based (Petäjä et al., 2009), however it is prone to be slightly inaccurate in environments with low $\text{CS}$ values. The second proxy (Mikkonen et al., 2011) has not been tested for conditions typical for Väriö, but we used it to test the sensitivity of the $\text{H}_2\text{SO}_4$ concentration given by the first proxy. The time-series analysis was done using two approaches: least squares covariance method that takes into account the seasonality and a non-seasonal Mann-Kendall -method which provides Sen’s slopes with lower and upper confidence intervals and the significance of the trend (Table 3). Significant trends ($P < 0.05$) were found from 18 independent variables. The least squares slopes were calculated using a Matlab function $\text{lscov}$. More detailed description of the fitting procedures can be read from Paper II.

$$[\text{H}_2\text{SO}_4]_P = \frac{k_3 \cdot \text{SO}_2 \cdot \text{Rad}}{\text{CS}} \quad (8)$$

$$[\text{H}_2\text{SO}_4]_M = 8.21 \times 10^{-3} \cdot k \cdot \text{Rad} \cdot [\text{SO}_2]^{0.62} \cdot (\text{CS} \cdot \text{RH})^{-0.13} \quad (9)$$

Overall, in 13 years, the number of clear, class I events per year decreased from more than
Table 3: Trends for various parameters obtained using both least squares covariance method where the seasonal trend has been taken into account, as well as Mann–Kendall method (Sen’s slope) without seasonality, as percentages per year. The slopes are for years 1998 – 2011, unless mentioned otherwise. The lower and upper 95th confidence intervals for the Sen’s slopes are also given. All of the trends are significant \((P<0.05)\). Table adapted from Paper II.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>l.s.cov slope</th>
<th>Sen’s slope</th>
<th>Lower conf. (SL)</th>
<th>Upper conf. (SL)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>[%/yr(^{-1})]</td>
<td>[%/yr(^{-1})]</td>
<td>[%/yr(^{-1})]</td>
<td>[%/yr(^{-1})]</td>
</tr>
<tr>
<td>(SO_2)</td>
<td>−10.9</td>
<td>−11.3</td>
<td>−12.4</td>
<td>−10.2</td>
</tr>
<tr>
<td>(H_2SO_4) (Petäjä)</td>
<td>−5.2</td>
<td>−6.2</td>
<td>−7.5</td>
<td>−4.9</td>
</tr>
<tr>
<td>(H_2SO_4) (Mikkonen)</td>
<td>−8.0</td>
<td>−8.6</td>
<td>−9.5</td>
<td>−7.7</td>
</tr>
<tr>
<td>Nucleation mode</td>
<td>4.3</td>
<td>3.7</td>
<td>2.7</td>
<td>4.7</td>
</tr>
<tr>
<td>(CN &gt; 80nm)</td>
<td>−4.1</td>
<td>−4.2</td>
<td>−4.9</td>
<td>−3.5</td>
</tr>
<tr>
<td>(CS)</td>
<td>−7.7</td>
<td>−8.0</td>
<td>−8.7</td>
<td>−7.4</td>
</tr>
<tr>
<td>(J_3) (2006 – 2011)</td>
<td>26.2</td>
<td>29.7</td>
<td>15.5</td>
<td>42.2</td>
</tr>
<tr>
<td>Event class I</td>
<td>−9.9</td>
<td>−14.2</td>
<td>−5.9</td>
<td></td>
</tr>
<tr>
<td>All events</td>
<td>−3.7</td>
<td>−5.7</td>
<td>−0.5</td>
<td></td>
</tr>
<tr>
<td>Non-events</td>
<td>3.1</td>
<td>1.2</td>
<td>5.1</td>
<td></td>
</tr>
</tbody>
</table>

40 to less than 10 events per year (−9.9%/year), while the number of non-events increased (Table 3). In the end of the timeseries, the amount of such events was around the same that is observed in Pallas, which is not experiencing pollution episodes from Kola. The decrease in anthropogenic \(SO_2\) and primary emissions decreased the \(CS\), leading to an increase in both event-time \(J_3\) and overall nucleation mode concentration. Despite of this, the particles did not grow enough and less events were observed in spring and autumn when the importance of sulphuric acid to the growth was the largest and the remaining vapour concentrations were not high enough to grow the particles up to CCN sizes. This could be seen as a clear decrease in the number concentration of aerosols larger than 50 nm and also in total aerosol concentration.

It should be noted that it is unlikely that the decrease in sulphur pollution would be resulting from changes in the filtering technology or deactivation methods, since the amount of deactivated emissions has remained the same. More likely, it is explained by the changes in the socio-economical factors, e.g. the decline in the economy and population in the Murmansk Oblast region (Paper II).

The event start time after sunrise was earliest in late winter – early spring when also the concentration of \(SO_2\) during the start of the event was highest (Fig. 13). Some events occurred also in the absence of sunlight and these events were connected to twice as high
SO$_2$ concentration as the event days on average. When looking at the event start time as a function of global radiation and sulphur dioxide, the high SO$_2$ concentrations advanced the onset of NPF by several hours, especially when there was more sunlight available (Fig. 13). When looking at the air mass history during event days and in general, nucleation seemed to occur more frequently in air masses that were coming over Kola Peninsula than on average trough late winter to autumn. Also, the nucleation mode particle number concentration and sulphur dioxide concentration were shown to correlate in air masses coming from Kola but not when the air was coming from clean sector. All of these are clear evidences that the rapid decrease in NPF frequency in eastern Lapland has been due to the decrease in sulphur pollution originating from Kola Peninsula. It is very likely that in areas with low background aerosol concentrations but close to large sources of anthropogenic sulphur emissions, the trends in NPF are governed by both the changes in the sulphur emissions due to overall human activity and cleaning of the emissions as well as natural biogenic emissions.

4.3 Aerosol dynamics during air mass transport

Paper III studied the time evolution of aerosol number size distributions by comparing two approaches and combines them with new particle formation analysis from three measurement stations — Abisko, Pallas and Värriö — located in northern Scandinavia. The first approach, a semi-Lagrangian method, was introduced by Tunved et al. (2006) where air mass trajectories that arrive at the stations are classified according to their transport time over land. In this approach, the initial condition is clean marine air and surface type is rural boreal forest. The second method is a Lagrangian method which combines simultaneous aerosol number size distribution measurements from station pairs (Tunved et al., 2004). In our study, the aerosol number size distributions at the upstream station were clustered to obtain internally similar initial states. The number of clusters was chosen so that each cluster was as tight as possible and clusters with less than 30 cases in the upstream station were discarded. With these approaches we investigated the differences between air masses that arrived at different stations after staying over land for a given time as well as aerosol dynamics when air masses were transported between different station pairs, both from east to west and west to east. We also investigated the timescales that it takes for source and sink processes to balance each other.

Overall, the apparent increase rates of particle mode diameters, accumulated mass concentrations and condensation sinks had similar behaviours between all the stations with the same overland times of air masses. However, when looking at the NPF probability distributions as a function of overland time, there was a difference between Abisko and the two other stations, although the different measuring periods may cause a bias to the results. In case of Värriö and Pallas, the NPF probability peaked when the air masses had stayed over
Figure 14: Mean aerosol number size distribution changes during summertime, when air mass transport is from west to east and east to west. The solid and dashed lines are the mean distributions at the upwind and downwind stations, respectively, and the blue and red lines are station pairs Abisko-Pallas and Pallas-Värröi, respectively.

land for 20 – 25 hours while for Abisko the probability distribution had highest values at the shortest overland times and decreased smoothly toward longer times. We concluded that this difference was due to the boundary layer meteorology: Pallas and Värröi are both located on top of a fjeld and affected mostly by horizontal winds, while Abisko is located on the lee side of the Scandinavian mountain range and affected by air that is descending from the mountains. The standing gravity waves generated by the mountain range (Schroeder et al., 2009) may compress the boundary layer, leading to enhanced accumulation of condensable vapours inside the PBL and allowing earlier onset of nucleation.

Even in the absence of any observable NPF, the Aitken mode grew 1 nm h\(^{-1}\) or less. The growth was most pronounced when the air mass had been transported from west to east. In general, when looking at the west to east transport, both Aitken mode number concentration and mode peak diameter grew distinctively (Fig. 14). When transported from east to west, several processes occurred simultaneously causing the apparent growth in the mode peak diameter to be very minimal. These differences were explained by the age of the aerosol population: air masses arriving from west to east were much younger and marine, while air masses arriving from east were well aged and were of more continental origin. The net effect of the source and sink processes seemed to be balanced when the air mass had been over land for 30 – 40 h.
4.4 Aerosol removal by snow

Sub-micrometre aerosol particle removal by snow is poorly known, yet it is an important removal mechanism in the polar and moutaneous regions as well as upper troposphere. Since most of the air pollution in the Arctic originates from lower latitudes, understanding of the efficiency with which snowfall scavenges the pollutants is essential. Paper IV introduced a parameterization for size-segregated snow scavenging coefficient for particle sizes 10 nm – 1 µm derived from four years of field observations and presents case-studies with UHMA (University of Helsinki Multicomponent Aerosol model) using the obtained parameterization.

We calculated the size-segregated snow scavenging coefficient $\lambda_s(d_p)$ using a semi-empirical approach given by Equation 10. A similar approach had been earlier applied to rain scavenging by Laakso et al. (2003) and later used in a snowscavenging study in an urban setting by Paramonov et al. (2011). Equation 10 results from integrating Eq. 6 from time $t_0$ to time $t_1$ with corresponding particle concentrations $c_0(d_p)$ and $c_1(d_p)$.

$$\lambda_s(d_p) = -\frac{1}{t_1 - t_0} \ln \left[ \frac{c_1(d_p)}{c_0(d_p)} \right]$$ (10)

$$\frac{dc}{dt} = -\Lambda_s c = -\lambda_s c \pm \left( \frac{dc}{dt} \right)_{\text{instr}} \pm \left( \frac{dc}{dt} \right)_{\text{turb}} \pm \left( \frac{dc}{dt} \right)_{\text{adv}} \pm \left( \frac{dc}{dt} \right)_{\text{cond}} \pm \left( \frac{dc}{dt} \right)_{\text{nucl}} \pm \left( \frac{dc}{dt} \right)_{\text{coag}} \pm \left( \frac{dc}{dt} \right)_{\text{hygr}}$$ (11)

This equation, however, is only valid if snow scavenging is the only process changing the particle number concentration. In practise there are many other sources and sinks that can take part simultaneously. These error sources are given in Eq. 11, where $\Lambda$ is the experimental snow scavenging coefficient. Namely, the processes that change the aerosol concentration are instrumental errors, turbulence, advection, condensation, nucleation, coagulation and hygroscopic growth. By using a careful data selection criteria and two consecutive measurements, we could neglect the error sources. Using two consecutive measurements, however, leads to a large range of $\Lambda_s$ values, often both positive and negative, since the particle concentration does not decrease monotonically. Therefore, we also used another approach where a linear fitting is made to Eq. 12 over the individual snowfall periods. With this approach, only one $\Lambda_s(d_p)$ is obtained for each snowfall episode thus reducing the amount of instrumental noise and atmospheric fluctuations.

$$\ln \left[ \frac{c(t, d_p)}{c(0, d_p)} \right] = \Lambda_s(d_p) t$$ (12)
Figure 15: Median snow scavenging coefficient and two parametrizations for $\Lambda_s$ from Paper IV and Paramonov et al. (2011). In addition, the parametrization by Laakso et al. (2003) for water scavenging is shown.

After choosing approximately 160 hours of snowfall (corresponding to around 960 particle size spectra), we then calculated the median $\Lambda_s(d_p)$ for the whole dataset. The obtained $\Lambda_s$s for 10 – 1000 nm sized particles ranged from $8.7 \times 10^{-6}$ s$^{-1}$ to $5.2 \times 10^{-5}$ s$^{-1}$ with Greenfield gap (minimum in the $\Lambda_s(d_p)$) at around 200 nm. Then, we fitted a parameterization (Eq. 13) into the median $\Lambda_s(d_p)$ (Fig. 15). Similar parameterization was already done for rain scavenging (Laakso et al., 2003), only the parameterization for water scavenging had 3 more parameters, one of them including the precipitation intensity. Our dataset consisted almost entirely of slight continuous snowfall (89% of the cases) having only a narrow range of intensities (median 0.2 mm/h), making it impossible to include it into the parameterization. However, our parameterization gave higher values than the parameterization for liquid precipitation (Laakso et al., 2003) or even similar parameterization than ours, but for more polluted air (Paramonov et al., 2011). Yet, the latter study had, on average, much lower values of RH during snowfall, which is more likely to be the reason for the lower $\Lambda_s$ than the high aerosol backround concentration. Also, a case study of a snowfall with intensities higher than 5 mm/h scavenged the air more efficiently compared with those cases with low snowfall intensity (median $\Lambda_s$ was $8 \times 10^{-5}$ s$^{-1}$). In addition, out of thousands of functions tested, another parameterization based on Pearson IV function was established since this function gave physically most reasonable fit (Fig. 15).

\[
\log_{10} \left( \frac{\Lambda}{\Lambda_0} \right) = a + dd_p^{-2} + \epsilon d_p^{-1} \tag{13}
\]
The obtained parameterization (Eq. 13) was implemented into UHMA (University of Helsinki Multicomponent Aerosol model) and tested in a few case studies. The parameterization was proven to reproduce the time-evolution of the aerosol number size distributions well in all the cases. We also tested the relative effect of the other aerosol dynamical processes by neglecting other processes than snow scavenging. These tests did not differ much from the studies with all aerosol dynamical processes included, demonstrating the great relative importance of snow scavenging in modifying the aerosol size distribution.

4.5 Climatic implications

This chapter ties up all the main results of this thesis and links them with future climate predictions. Also, this chapter speculates the significances of the results from the climatic perspective especially in the polar regions but some of the results have even global importance. Figure 16 depicts the areas of interest and climatic implications of the papers presented in this thesis.

Figure 16: Schematic figure illustrating the areas of interest and climatic implications of the papers. Paper I contributes to the understanding of NPF from biogenic precursors in glaciated areas as well as from freshwater vegetation. Paper II focuses on the effect of large sources of anthropogenic sulphur on NPF trends. Paper III is a throughout study of the aerosol dynamical processes during atmospheric transport and Paper IV adds to the understanding of snow scavenging of aerosols and the results are applicable to GCMs.

Paper I addresses essentially the aerosol formation and growth in continental Antarctica from biogenic precursors. It shows that meltwater ponds that form during summer, contribute to a great extent in NPF and subsequent growth and that the particles can grow to CCN sizes within 9 hours with the help of organic vapours that are emitted from the ponds. There is already evidence of increased amount of meltwater and also increased amount of biological activity in meltwater ponds in Antarctica, related to climate change. However, similar glaciated areas with meltwater production are also found from the Arctic, e.g.

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Greenland, Svalbard or Canadian Arctic. Thus, this new source for secondary aerosols and CCN can be of great importance also in the Northern Hemispheric polar regions and since Arctic areas are warming faster than Antarctica, these sources can also grow in importance faster in the Arctic. All in all, more particles are expected to be formed from biogenic precursors during summertime in both Antarctic and Arctic regions in the future. This, combined with possibly increasing atmospheric transport to and within the Arctic, will likely affect the cloudiness and cloud properties in large areas.

Moreover, **Paper I** presents the first observations of freshwater vegetation as an aerosol precursor source. Since the species *Nostoc commune* (Vaucher) is living abundantly in various habitats around the world, it can be responsible for NPF and aerosol growth also elsewhere, especially in non-polluted areas such as large Siberian or Canadian wetlands. The aerosol forming potential of the vapours emitted by *Nostoc commune* should be studied further both with laboratory and field experiments.

**Paper II** shows that in areas with low background aerosol number concentrations but close to large sources of sulphur pollution, the concentration of sulphur dioxide has a major impact on the NPF frequency and aerosol formation time. *E.g.* in Värriö, from 1998 to 2011, the number of clear event days has decreased to 16% of the initial value while the concentration of sulphur dioxide has decreased down to 20% of the original concentration. Changes in the sources of sulphur pollution can affect the trends in the potential CCN in at least two ways. On one hand, the decreasing sulphur pollution may decrease NPF in a way explained in section 4.2 and thus decrease the concentration of the potential CCN particles, while on the other hand, it has the possibility to increase the number of potential CCN by increasing the formation rate due to lower $CS$ and $CoagS$. In **Paper II** it was shown that the decreasing pollution leads to a decrease in the number of potential CCN particles because, in those pristine areas which experience occasional sulphur pollution episodes, sulphur dioxide and sulphuric acid seem to have a large role also in the growth of particles.

Maritime activity in the Arctic Ocean has been under scientific interest for some time already, since the climate predictions show that Arctic will be free of sea-ice during the summer by the mid 21st century (Stroeve et al., 2012) and the Northern Sea Route (also known as the Northeast Passage) as well as the Northwest Passage have been already navigable for several summers. As shown in **Paper II**, the trends in NPF in pristine areas close to the large source(s) of sulphur pollution are governed by the overall human activity, cleaning of the emissions and changes in natural biogenic emissions. This in mind, the increased shipping in the Arctic (Smith and Stephenson, 2012) will inevitably bring more aerosol sources to the Arctic. On the other hand, if the sulphur emissions from Kola Peninsula continue to decrease, the NPF frequency can decrease further, or at least stay at the present level if the natural background level is already reached. However, since there is no evidence that the smelters in Kola Peninsula would be using more efficient filtering techniques or deactivation
methods, the NPF frequency over those areas can also increase if warming Arctic climate draws more human activity back to Murmansk-Oblast region.

As discussed already in the Introduction, Arctic amplification leads to decreased temperature gradient between the Arctic and mid-latitudes, which then changes the atmospheric dynamics and enhances the atmospheric transport of pollutants and aerosols in general, into and within the Arctic. It is important to understand the aerosol dynamics related to atmospheric transport when making predictions of the future aerosol and cloud properties in the Arctic. **Paper III** shows that aerosol condensational growth is important even in the absence of observable NPF especially during summertime. This was assumed to be an indication that biogenic volatile emissions are tightly linked with the particle growth. The same study also showed that in airmasses that are transported from maritime areas to more continental areas, aerosol sources are more important than aerosol sinks, increasing the aerosol concentration up to $30 - 40$ hours of overland time. These results, combined with likely increasing biogenic emissions and faster atmospheric transport, suggest that the spatial distribution of aerosols and aerosol number concentrations will change in the Arctic in the future. Also, if significant amounts of biogenic volatile organics are emitted over longer periods of time during a year, the seasonal cycle of the aerosol number concentration and number size distribution will likely change.

Already now snowcover appears earlier in autumn in Siberia due to the shrinking Arctic sea ice (Cohen et al., 2012). **Paper IV** shows that compared with liquid precipitation, even light snowfall is more efficient in scavenging aerosol particles. This would imply that in the future, the aerosol removal is more efficient in Siberia, especially during late autumn and early winter. As currently pollution originating from as far as southern Russia and China reaches the Arctic, one could speculate that with increasing aerosol removal due to more solid precipitation in Siberia, less pollutants could reach the Arctic if they are already scavenged in Siberia. However, it is expected that in *e.g.* Scandinavia the amount of precipitation will increase even though most of it will fall in liquid form. For aerosol removal, this could still mean more efficient scavenging if the increase in liquid precipitation counteracts the decrease in the scavenging efficiency when more precipitation fall as liquid water instead of snow. Especially this is possible if the rainfall rates increase in the future, as it is expected (IPCC, 2013). At the same time, for mountainous areas where most of the precipitation falls in solid form, the aerosol scavenging efficiency is subject to change if the precipitation comes in liquid form instead. The biggest changes are expected to occur around areas with temperatures close to zero degrees during winter.
5 Review of papers and the author’s contribution

Paper I investigates new particle formation in Antarctica during a two-month FINNARP 2009 expedition. It introduces the first observations of NPF of continental origin in Antarctica, and shows that summertime melt water ponds and organic matter in them are responsible for a large fraction of both secondary aerosol formation and subsequent growth even some hundreds of kilometres downwind of the sources. The study also shows how the particles that have been born over the ponds can grow within a matter of few hours into sizes where they can act as CCN. In this paper I was responsible for partly building the DMPS instrumentation, performing the measurements during the FINNARP 2009 expedition, data analysis and writing most of the paper.

Paper II studies the long-term trends in the quantities related to new particle formation in eastern Lapland, Finland, and the effect of the reduction of large sulphur emissions from Kola Peninsula to these quantities. The study shows how sulphur species have a large effect on the number of NPF event days, the formation time of secondary aerosols and the growth of the particles in eastern Lapland close to the Kola Peninsula smelters. I made the data analysis and wrote most of the paper.

Paper III focuses on aerosol dynamical processes during air masses transportation from ocean to individual measurement stations and between different sites. The paper offers a new Lagrangian way to study the air mass transport from one station to another by clustering the initial size distributions at the upwind station and shows that the aerosol condensational growth is important even in the absence of observable new particle formation. In this paper I was responsible for the meteorological background, contributed writing the paper and produced one figure.

Paper IV is a field study of snow scavenging of sub-micron aerosols. In this paper we also made couple of simple parameterizations that are easy to use in modelling studies. We also applied one of the parameterization into Helsinki University Multicomponent Aerosol model (UHMA) to test the applicability and showed that even with simple parameterizations it is possible to reproduce the aerosol number size distributions during snowfall. In this study I did the data-analysis and wrote most of the paper.
6 Conclusions and future outlook

This thesis focuses on understanding the aerosol processes in the polar regions from their formation until their removal from the atmosphere and, on a broader perspective, their potential effects on clouds and climate. The thesis combines campaign-wise (Paper I) and long-term field measurements (Papers II-IV) of aerosol size distributions from five different measurement stations in the Arctic and Antarctica but also modelling using both back-trajectory calculations (Papers I-III) and University of Helsinki Multicomponent Aerosol model (UHMA) (Paper IV).

This PhD thesis presents several advances in understanding the aerosol processes and aerosol–cloud–climate interactions and thus helps in predicting the future climate and the consequences of the ongoing climate change not only in the polar regions but also globally. More specifically, this thesis presents:

i) First observations of Antarctic NPF from continental biogenic precursors.

The Antarctic continent was previously thought to be free of precursor sources for secondary aerosols. However, during FINNARP 2009 expedition (Paper I), we found that meltwater ponds in mountaneous and blue ice areas and cyanobacteria *Nostoc commune* (Vaucher) living in the ponds and mountaintops are, in fact, a major source for the observed summer-time new particle formation in Antarctica within the distance of few hundreds of kilometers from the coast, both on local and regional scales. Organic vapors evaporating from such ponds were also shown to make particles to grow only in a matter of few hours into sizes where they were able to act as cloud condensation nuclei and thus have climatic effects. This new source of NPF is likely to be important also elsewhere in glaciated areas such as Greenland, northern Canada or Svalbard. Its importance in terms of temporal and spatial scales is expected to increase in the future due to climate change as the ponds thaw earlier and freeze-up later and larger areas experience surface melting in continental ice sheets.

ii) Globally first observations of freshwater vegetation as an aerosol precursor.

Although atmospheric NPF has been observed almost everywhere in the planet in different environments and from various precursor sources, freshwater vegetation has not been observed to emit any chemical components that would take part into NPF. However, in Paper I we observed that cyanobacteria *Nostoc commune* (Vaucher) is likely to be responsible for a large fraction of NPF observed in Antarctica. This cyanobacteria is living in various habitats around the world, both on ground and in freshwater bodies, and is able to survive even in the polar regions. The species emits various organic compounds including *DMS*, which is later oxidized into *H₂SO₄* once in the atmosphere and can take part in NPF. The emitted vapours can thus participate both in the early stages of NPF as well as the subsequent con-
densational growth. This was the first time that freshwater vegetation was identified as a source for aerosol precursor gases.

iii) Throughout understanding of the effect of large sulphur pollution sources to the trends in NPF in the scale of few hundred kilometers.

Sulphuric acid has been known to be the most important component in the atmospheric new particle formation, yet throughout long-term studies on the effect of sulphur pollution to the trends in NPF have not been made until now. The SMEAR I station in Värriö, eastern Lapland, is an ideal, and in fact currently the only place in the world, where such research can be made. The station is close to the smelter industry in Kola Peninsula, and it has one of the longest datasets of aerosol number size distribution measurements in the world. In Paper II, we observed that the number of NPF days in eastern Lapland has been decreasing consistently with the decrease of sulphur pollution originating from Kola Peninsula and that the air masses that pass over Kola favour NPF compared with the air masses that are coming from other areas. Whilst the fraction of NPF days has been decreasing, the decrease in sulphur pollution has lead to lower $CS$, resulting in higher event-time values of $J_3$. In this study, it was shown that the events start earlier after sunrise if the $SO_2$ concentration is high. We also showed that the decrease of sulphur pollution from Kola Peninsula is a result of socioeconomic changes in the area and not due to better filtering or deactivating methods.

iv) A new way of studying the evolution of aerosol number size distribution during air mass transport.

Paper III presents a new Lagrangian way to study the condensational growth of aerosol populations during air mass transport from one station to another. In this approach, the number size distributions from the upwind station are first clusterized into sets of number size distributions similar to each other. Then the different initial conditions generated by the clustering algorithm are compared with number size distributions measured at the downwind station at a time that corresponds to the air mass transport from the upwind station to the downwind station. The transport time is obtained from back-trajectory calculations. Finally, the evolution of the aerosol population can be studied by comparing the initial conditions at the upwind station to the corresponding set of number size distributions at the downwind station. The advance of this approach is that the initial condition is completely independent on any assumptions and the number of initial conditions are not limited.

v) Observation of aerosol condensational growth in the absence of evident NPF.

Paper III shows that even in the absence of observable NPF, the aerosol population, and in particular Aitken mode particles, grow significantly by condensation during summertime in the sub-Arctic region. This result was obtained by studying the evolution of aerosol number size distributions during atmospheric transport with two different methods: 1) by investi-
gating the aerosol population during atmospheric transport from the ocean to the individual stations with different overland transport times and analyzing the changes in the aerosol number size distributions when the air masses were transported from one measurement station to another and 2) by comparing them to the analysis of NPF events. The Aitken mode was observed to grow approximately $0.6 - 0.7 \text{ nm h}^{-1}$ when travelling from the ocean to the individual stations and approximately $0.5 - 1.3 \text{ nm h}^{-1}$ when travelling from one site to another.

vi) First parameterization of snow scavenging in a way that is easily applicable to climate models.

Due to the complex nature of snowfall and the vast varieties in different snowfall types, studying and parameterizing the scavenging of aerosols due to solid precipitation is extremely challenging. In Paper IV we tried to overcome the challenges by choosing snowfalls with as stable conditions as possible to avoid any particle concentration changes caused by other factors than snow scavenging itself. We calculated size-dependent snow scavenging coefficients from approximately 160 hours of continuous, light snowfall and made a computationally easy parameterization into these coefficients. This parameterization was also applied to UHMA (University of Helsinki Multicomponent Aerosol model) and it was proven to simulate well real changes in aerosol number size distribution during snowfall. This parameterization can also be applied to other models that include aerosol dynamics, to be used in estimating the aerosol removal rates due to snowfall.

The polar regions remain untouched and this is how they also should be. This, of course, makes it a challenge to study the atmospheric physics in such areas. The advances presented in this thesis bring new insight into our understanding of the aerosol phenomenology in the remote areas of our planet. However, much is still to be discovered. In the future, more emphasis should be put on understanding of complicated feedback-mechanisms that are related especially to the Arctic amplification and melting of the sea ice, as well as on studying the cryosphere–atmosphere interactions not only in small increments individually, but more efficiently as a whole, from small scale to global scale. Although there is great effort in doing this in the scientific community e.g., in the form of Nordic centres of excellence CRAICC (Cryosphere–Atmosphere–Interactions in a Changing Arctic Climate), DEFROST (Impact of a Changing Cryosphere: Depicting Ecosystem–Climate Feedbacks from Permafrost, Snow and Ice) and SVALI (Stability and Variations of Arctic Land Ice), more co-operation is still needed. Most importantly, the changes in synoptic-scale atmospheric dynamics that are related to the Arctic sea ice melt, and their effect on aerosol transport and removal processes in the Arctic, should be studied more in detail. Also, the new or enhanced sources of anthropogenic and biogenic aerosols, and their precursors, should be studied on a regional scale as now understanding their geographical locations remain to be more on a somewhat theoretical level. Especially, the possibility of meltwater ponds as a significant source for
NPF and subsequent aerosol growth in glaciated areas elsewhere than in Antarctica, should be studied.
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