NEW PARTICLE FORMATION AND ITS CONNECTION WITH CLOUD DROPLET POPULATION IN A REMOTE CONTINENTAL SITE IN NORTHERN FINLAND

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Helsinki, September 2005

Mika Komppula
Abstract

In order to predict the current state and future development of the Earth’s climate system, detailed knowledge on how atmospheric particles from various sources are entered into the atmosphere and how they interact with clouds is needed. This thesis provides new insight into the characteristics and geographical extent of new particle formation events observed in a remote site in Northern Finland during almost four years of continuous measurements. Aerosol-cloud interactions are studied by looking at the activation behaviour of sub-micron size aerosol particles as a function of particle size. Ultimately, direct observational evidence of link between newly-formed particles (after some growth) and cloud formation is presented.

The first part of the thesis concentrates on the local effects on new particle formation studied at Pallas (paper I) and takes a look to more regional scale new particle formation combining events observed at Pallas and at Värriö (papers II and III). Also an approach to model new particle formation in air mass transported between the two sites is presented (paper III).

In the second part of the thesis an attempt was made to link new particle formation to the formation of cloud condensation nuclei (CCN). This was done by investigating the growth of newly formed particles to CCN sizes and their contribution to CCN population, and ultimately their connection with cloud droplet formation. Measurements of cloud droplet activation of aerosol particles were carried out in Pallas site located in Northern Finland. Size-segregated activation efficiencies of sub-micron size aerosol particles were evaluated from DMPS measurements. Finally, direct evidence of link between new particle formation and cloud formation was presented.

This study indicates that atmospheric new particle formation has to be taken seriously when estimating the climatic effect of aerosol particles. New particle formation can occur simultaneously over large geographical scales. Evidence on large numbers of newly formed particles growing to CCN sizes was obtained. Particles down to about 50 nm in diameter were observed to activate into cloud droplets and therefore the role of ultrafine particles (<100 nm) in this activation process seems to be very important, at least in these clean or moderately polluted environments.
List of publications

This thesis consists of an introductory review, followed by six research articles. Papers are reproduced with the kind permission of the journals concerned.


1 Introduction

Atmospheric aerosol is defined as a mixture of liquid or solid particles suspended in the air. These particles are present everywhere, with number concentration varying from few cm$^{-3}$ in remote areas up to $10^6$ cm$^{-3}$ in heavily polluted urban areas. Their size varies from molecular clusters (~1 nm) to hundreds of micrometers in diameter. Aerosol particles can be emitted to atmosphere directly by an emission source as primary particles or they can be formed through gas-to-particle conversion as secondary particles.

Formation and subsequent growth of atmospheric aerosol particles have recently been an active research topic (Kulmala et al., 2004a). Over the past decade, observations of the formation and growth of nanometre-size atmospheric particles have been reported in many different environments around the world. New particle formation has been shown to be a notable source of Aitken mode particles and therefore a potential source of cloud condensation nuclei (CCN).

Atmospheric aerosol and clouds have a mutual influence on each other. Aerosol particles modify many climatically important cloud properties, including the cloud reflectivity and lifetime (Rosenfeld, 2000; Norris, 2001; Harshvardhan et al., 2002; Kruger and Grassl, 2002; Lohmann, and Feichter, 2005). Clouds, in turn, affect aerosol particle populations by altering their chemical composition, by enhancing their activation efficiencies in subsequent cloud encounters, by transferring ultrafine particles into optically more active sizes, and by removing particles from the atmosphere by precipitation (Choularton et al., 1998; Flynn et al., 2000; Krämer et al., 2000, O’Dowd et al., 2000; Kerminen, 2001; Andronache, 2003; Kerkweg et al., 2003).

Aerosol-cloud interactions are crucially dependent on which fraction of the aerosol population activates into cloud droplets and which fraction remains as cloud interstitial particles. Despite the recent theoretical advances made in understanding and simulating cloud droplet activation (Kulmala et al., 1993; Chuang et al., 1997; Liu and Daum, 2000; Charlson et al., 2001; Abdul-Razzak et al., 2002; Feingold et al., 2002; Nenes and Seinfeld, 2003), we are still unable to predict the exact division between cloud droplets and cloud interstitial particles in the real atmosphere (Snider et al., 2003). One reason for this is the lack of accurate field data on cloud droplet activation, especially when it comes to ultrafine particle size range that is relevant to clouds forming in clean or moderately polluted environments.

In order to predict the current state and future development of the Earth’s climate system, detailed knowledge on how atmospheric particles from various sources interact with clouds is needed.
This thesis provides an insight to characteristics and geographical extent of new particle formation events observed in a remote site in Northern Finland. The aerosol-cloud interactions are studied by the activation behaviour of sub-micron size aerosol particles as a function of size. Ultimately, direct observational evidence of link between newly-formed particles (after some growth) and cloud formation is presented.

This study had two main goals:

1) To characterize new particle formation in a subarctic area in Northern Finland (Papers I–III), including
   - frequency of occurrence and seasonal/diurnal variation
   - effects of local surrounding and altitude
   - effects of the air mass type and meteorological conditions
   - geographical extent of new particle formation events.

2) To study aerosol-cloud interactions (Papers IV–VI), including
   - investigating the growth of newly formed particles to CCN sizes and their contribution to CCN population and connection with cloud droplet formation
   - activation behaviour of particles as a function of size
   - connecting atmospheric new particle formation to cloud droplet activation.

2 Methods

This thesis is mainly based on the continuous DMPS (Differential Mobility Particle Sizer) measurements conducted at Pallas in Northern Finland (Figure 1). In papers II and III a look to geographical extent of new particle formation was done by comparing the data with the similar measurements conducted in Värriö, 250 km east from Pallas. This section presents the measuring sites, the instrumentation used and the methods used for data evaluation.
2.1 Measuring sites

Finnish Meteorological Institute's (FMI) measurement station in Pallas is part of the Pallas-Sodankylä Global Atmosphere Watch (GAW) station located in Northern Finland (Figure 2). Aerosol measurements are carried out mostly at the Sammaltunturi station (67°58’ N, 24°06’E; Figure 3), which is the main station of the four measuring sites in Pallas. The Sammaltunturi station resides on a top of the second southernmost fjeld (an arctic round-topped treeless hill) in a 50-km-long north and south chain of fjelds at an elevation of 565 m above sea level (a.s.l.), and ca 300 m above the surrounding area. The highest fjelds in the chain are located 600 to 800 m a.s.l. The timberline lies about 100 m below the Sammaltunturi station. The surrounding forest is mixed pine, spruce and birch. The vegetation on the fjeld top is sparse, consisting mainly of low vascular plants, moss and lichen. Due to its elevation, the Sammaltunturi station is from time to time within the cloud cover. The station is within the cloud cover for at least part of the day in about 10 % of all days. The other station, Matorova (68°00’N, 24°14’E), is about six kilometres
northeast of Sammaltunturi at an elevation of 340 m a.s.l. The station lies in a forested area. The stations are located inside the Pallas-Ounastunturi National Park (total area 501 km$^2$), in a subarctic region near the northern limit of the boreal forest zone. A more detailed description of the Pallas area and its infrastructure can be found in Hatakka et al. (2003).

![Figure 2. Location of the measuring stations at Pallas area.](image)

One of the main criteria for selecting the Pallas area as the site for these measurements was the absence of large local and regional pollution sources. The distance to the nearest town, Muonio, with some 2500 inhabitants, is 19 km to the west. The closest major sources of pollutants are the smelters in Nikel and Montshegorsk in Russia, located more than about 350 km away from Pallas, Nikel to the northeast and Montshegorsk to the east.

The Värrö SMEAR I environmental measurement station (67°46’N, 29°35’E), managed by the University of Helsinki, is located 250 kilometres away and almost directly to the east of Pallas (Hari et al., 1994; Ahonen et al., 1997) (see Figure 1). The station is near the Russian border at the height of 390 m a.s.l. The station is located just below the alpine timber line, which is at about 400 m a.s.l. The main tree species in the area is Scots pine. The station area belongs to the Värrö National Park. There are practically no local sources of pollutants close to the station. The closest
major pollutant sources are the smelters in Nikel and Montshegorsk in Kola Peninsula, Russia, located at 200 kilometres to the north and 140 kilometres to the east of the Värriö station, respectively.

![Figure 3. A picture of the main station at Pallas area, Sammaltunturi.](image)

### 2.2 Instruments

Similar DMPS (Differential Mobility Particle Sizer) systems are used at both Pallas stations measuring sites for nanometre particle sizing. The DMPSs consist of a 28-cm-long Hauke-type differential mobility analyzer (DMA) (Winklmayr et al., 1991) with a closed loop sheath flow arrangement (Jokinen and Mäkelä, 1997) and a condensation particle counter (CPC), TSI model 3010. The closed loop sheath air is dried with silica gel dryer and the relative humidity is kept below 20%. This sheath air setup also keeps the temperature more constant. The sheath air relative humidity, temperature and pressure are monitored continuously. The sheath air flow rate is about 11 litres min\(^{-1}\) and the sample aerosol flow rate is 1 litre min\(^{-1}\). Before sizing, the aerosol is neutralized with a radioactive source (370 MBq Ni-63 beta source), to provide a known charge distribution for the aerosol. The measured particle size range is from 7 to 500 nm, which is divided into 30 discrete bins. It takes around five and a half minutes to measure one particle size spectrum.
Table 1. Measurement programmes at Sammaltunturi station (list from year 2003).

<table>
<thead>
<tr>
<th>Programme</th>
<th>Instrument or measurement method</th>
<th>Sampling frequency</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Gases and aerosols</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Surface O3</td>
<td>Two Dasibi ozone meters</td>
<td>Continuous</td>
</tr>
<tr>
<td>SO2</td>
<td>Thermo Environmental Instruments 43 S</td>
<td>Continuous</td>
</tr>
<tr>
<td>CO2, N2O, CH4</td>
<td>Glass flask samples</td>
<td>1/week</td>
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<tr>
<td>CO2</td>
<td>Li-Cor infra-red gas analyser</td>
<td>Continuous</td>
</tr>
<tr>
<td>VOCs</td>
<td>850-mL stainless steel flasks</td>
<td>2/week</td>
</tr>
<tr>
<td>Condensation nuclei</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Da &gt; 10 nm</td>
<td>TSI CPC model 3010</td>
<td>Continuous</td>
</tr>
<tr>
<td>Da &gt; 0.3 µm</td>
<td>TSI Laser Particle Counter</td>
<td>Continuous</td>
</tr>
<tr>
<td>Particle size distribution 7-500nm</td>
<td>DMPS</td>
<td>Continuous</td>
</tr>
<tr>
<td>Aerosol scattering coefficient</td>
<td>TSI Nephelometer 3560</td>
<td>Continuous</td>
</tr>
<tr>
<td>Black carbon (BC)</td>
<td>Aethalometer (Magee Scientific)</td>
<td>Continuous</td>
</tr>
<tr>
<td>Radon-222</td>
<td>Aerosol beta activity measurement</td>
<td>Continuous</td>
</tr>
<tr>
<td>Radon-222</td>
<td>Accumulation chamber, alfa counting (with EML)</td>
<td>Continuous</td>
</tr>
<tr>
<td><strong>Solar radiation</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Global irradiance</td>
<td>Kipp &amp; Zonen pyranometer CM11</td>
<td>Continuous</td>
</tr>
<tr>
<td>J(NO2)</td>
<td>Radiometer (Meteoroloogie Consult GmbH)</td>
<td>Continuous</td>
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<tr>
<td><strong>Surface weather parameters</strong></td>
<td></td>
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<td>Vaisala MILOS500+sensors</td>
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<td>Continuous</td>
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</table>
In Värriö, the size distribution is measured with similar setup as in Pallas. The measured size range (7–500 nm) was divided to 20 bins until April 26, 2003. After that, the size distribution measurements were extended to cover a size range from 3 nm to 500 nm with 24 size bins. This was done by using a Twin-DMPS system (e.g. Kulmala et al., 2001a).

At Sammaltunturi station there is also a Condensation Particle Counter (TSI 3010) measuring the total aerosol number concentration. It gives a good comparison and monitoring value for the DMPSs. The aerosol scattering and backscattering coefficients were measured with an Integrating Nephelometer (TSI model 3563). It measures the scattering and backscattering coefficients $\sigma_{sp}$ and $\sigma_{bsp}$ at three wavelengths: 450, 550 and 700 nm. Aerosol black carbon (BC) concentration, or optical absorption, is measured with an Aethalometer (Magee Scientific). The station is equipped with an automatic weather station (Vaisala, Milos 500) and present weather sensor (Vaisala, FD12P). The present weather sensor measures visibility (up to 50 km) and type of precipitation using light scattering. Concentrations of gaseous CO$_2$, SO$_2$, O$_3$, NO and NO$_X$, as well as the global solar radiation, are continuously monitored. Table 1 presents a list of measurement programmes at Sammaltunturi station in year 2003.

At Sammaltunturi station, all the air samples into the instruments are taken from one main sampling line. The inlet of the main sampling line is about 7 meters above the ground and about 3 meters above the roof of the station building. The inner diameter of the stainless steel inlet nozzle and the main sampling line is 56 mm. The flow rate in the main sampling line was $90 \text{ m}^3 \text{ h}^{-1}$ until June 17, 2003. From there on the flow rate was $155 \text{ m}^3 \text{ h}^{-1}$. The inlet nozzle is H-shaped, leading to two downward and two upward orientated inlets with a flow rate of one fourth of the total flow through each. The outer surface of the inlet is heated about 1–2 °C above zero to avoid the build up of ice and snow. Because the residence time of the sample air in the H-shaped nozzle is <0.1 s, the effect of heating on the temperature of the sample air or size of aerosol particle is negligible. There is no special rain protection for the inlet due to the problem with ice and snow. The nozzle is followed by about five meter long vertical line. After this, there is a T-connector, in which the sample air makes a 90° turn into the station building. The other branch, directed downwards and sealed at the end, collects possible condensed water. This is also the point where almost all the particles larger than 10 μm are separated from the sample air by inertia. The water collector is heated to avoid freezing. The heating does not affect the sample air. The connector is followed by about four meters of horizontal tube with two 90° turns before the sample air arrives to the point, from which it is drawn into the aerosol instruments. The tubing inside the station building is
insulated to avoid the condensation of water during summer. The diameter of 50 % particle transmission efficiency with this arrangement was calculated to be about 7 μm for ambient air conditions and with an average wind speed (6.4 m s⁻¹) in the data analyzed here. This diameter was calculated to vary slightly, by 1–2 μm, with wind speed extremes. The calculations were made according to the equations presented in Baron and Willeke (2001). At Matorova station the air sample was drawn straight to the DMPS through stainless steel tube with 4 mm inner diameter about half a meter away from the station building outer wall and about 2 m above the ground.

At both stations the sample air was heated to station indoor temperature (about 20 °C) before it entered the aerosol instruments. The average outdoor temperature during the cloud events, studied in paper V, was about −5 °C and the highest temperature about 9 °C. In addition, the sheath air of the DMPS was kept below 20 % relative humidity (16 % on the average) and temperature above 20 °C (21.2 °C on the average). These facts ensured that the particle dry diameter was measured at both the sites.

2.3 Data analysis

The DMPS data used in this thesis covers the period from April 2000 to December 2003 for both Pallas and Värriö sites. Simultaneous measurements at the two Pallas stations, Sammaltunturi and Matorova, cover the period from April 2000 to February 2002. This section presents the main objectives in the DMPS data inversion, how to identify a new particle formation event, and gives an introduction to modelling setup used paper III. Finally, methods used in CCN and aerosol-cloud interaction calculations in papers IV and V are presented.

2.3.1 Data inversion

Data analysis and quality control of the Pallas DMPS data was performed at the FMI. The data inversion was based on the routines and programs developed at the University of Helsinki (Aalto et al., 2001). The routines for further analysis involving cloud droplet activation and CCN calculations were made at the FMI. The raw data of the DMPS includes concentrations for a certain voltage. In order to get the particle size distribution dN/dlogDp from the raw data, an inversion has to be done. One has to know the charge distribution after the neutralizer, DMA transfer function and detection efficiency of the CPC. Also losses in the sampling lines must be taken into account. Charge distribution of Wiedensohler (1988) and DMA transfer function of
Stoltzenburg (1988) were used in the data inversion. The whole DMPS-systems were calibrated with monodisperse Ag and PSL particles prior to installation at Pallas.

2.3.2 Identification of particle formation events

A new particle formation event can be visualized with a surface plot (see Figure 4), in which the sub-micron particle size distribution is presented as a function of time (Mäkelä et al., 1997; Kulmala et al., 1998). An event is seen as increasing particle concentrations in the smallest size channels of the DMPS system. On a typical particle formation day, newly-formed particles enter the measurement range at around midday at initial sizes below 15 nm. During the rest of the day a subsequent growth of these particles is seen at a rate of a few nanometres per hour. Classifying between a particle formation event and non-event is sometimes difficult and somewhat subjective. The classification method applied here has been explained in more detail by Mäkelä et al. (2000).

Figure 4. A new particle formation event observed at Sammaltunturi, Pallas, on August 29, 2002.
Events were classified into three classes from 1 to 3 according to their intensity and the distinctness of growth. The best ones, showing a clear formation of small particles and clear subsequent growth, were classified as class 1 events (e.g. Figure 4). Class 2 events do not have such intense formation of new particles or particle growth as class 1 events. In class 3 events some new particles are formed, but no growth can be seen or it is weak. However, class 3 events still have some indications of particle formation. Because of the subjectivity of the classification some overlapping within the classes may occur. In detailed analysis, only class 1 and 2 events were considered.

The particle formation rate (particles cm$^{-3}$ s$^{-1}$) and particle growth rate (nm h$^{-1}$) were estimated from the size distributions. The formation rate of 7 nm particles ($J_7$) is a quantity calculated from the observations, which refers to how many new particles appear in the measurement range (starting at 7 nm) during a formation event. $J_7$ is determined as,

$$J_7 = \frac{dN_7}{dt} + K_7N_7,$$  \hspace{1cm} (1)

where $dN_7/dt$ is actually the observed formation rate on 7 nm particles and $N_7$ is the number of nucleation mode particles (>7 nm). $K_7$ is the typical coagulation sink during the growth and $t$ is the particle growth time. Particle formation days were mainly examined with a program made for the purpose at the University of Helsinki (Mäkelä et al., 2000). This program calculates the apparent formation rate of 7 nm particles and the growth rate. The starting and ending times of the events and upper diameter limit for the event mode (maximum new particle diameter) for every hour from the start of the event were manually determined from the one-day spectrum. Defining the starting and ending times of an event is sometimes difficult due to fluctuation in the smallest size classes due to measurement uncertainties. Furthermore, the total number concentration of new particles formed during the event was calculated. The number concentration of pre-existing particles affecting the condensation sink just before the formation event was also calculated. The condensation sink determines how rapidly molecules condense onto pre-existing aerosols and depends strongly on the shape of the particle size distribution (Pirjola et al., 1999). The condensation sink (CS) is $4\pi D$ CS', and CS' is integrated over the aerosol size distribution:

$$CS' = \int_0^\infty r \beta_M (r)n(r)dr = \sum_i \beta_M r_i N_i.$$  \hspace{1cm} (2)
Here, $D$ is the diffusion coefficient and $\beta_M$ is the transitional correction factor by Fuchs and Sutugin (1971). A detailed calculation procedure for the condensation sink has been presented by Kulmala et al. (2001a). The condensable vapour concentration required for the observed growth during each event day was estimated. Also the source rate of condensable vapour required to sustain the concentration needed for the observed growth was estimated as presented by Kulmala et al. (2001a).

It has been suggested that 1 nm is the size of a thermodynamically stable clusters, which under certain conditions grow to detectable sizes (Kulmala et al., 2000). For this reason, the approximate starting time of the formation of 1 nm particles was calculated. This was made to estimate the starting time of the nucleation process and, for example, how close to the sunrise nucleation starts. Calculations were made backwards in time with the estimated growth rate. Although Kulmala et al. (2004b) have found that particles below 7 nm in diameter have smaller growth rate compared to larger particles, our estimate should be reasonable because of the quite stable growth rates observed during the events at the Pallas site.

2.3.3 A modelling approach

The size-segregated aerosol dynamics model UHMA (University of Helsinki Multicomponent Aerosol model) (Korhonen et al., 2004) can be used for Lagrangian studies of the tropospheric aerosol particles. The model includes the major clear sky processes: multicomponent condensation of organic and inorganic species (Grini et al., 2005), coagulation, new particle formation through nucleation and primary emissions, dry deposition as well as a simple treatment of entrainment and detrainment. Recently, the model has been extended to treat cloud processing in non-precipitating clouds (Korhonen et al, 2005).

In the current study, the model was applied to two case studies to investigate nucleation and particle growth in air masses travelling from Pallas to Värriö measurement site. The air mass trajectory information was obtained from NOAA ARL trajectory model Hysplit 4 (Draxler and Rolph, 2003). The meteorological conditions (temperature, pressure, relative humidity, boundary layer height) for the model were interpolated from the calculated trajectory data. The particle size distribution and SO$_2$ concentration measured at Pallas were used as model input and the modelled results were compared to similar measurements made at Värriö. The kinetic nucleation rate was
assumed in simulations and different production rate profiles of condensable organics were tested.

2.3.4 Growth of particles to CCN sizes

In Paper IV, new particle formation events were used to link particle formation and CCN. The criteria for selecting the events were that the event was very clear and that the particle formation was followed by a relatively steady particle growth of at least 12 hours in duration (see Figure 5). A CCN formation event was judged to have ended when the particle growth was stopped, or when it was interrupted by either a clear change in the air mass origin or by cloud droplet activation. Changes in the air mass origin could be identified from sudden changes in the particle size distribution spectrum or from increases in the black carbon (BC) concentration and particle scattering coefficient caused by anthropogenic pollution. The site is relatively frequently within a cloud (at least part of the day in about 10% of all days), which could be seen directly from the present weather sensor (visibility) data. A cloud occurrence could also be observed straight from the measured size distribution spectrum as a loss of particles in the largest size classes.

Figure 5. An example of CCN formation event starting on August 3–6, 2001.
Quantitative determination of a CCN concentration as a function of water vapour supersaturation (CCN spectrum) is not possible without direct CCN measurements or without detailed information on the aerosol chemical composition as a function of particle size. Since such data were not available, we had to rely on some surrogate measure for the actual CCN concentration. Two quantities, CCN$_{50}$ and CCN$_{80}$, namely the concentration of particles larger than 50 and 80 nm in dry diameter, respectively, were selected to represent the number of particles that are “potential” CCN. This selection can be considered justified for several reasons. First, the critical supersaturations needed to activate particles at the size range 50–80 nm (0.2–0.5 % for dry ammonium sulphate particles (see Seinfeld and Pandis, 1998)) are close to those estimated for clean and moderately polluted clouds (e.g. Hudson and Yum, 2002; Yum and Hudson, 2002). Second, the minimum dry diameter of particles forming cloud droplets is usually between about 50 and 100 nm under clean conditions (e.g. Covert et al., 1998; Schwarzenboeck et al., 2000; Twohy et al., 2001; Henning et al., 2002; Lowenthal et al., 2002; Glantz et al., 2003; Paper V). Third, observations at our site have demonstrated that major part of particles >80 nm in diameter became incorporated into cloud droplets when the site was within a cloud (Papers I and V).

2.3.5 Aerosol-cloud interactions

The existence of a cloud was identified as a sudden drop in the particle number concentration corresponding to the largest size classes of the DMPS at the in-cloud station (Figure 6). Simultaneously, a drop in the visibility was seen. To verify the existence of a cloud, weather and other related data were examined. A decrease in aerosol light scattering coefficient and an increase in the relative humidity were clear indications of the presence of a cloud. A web-camera, taking still-photos in 0.5–3 hour intervals, was also helpful in determining the cloud. The presence or absence of rain was identified from weather data.

The selection procedure for cloud events was started by checking the DMPS data day by day. If a sudden decrease in the number concentration of largest particles was observed at Sammaltunturi the day passed the first stage. After that a comparison with the reference DMPS data at Matorova was done to verify that a difference in the size spectra between these two sites existed. From weather data it was then checked whether a rapid increase in the relative humidity close to 100 % (>98 %) and a sudden drop in the visibility close to zero (<200 m compared with a typical value of about 50 km) was found at Sammaltunturi. Finally, the web-camera photo was checked to ensure the cloud occurrence. If all these criteria were fulfilled, the day was classified as a cloud.
event day. After this all rainy days were left out. This was done because during the rain, falling rain drops remove unactivated aerosol particles, which would cause errors in activation calculations. Out of the remaining days, all days with fluctuations in the DMPS data during the cloud time were ruled out. We are confident that this procedure ruled out the days with thin clouds (very probably with fluctuations in the DMPS data) and mixed-phase clouds with very rough and varying interstitial particle size distribution as seen in Henning et al. (2002). This very critical selection procedure decreased the number of cloud events from 200 in the beginning to only 50 remaining for this study. An uncertainty estimate was calculated from the concentration difference between the sites before the cloud appeared to confirm that the air masses were homogenous on both sites (paper V).

Figure 6. An example of a cloud droplet activation event, October 15, 2000. In a) in-cloud particle size distribution, b) total particle concentration in- and out-of-cloud, c) out-of-cloud particle size distribution and d) visibility.

The size distribution of activated particles (cloud residual particles) was deduced indirectly by subtracting the in-cloud (Sammaltunturi station) DMPS size spectra from the corresponding out-of-cloud (Matorova station) size spectra (Figure 7). From the same data, the fraction of activated...
particles as a function of size could be calculated, including the diameter corresponding to 50% activation efficiency ($D_{50}$) (see Figure 7). The number of activated particles and the activated fraction were also estimated separately for different modes. All these comparisons and estimations were done for average values during the whole cloud event. Numerous weather-related and other parameters (temperature, temperature lapse rate, wind, sun radiation, relative humidity, absolute water vapour concentration, visibility, rain, pressure, air mass origin and concentrations of $SO_2$, NO, $NO_2$ and $O_3$) were studied for correlation analysis to see their effects on cloud droplet activation.

![Graphs showing data analysis procedure](image)

**Figure 7.** Two examples of the data analysis procedure: (a-b) typical continental air masses on September 18, 2001 and (c-d) typical arctic air masses on November 6, 2001. Figures 7a and 7c present the spectra out-of-cloud, in-cloud and the spectra of activated particles (out-of-cloud minus in-cloud size spectra). Figures 7b and 7d present the activated fraction in each size class.

One critical assumption that was made was that all particles larger than 7 µm in diameter are cloud droplets. This had to be assumed because of the aerosol inlet cut-off diameter of 7 µm. It should be noted that we did not measure the cloud in a lagrangian sense. The cloud was moving and the measurements were made in one fitted location on the ground. This forced us to make the assumption that the cloud was homogenous by its physical properties. The drift of the cloud
makes it difficult to estimate any changes that the cloud droplet population, or the cloud droplet residual particle size distribution, would experience during the cloud evolution. However, a comparison of the DMPS evaluated and directly measured (Forward Scattering Spectrometer Probe, FSSP) droplet concentrations was performed. The results were surprisingly similar which verifies the used method to be functional (see Figure 8).

Figure 8. Comparison of cloud droplet number concentration evaluated from the DMPS data (red stars) and counted by FSSP (blue circles). Two example days, October 30, 2004 and November 7, 2004.

3 Atmospheric new particle formation in a subarctic area

This chapter presents the characteristics of new particle formation and particle number concentration observed in Northern Finland. The chapter is divided to two parts: the first concentrating on the local effects on new particle formation studied at Pallas (paper I) and the second part takes a look at more regional scale new particle formation, combining events observed at Pallas and at Värriö (papers II and III). In the second part, an approach to model new particle formation in air mass transported between the two sites is also presented (paper III).

3.1 Local scale new particle formation and particle concentration

Basic characteristics of particle number concentration and new particle formation events observed at Pallas are presented. Local scale new particle formation and the effect of surrounding
area are studied between two sites, Sammaltunturi and Matorova, in different surroundings and at different heights at Pallas area.

3.1.1 Overall characteristics

In total, 65 particle formation events were recorded at Pallas during the period of 22 months (Apr 2000-Feb 2002). The formation events were not as frequent and intense as seen in the measurements in Southern Finland. In Hyytiälä 50–100 events per year has been observed (Mäkelä et al., 1997; Kulmala et al., 1998; Mäkelä et al., 2000). The low number and low intensity of events made the event classification somewhat difficult. Event days were divided into three classes. Nine event days were classified as class one events, 13 as class two events and 43 as class three events. In further analysis, only class one and two events are used because they are the most obvious ones.

The largest number of particle formation events occurred in April and May. Another slight peak was observed in August. In winter, practically no particle formation events were detected. The particle formation season from April to September is shorter than what has been observed in Southern Finland, where particle formation is already occurring frequently in early March (Mäkelä et al., 2000).

The starting times for the formation of 7 nm particles varied between 8:25 and 15:50 local time (UTC+2), whereas the calculated starting times of 1 nm particle formation varied between 4:50 and 14:20. The length of the events varied from about 3 hours to 9.5 hours with an average of about 5 hours. The particle growth rate values varied in the range 1.4–8.2 nm h$^{-1}$. The formation rate of 7 nm particles varied in the range 0.06–0.40 particles cm$^{-3}$ s$^{-1}$. These events produced new particles between 1100 and 4000 cm$^{-3}$. Condensation sink varied in the range 0.8–17.0×10$^{-4}$ s$^{-1}$. The pre-existing particle number concentration at the start of the events showed a large variation, being 70–1000 cm$^{-3}$. The observed pre-existing particle number concentrations during event days were considerably below the monthly average of the month in question.

The formation events at Pallas started within a maximum of 8.5 hours after the sunrise. The start time of formation events did not depend on the time of the sunrise, although the presence of solar radiation was clearly necessary. No events were observed during dark hours. The minimum global solar radiation intensity on the time of formation was 220 W m$^{-2}$ and the average was 450 W m$^{-2}$. For comparison, the average solar radiation was 80 W m$^{-2}$ for the two-year period and
160 W m\(^{-2}\) for the main particle formation season (1 Apr–15 Sep). In Hyytiälä, Southern Finland, particle production was not observed when global solar radiation was below 400 W m\(^{-2}\) (Aalto et al., 2001).

The ambient one-hour average air temperature before the particle formation events varied between –13.0 °C and 13.5 °C depending on the season. The mean temperature on the particle formation days was 2.3 °C, which is significantly lower than the average temperature for the whole particle formation season (1 Apr–15 Sep) of 5.8 °C. The lower temperature on formation days indicates colder polar or arctic air masses from the north. The air mass trajectory analysis supports this. This is the usual air mass pattern on particle formation days observed also in Southern Finland (Mäkelä et al. 2000; Kulmala et al., 2001a). The wind speed did not show any difference between the formation days and other days. The relative humidity was observed to be a little lower on average on formation days, although some high relative humidity values were also found. The average relative humidity on formation days was 74.9 % and for the whole formation season slightly below 80 % at Sammaltunturi.

The weather data showed a temperature rise of 2 to 6 °C to be a normal pattern before or during the start of the formation event. Also an increase of solar radiation before the event was observed. Some variation in solar radiation, i.e. variable cloudiness, was observed during most event days. The visibility on the formation days was usually high (over 30 km). On event days, the relative humidity was usually observed to take a large dive of 10–40 %. On most occasions there was no precipitation observed during the whole day. Low levels of SO\(_2\), NO and NO\(_2\) concentrations during particle formation days indicate also clean polar or arctic air masses. These concentrations were most of the time below the detection limit of the instruments, especially on formation days.

### 3.1.2 Influence of local surroundings

The influence of local surroundings on new particle formation and particle concentration was studied by comparing the data from the two Pallas sites six kilometres apart from each other. The Sammaltunturi station is on the top of a treeless hill while the Matorova station is surrounded by a forest.

All the events started at almost the same time in the two measuring sites, within 30 minutes of each other (e.g. Figure 9). On average the starting times were equal though some day-to-day variation was seen. No event was found that would occur only at one of the sites. Basically all the
parameters presented in section 3.1.1 were very close to each other between the sites. The average number of new particles produced during the particle formation events was slightly higher at Matorova (2700 cm$^{-3}$) compared with Sammaltunturi (2360 cm$^{-3}$). Also the particle growth rate was, on average, slightly higher in Matorova (4.0 nm h$^{-1}$) than in Sammaltunturi (3.7 nm h$^{-1}$). The effect of surroundings to new particle formation between these two sites was found to be insignificant.

![Figure 9](image)

Figure 9. A simultaneous event observed at Sammaltunturi and at Matorova six kilometres apart from each other, on August 8, 2001.

When studying the small time shift in the starting times of formation events between the stations, wind speed and direction were used to calculate the time for an air parcel to travel from one station to the other. This was carried out to find out if newly formed particles could have been transported in horizontal direction from one station to another. Also the possibility of different growth rates between the stations was checked. Only about 25 % of the events could be reasonably explained using these features. This leaves the vertical movement of the air for the most likely explanation. It has to be noted that the starting times of the events are manually
picked from the daily surface plot, and that one measurement cycle takes as long as five and a half minutes to measure. These two factors are causing some error to the starting times, but they still do not fully explain the observed differences.

In Southern Finland it has been observed that during formation events particle concentration is not increasing equally at different heights, and it has been suggested that particles are not formed in the canopy level but higher (Aalto et al. 2001). Earlier studies in Southern Finland also exclude free troposphere as a possible origin of the new particles and assume that the new particles are originating somewhere between the canopy layer and free troposphere (Nilsson et al., 2001). One explanation for the starting time differences could be vertical scale variations in particle formation. Particle formation could be occurring at different height levels in different days and then spreading by turbulent mixing towards other vertical layers.

The annual average particle number concentration measured at Sammaltunturi was 20 % lower than what was observed at Matorova, being 700 cm\(^{-3}\) and 870 cm\(^{-3}\), respectively. A great deal of this difference was caused by the higher altitude of the Sammaltunturi station. It was observed that from time to time Sammaltunturi is within the cloud cover. In low cloud cover days particles larger than ca. 80 nm on average are activated as cloud condensation nuclei and are growing rapidly outside the detection limit, which lowers the number concentration at the measured range (see Figures 6 and 7). On clear conditions the size distributions are almost identical at both sites. Overall, this finding led to a more detailed cloud droplet activation studies presented later in papers IV, V and VI. In general, the Pallas area has lower particle number concentrations compared with those in Southern Finland due to the dominance of relatively clean air. In Southern Finland at the Hyytiälä station, the yearly-average particle number concentration (2000 cm\(^{-3}\)) is more than twice that in Pallas (e.g. Laakso et al., 2003).

At Pallas a clear seasonal variation in the particle number concentration was observed, with high values in spring and summer and lower values in winter. This variation is similar in both the stations, as expected. A maximum one-hour average particle number concentration of 9290 cm\(^{-3}\) was found in May 2000 at Matorova and minimum of 10 cm\(^{-3}\) in late October 2000 at Sammaltunturi. Generally speaking, in spring and summer the daily average number concentration may rise up to 3500 cm\(^{-3}\), whereas in winter the daily average may drop well below 100 cm\(^{-3}\). Modal concentrations were compared using fixed mode size limits: nucleation mode (7–25 nm), Aitken mode (25–95 nm) and accumulation mode (95–500 nm). All modes have their minimum concentration during winter when total concentrations were low and no nucleation
events occurred. The nucleation mode had its highest concentrations during April and May when most of the new particle formation events took place. Nucleation mode average concentrations in 2001 were very similar at both stations. In Aitken mode the monthly average concentrations were a bit higher at Matorova compared to Sammaltunturi almost throughout the whole measuring period, the yearly averages being 420 cm\(^{-3}\) and 350 cm\(^{-3}\), respectively. In accumulation mode the concentration difference between the stations was the largest. The average accumulation mode concentrations in year 2001 were 320 cm\(^{-3}\) at Matorova and 200 cm\(^{-3}\) at Sammaltunturi. The occasional low cloud cover at Sammaltunturi decreased the accumulation mode and partially also the Aitken mode concentrations (see Figures 6 and 7).

In the diurnal variation of particle concentration, no difference between the stations was found. The diurnal variation of the accumulation and Aitken mode was low. Accumulation mode had a minimum at around 07:00 and maximum around 15:00 from spring to autumn. The Aitken mode had a slight minimum around 10:00 and maximum around midnight in summer and autumn. During winter no diurnal variation was seen. Most of the diurnal variation in nucleation mode particle number concentrations was seen in spring and early summer caused by the particle formation events occurring always within few hours around noon. The nucleation mode had its minimum around 09:00 and maximum around 18:00, as did the total number concentration. It was clearly seen that high variation occurs in spring (April and May), whereas low concentration and variation prevail in winter (November). The diurnal maximum peak of nucleation mode particle concentration is getting earlier, when moving towards summer. This is probably due to earlier sunrise, meaning larger amount of solar radiation. However, the time of the minimum value stays at around 09:00. This is a combination of higher nucleation rate, shorter duration of formation events and higher growth rate of newly formed particles.

### 3.2 Regional scale new particle formation

The regional scale of new particle formation is studied by combining the events observed at Pallas and at Värrö 250 km apart from each other. The effects of different air masses (continental vs. marine) are studied. Seasonal variations in observed new particle formation event parameters are presented. An approach to model new particle formation in an air mass transported between the two sites is presented. The idea was to find out whether the aerosol particle physical
properties observed at the Värriö station can be generated from those observed at the Pallas station, or if something is taking place which we can not explain.

### 3.2.1 New particle formation in Northern Finland

In total, 87 classified event days were observed at Värriö and 71 event days at Pallas during the 21 month measuring period (Apr 2000–Dec 2001). The largest number of events was observed in spring (April–May). Another peak was observed in late summer, in August and September. Among these, only the days that had class 1 or 2 event occurring at one of the stations and a clear event (classes 1–3) or non-event at the other, were taken into further inspection (41 days). At Värriö 35 and at Pallas 29 event days filled these selection criteria. On 23 of these days new particle formation was observed at both stations. On 12 days the event was observed only at Värriö (see Figure 10) and on 6 days only at Pallas.

![Figure 10. A typical polluted new particle formation event observed only at Värriö site, on March 19, 2001.](image-url)
First we provide a short overview of parameters related to nucleation events during those 23 days at which particle formation events were observed at both sites. The largest differences and variation were seen in the starting times of the events. The difference between the two sites varied from 0 to 5.2 hours. The average starting time was 12:10 at Värrö and 12:50 at Pallas. This 40-minute difference compares somehow with the fact that at Värrö sun rises 18 to 23 minutes earlier than at Pallas. On average, the number of pre-existing particles before the event was almost double at Värrö when compared to Pallas. This leads to higher condensation sinks (averages $8.2 \times 10^{-4}$ s$^{-1}$ at Värrö and $4.8 \times 10^{-4}$ s$^{-1}$ at Pallas) and higher vapour sources needed for a similar particle growth. The average formation rate of 7 nm particles ($0.08$ cm$^{-3}$ s$^{-1}$ at Värrö and $0.09$ cm$^{-3}$ s$^{-1}$ at Pallas) and the number of new particles produced during the event ($1750$ cm$^{-3}$ at Värrö and $1700$ cm$^{-3}$ at Pallas) were very similar. The average growth rates were equal (2.9 nm h$^{-1}$) at both sites. Due to the low levels of SO$_2$, the observed particle growth rate could not be explained by sulphuric acid from SO$_2$ oxidation. Other vapours are clearly needed. Overall the simultaneous events indicated that in clean air masses the geographical extent of a nucleation event could be at least 250 kilometres which was the distance between Pallas and Värrö stations.

On the days new particle formation was observed only at the Värrö site, high SO$_2$ concentrations (up to $70$ µg m$^{-3}$) were present on 75% of the days, while Pallas had normally very low (< 2 µg m$^{-3}$) concentration. On most of these days the air masses at Värrö had been travelling through the Kola Peninsula industrial area. On some of these days also Pallas was affected by air masses coming from the Kola area, but it seems that the distance from Kola to Pallas decreases the gas concentration lower than what is needed to launch a nucleation process at Pallas. Also on few days Pallas was affected by a fog or low-cloud cover. These events observed only at Värrö are an indication of new sulphate particles formed in plumes originating from the Kola industry (e.g. Figure 10). Previous studies indicate that nucleation involving H$_2$SO$_4$ could be a viable mechanism in plumes containing high concentrations of SO$_2$. H$_2$SO$_4$ is also expected to participate in particle growth by condensation on existing particles. (Mueller and Imhoff 1994, Kerminen and Wexler 1996, Pirjola et al. 1998).

The days new particle formation was observed only at the Pallas site could mostly be explained by the fact that the stations were exposed to air masses of different origin. On few cases, the air masses came from north, but travelled over the Kola Peninsula industrial area prior to arriving at Värrö. This polluted area may also act as a sink for condensable gases formed above the sea and thereby suppress new particle formation as well as cloud/rain.
A summary of seasonal variation of some parameters related to class 1 and 2 particle formation events observed during years 2002–2003 is presented here. The starting time of particle formation (7 nm particles) was earliest in summer, following the annual pattern of sun rise. The duration of these events seemed to have no seasonal pattern. The formation rate of 7 nm particles (J7) had no clear seasonal variation, though slightly higher average values were observed in spring and summer in Värriö. The growth rate had a similar seasonal pattern at both sites. The highest growth rates were observed during summer. The production of new particles per event was highest in spring and summer at both sites. Condensation sink (and the pre-existing particle concentration) had slightly higher values in spring and summer. This together with higher growth rates indicates higher vapour source rate needed in spring and summer.

3.2.2 The effects of different air masses

An additional study with new particle formation data from years 2002–2003 (Paper III) provided results close to similar as shown earlier (Paper II). This study concentrated only on the formation events that were occurring in the same air masses at both sites. With a large set of trajectories we picked up all the trajectories that had passed the Pallas site within few tens of kilometres prior to entering Värriö, and vice versa from the air mass transport from Värriö to Pallas. With this set of trajectories we can compare the particle formation events occurring in air masses with similar trajectory paths at both sites. Average values of the event parameters were close to each other at the sites. With the obtained set of trajectories, also differences between eastern (continental) and western (marine) air masses were observed.

First, we take a look at the western air masses originating mostly from the Atlantic Ocean. The particle growth rate and J7 were equal in both sites during these western air masses. However, Värriö had a higher pre-existing particle concentration and higher condensation sink (CS) compared to Pallas. Within these western air masses the emissions are mainly biogenic, therefore Värriö (deeper in the boreal forest than Pallas) has higher source rates of condensable vapour. However, higher vapour source rates combined with higher condensation sinks compensate each other, which might partly explain why, on average, no difference in 7 nm particle formation rate and growth rate was observed between Pallas and Värriö in these western air masses.

The eastern air masses were mostly continental and travelled sometimes over the Kola Peninsula industrial area. These air masses arrive first to Värriö before arriving at Pallas. Värriö is closer to
the Kola Peninsula area being influenced more strongly by the industrial plumes. As mentioned earlier, the oxidation of SO$_2$ to H$_2$SO$_4$ in these plumes could boost the particle formation in Värriö. This is seen as higher J7 and growth rate than in Pallas during these eastern air masses. On average, a double amount of new particles were produced in Värriö compared to Pallas. This is partly due to a higher CS and slightly higher pre-existing particle concentration observed in Pallas during these eastern air masses. Higher values in Pallas refer either to some primary particle sources or to secondary formation of particulate matter between the sites.

In eastern air masses almost all events were observed to start earlier at the eastern station Värriö, whereas in western air masses most of the events were observed to start earlier at the western station Pallas. This demonstrates that particle formation in a certain air mass type depends not only on the diurnal variation of the parameters causing the phenomenon (such as photochemistry) but also on some properties carried by the air mass itself. The formation rate of 7 nm particles showed clear differences between the two sites, but no systematic pattern can be found. It seems that the particle formation rate is not very conservative parameter in an air mass. On the other hand the correlation in growth rates between the two sites was relatively good, which suggests that the amount of condensable vapour causing the growth must have been at about the same level in both sites.

When looking at the average values on both sites, the eastern (continental) air masses had higher pre-existing particle concentration and CS, compared with western (marine) air masses. The eastern air masses had also higher J7 and growth rate. Also a larger number of new particles were produced during eastern air masses, compared with western air masses. This requires higher concentration and source rate of condensable vapour and suggests that on average in continental (eastern) air masses, there is more nucleating and condensable vapour available for particle formation and subsequent growth.

### 3.2.3 New particle formation in air mass transported between two measuring sites

One of the ideas of paper III was to find out whether the aerosol particle physical properties observed at the Värriö station can be generated from those observed at the Pallas station, or if something is taking place which we can not explain. In our previous study we found lots of similarities, but also some major differences between the sites (paper II). Observed differences were mostly explained by different air masses at the sites. Paper III analyses 24 months of data
and presents two different case studies in more detail, in which the same air mass trajectories cross both the sites. An effort is made to model new particle formation observed in Värriö site based on size distributions measured in Pallas site. The selected events were simulated with the UHMA aerosol dynamics model. These different cases were selected to test phenomena, if the cases could be explained with similar modelling setups.

Figure 11. The used concentration profiles of condensable organics and sulphuric acid during the simulation.

The concentration profiles of condensable organics and sulphuric acid used during the simulation are presented in Figure 11. In the selected cases, the simulated concentrations of sulphuric acid explained less than 10% of the nucleation mode growth indicating that organic compounds are the main contributor to particle growth in this clean yet relatively sparsely vegetated subarctic environment (see paper III). This observation is also in line with earlier analysis for more densely vegetated environments (Birmili et al., 2003; Boy et al., 2005). The crucial role of organics in new particle growth offers a tentative hypothesis to the distinct features of new particles at the two sites: At the source station Pallas no particle formation at 7 nm is detected in one of the cases and in the other case the growth of newly formed particles is not as clear as at Värriö. Since the
air mass in both cases reaches Pallas after travelling over land for only 9–10 hours, the time for the organic vapours to accumulate in the air and to interact with the particles is relatively short. As a result, the growth rate of newly formed clusters remains low, they are effectively scavenged by pre-existing particles and may not reach the detection limit of the measurement instrumentation.

4 Aerosol-cloud interactions

An attempt was done to link new particle formation to formation of cloud condensation nuclei (CCN). This was done by investigating the growth of newly formed particles to CCN sizes and their contribution to CCN population, and ultimately their connection with cloud droplet formation. As explained earlier, the two measurement sites (Sammaltunturi and Matorova) provide an opportunity to investigate the aerosol-cloud interactions, since Sammaltunturi is inside the cloud during 10 % of all days (5 % of the time), while the lower-altitude Matorova station is practically always outside the cloud (e.g. Figure 6). Size-segregated activation efficiency of sub-micron size aerosol particles could be measured with similar DMPS systems at both the stations. Finally, direct evidence of link between new particle formation and cloud formation is presented.

4.1 Formation of potential CCN via new particle formation

A total of 103 new particle formation events were observed during the 33-month period considered in this CCN study. From all the particle formation events observed, 19 were selected for a closer inspection, which is 18 % of all events. The criteria for this selection are presented in section 2.3.4.

For the selected CCN formation events, CCN$_{50}$ and CCN$_{80}$ increased on average by factors of 11.7 and 4.1, respectively, during the events. This result indicates very clearly that atmospheric new particle formation is an extremely important source of new “potential” CCN at our site during the periods of clean air masses influenced by new particle formation. Laaksonen et al. (2005) have demonstrated that new particle formation can also be an important source of CCN even in a polluted environment. In absolute terms, the average CCN$_{50}$ increase of 794 cm$^{-3}$ caused by these CCN formation events in Pallas is more than double the overall average CCN$_{50}$
Contrary to this, the average CCN\(_{80}\) increase of 197 cm\(^{-3}\) is close to average CCN\(_{80}\) (235 cm\(^{-3}\) in year 2001).

An interesting feature is the huge variability in CCN\(_{50}\) and CCN\(_{80}\) produced by each event, both in absolute terms and relative to the number of CCN originally present in that air mass. Increases in CCN\(_{50}\) and CCN\(_{80}\) were correlated to each other (R\(^2\)=0.71) but less so with the number of new particles produced (R\(^2\)=0.27 and R\(^2\)=0.30, respectively).

One explanation for this very different CCN formation behaviour between different events tested with two example cases is the initially very different condensation sinks. When the condensation sink is low, concentrations of low-volatile vapours can build up more easily so that the particle growth is accelerated. On the other hand, no major differences were seen in the number of new particles formed between the two example events. These two features together suggest that vapours responsible for the formation and early growth of new particles may be different from vapours responsible for their growth to CCN sizes, and that the sources of these two vapour types are likely to have been somewhat different.

While sulphuric acid has been demonstrated to be a key compound in atmospheric new particle formation, the subsequent growth of these particles to larger sizes is in most cases driven by some other yet largely unidentified vapours (e.g. Weber et al., 1997; Kavouras et al., 1998; Kulmala et al., 2001b; O’Dowd et al., 2002). To evaluate the potential role of sulphuric acid in the CCN formation events considered here, an example case was studied. Using simple aerosol dynamical arguments (e.g. Kulmala et al., 2001a), our estimations show that in this case the sulphuric acid concentration corresponds to a particle growth rate of about 1.4 nm h\(^{-1}\), which is well below the average observed growth rate of 3.6 nm h\(^{-1}\). In reality, the contribution of sulphuric acid to the formation of “potential” CCN is likely to have been considerably smaller, since the concentration of SO\(_2\) and probably also of OH were somewhat lower than what was used in these estimations. In Hyytiälä, Southern Finland, Boy et al. (2005) have estimated sulphuric acid to be responsible for about 10 % of the nucleation mode particle growth on average. This supports our finding.

Several important findings were made and can be summarised as follows: 1) all CCN formation events took place in originally quite clean air coming from the Northern Atlantic/Arctic Ocean, 2) gaseous sulphuric acid resulting from SO\(_2\) oxidation gave only a minor contribution to the production of “potential” CCN, 3) the number of new “potential” CCN formed varied significantly between different events and correlated weakly with the number of new particles formed during the same event, and 4) the concentrations of the smallest “potential” CCN were
affected substantially more by new particle formation than the concentrations of larger "potential" CCN.

The apparent reason for the finding (1) is that no new particle formation was observed to take place in continental air masses, or in air affected significantly by anthropogenic sources. It remains to be studied whether it is pollution that suppresses new particle formation at our site, or whether some meteorological factors associated with the northerly/oceanic air masses make them more favourable to new particle formation and subsequent CCN production. The finding (2) is consistent with observations by others (e.g. Kavouras et al., 1998; Kulmala et al., 2001b; O’Dowd et al., 2002), which have shown that the growth of newly formed particles to larger sizes is driven usually by vapours other than sulphuric acid. Finding (3) is suggestive of some sort of decoupling between new particle formation and growth, being in line with our current understanding of the processes affecting atmospheric particle formation (e.g. Kulmala et al., 2000; Kerminen et al., 2001). By combining the findings (1), (2) and (3) together, it might be tempting to conclude that new particle formation took place via a nucleation mechanism involving gaseous sulphuric acid and that the subsequent production of "potential" CCN was tied to some organic (or other) vapours of mostly natural in their origin. This conclusion cannot, however, be drawn before identifying the vapours participating into these processes.

The finding (4) is not a surprise, since primary and secondary particle sources contribute to very different parts of the particle size spectrum. Both observations and model studies have shown that the sizes of the smallest CCN are considerably lower under clean conditions than under polluted conditions (e.g. Covert et al., 1998; Cohard et al. 2000). This means that, if applicable to other continental sites as well, findings (1) and (4) act to support each other when looking at the climatic effects caused by atmospheric CCN formation.

4.2 Activation efficiency of aerosol particles

After a careful classification and selection, described in section 2.3.5, a total of 50 “cloud droplet activation days” were selected for this study of which 39 are inspected in more detail. The number of these days peaked in late autumn (October–November) and was lowest in summer (May–August). In November, cloud droplet activation was observed during one third of the days. In summer only 0–2 events were observed per month. The annual variation of “cloud droplet activation” events followed the annual pattern of low clouds (those below 1000 m). On average
the cloud base was lowest during late autumn (October–November) and highest in summer (June). Orographic clouds were not observed, but orography may have some effect on the cloud layer. Cloud events were observed to start throughout the day. The duration of cloud events varied from one hour to 47 hours being 11 hours on average.

The diameter corresponding to 50 % activation efficiency, $D_{50}$, is a useful quantity especially for modelling purposes, since it provides a simple way to divide the particle population into two classes: those that activate into cloud droplets (particles with a dry diameter larger than $D_{50}$) and those that remain as cloud interstitial particles. Two examples of these activation curves are seen in Figure 7. In our measurements, $D_{50}$ varied between 50 and 128 nm with an average of 80 nm. The value of $D_{50}$ increased typically with increasing pre-existing particle concentration. The cleaner marine air masses from Northern Atlantic or the Arctic Ocean had an average $D_{50}$ of 76 nm, while the more polluted continental air masses from south and east had an average $D_{50}$ of 91 nm (e.g. Figure 7). Although all rainy days were taken out of the final data set, $D_{50}$ was also calculated for 23 rainy cloud days for a general interest. On rainy days particles will experience also below cloud scavenging additional to in-cloud scavenging/activation. For these rainy days $D_{50}$ was on average 92 nm and varied between 49 and 137 nm.

The pre-existing particle concentration varied between 60 and 1330 cm$^{-3}$ with an average of 410 cm$^{-3}$ during the cloud events. The average number of activated particles was 154 cm$^{-3}$ and varied between 30 and 610 cm$^{-3}$. The resulting percent of activated particles varied from 9 to 86 % with an average of 47 %. A clear relation between the total pre-existing particle number concentration ($N_{tot}$) and the number of activated particles ($A_{tot}$) was found. Also the percent of activated particles and $D_{50}$ were related to total number concentration. On average, a higher pre-existing particle number concentration was associated with a higher number of activated particles, lower activation percent and larger $D_{50}$ (see Table 2).

The cleaner air masses from the Northern Atlantic or Arctic Ocean had, on average, 120 cm$^{-3}$ activated particles with an activation percent of 42 %. In the more polluted air masses from south and east, the average $A_{tot}$ was 200 cm$^{-3}$ and the activation percent was 53 %. The slightly lower activation percent in clean marine air compared with continental air is somewhat surprising and not consistent with the general trend found. The most likely reason for this behaviour can be found in the different shape of the particle number size distribution between these two air mass types: marine air masses had typically a more pronounced Aitken mode and thereby a larger fraction of particles that are too small to act as cloud condensation nuclei (see Figure 7). Another
factor that affects the activation percent is the chemical composition of particles, which might be different between the marine and continental air masses.

Table 2. Concentration out-of-cloud (pre-existing particle concentration, N) compared with cloud droplet activation (number and fraction) and the 50 % activation efficiency diameter.

<table>
<thead>
<tr>
<th>N (cm$^{-3}$)</th>
<th>A (cm$^{-3}$)</th>
<th>A (%)</th>
<th>D$_{50}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt; 150</td>
<td>66</td>
<td>60</td>
<td>71</td>
</tr>
<tr>
<td>150 – 300</td>
<td>97</td>
<td>49</td>
<td>78</td>
</tr>
<tr>
<td>300 – 600</td>
<td>181</td>
<td>41</td>
<td>85</td>
</tr>
<tr>
<td>&gt; 600</td>
<td>290</td>
<td>34</td>
<td>87</td>
</tr>
</tbody>
</table>

The modal cloud droplet activation of aerosol particles was calculated for three fixed modes: the nucleation mode (7–25 nm), Aitken mode (25–95 nm) and accumulation mode (95–500 nm). In the accumulation mode, 87 % of the particles were activated on average, varying from 71 to 97 %. The average number of activated accumulation mode particles was 110 cm$^{-3}$ varying between 20 and 310 cm$^{-3}$. If the nucleation mode was left out of N$_{tot}$ and the correlation for A$_{acc}$ was done with N$_{ait+acc}$ (the number concentration of particles larger than 25 nm in diameter), the correlation coefficient ($R^2$) was 0.42. A very high correlation ($R^2$=0.97) was found between N$_{acc}$ and A$_{acc}$, as one might expect.

Even in Aitken mode, notable activation was found to take place, since on average 30 % of the Aitken mode particles were activated. The average number of activated Aitken mode particles was 35 cm$^{-3}$ varying from zero to 200 cm$^{-3}$. Aitken mode particles were observed to cover up to 55 % of the total number of activated particles (i.e. number of formed droplets), 22 % on average. All this demonstrates that Aitken mode particles have a significant influence on cloud droplet activation, being important when estimating the indirect climatic effects by aerosols. This observation agrees nicely with the earlier model predictions by Kulmala et al. (1996).

A compilation of cloud droplet measurements done in marine and continental clouds has been presented by Miles et al. (2000). They found significant differences between these two cloud types, with average cloud droplet number concentrations being equal to 74 and 288 cm$^{-3}$ for marine and continental clouds, respectively. These results are in qualitative agreement with our
values of 120 cm$^{-3}$ and 200 cm$^{-3}$ for marine and continental air masses, respectively. The slightly higher droplet concentration in marine air masses measured by us may be due to the fact that these air masses have travelled over the continent prior to arrival at Pallas. The relatively clean continental air at Pallas explains the lower number of activated particles in continental air masses compared to the results in Miles et al. (2000). The range of the number of activated particles in Pallas, 30–610 cm$^{-3}$, was in the range of the average values reported by Miles et al. (2000). Droplet concentrations up to 2500 cm$^{-3}$, observed in polluted conditions (Bower et al., 2000) were not reached at Pallas site. On the contrary, the extremely low average cloud droplet concentration of 20–70 cm$^{-3}$ measured in the Southern Ocean by Yum and Hudson (2004) is in the range of the lowest values (30 cm$^{-3}$) found at Pallas. The average value at Pallas, 154 cm$^{-3}$, could be classified as clean continental or slightly polluted marine clouds.

Literature data on the activated fraction of aerosol particles is available almost solely for the accumulation mode. In these studies, the activated fraction of accumulation mode particles has been found to vary from <0.2 to values close to unity, with higher fractions typically encountered in cleaner air and for higher cloud liquid water contents (Anderson et al., 1994; Hallberg et al., 1994, Gillani et al., 1995; Schwarzenboeck et al., 2000; Henning et al., 2002; Glantz et al., 2003). The activated fractions measured by us for the accumulation mode are at the upper end of those found in the earlier studies, averaging 93 % for the cleanest air mass category and 83 % for the most polluted air mass category.

Glantz et al. (2003) found a poor correlation between the number of cloud droplets and the mass (volume measured) of cloud residual particles for clean air, based on which they suggested that a major fraction of particles determining the cloud droplet number concentration were below the accumulation size range (<100 nm). In our measurements, the contribution of Aitken mode particles to the cloud droplet population was found to be significant, being 22 % on average. The activated fraction of combined Aitken and accumulation mode particles ranged from 12 to 93 % with an average of 58 %. These values are larger than those measured by Schwarzenboeck et al. (2000) but comparable to those measured by Lowenthal et al. (2002) at a high-altitude site (3210 m a.s.l.) in north western Colorado. Direct evidence of the activation of Aitken mode particles at clean conditions has also been reported by Henning et al. (2002) at a high-Alpine site Jungfraujoch in Central Europe.
4.3 Direct evidence of link between particle formation and clouds

One source for ultrafine particles is atmospheric aerosol formation which includes the nucleation of nanometre-size clusters from gaseous precursors and subsequent growth of these clusters to larger sizes. Observationally, it is extremely difficult to follow the time evolution of nucleated clusters up to the point at which they eventually activate into cloud droplets. As a result, although aerosol formation appears to take place throughout the Earth’s troposphere (Kulmala et al., 2004a), no direct evidence has been obtained so far that these same particles would contribute to cloud droplet populations. This section will provide such evidence based on measurements made in a remote continental site. By combining the measurement data with theoretical calculations, we further suggest that atmospheric aerosol formation is able to enhance cloud albedo and thereby affect the radiative forcing by clouds.

Since the beginning of our measurements in the Pallas in April 2000, more than a hundred aerosol formation events have been observed, and several tens of individual cloud encounters have been identified. An example of observed aerosol formation and growth event followed by a cloud encounter is shown in Figure 12. During this 60-hour period, very clean Arctic air was being transported to the measurement site. The air temperature varied between −10 and 0 °C and the relative humidity was >80 %. No rain was encountered, even though some intermittent drizzle could be observed during the cloudy period on the day of year (DY) 121–121.5. The measured aerosol number size distributions were characterized by a stable yet weak accumulation mode (average particle number concentration <150 cm$^{-3}$), together with an ultrafine particle size range (particle diameter <100 nm) consisting of a very weak “background” Aitken mode embedded in a more pronounced and variable group of particles (number concentration of up to 2000 cm$^{-3}$) formed in the atmosphere by nucleation.

Atmospheric aerosol formation can be seen as a series of three events in Figure 12, the first of which took place on DY 119, and the next two ones on the following day. During all the events, new particles appeared first at the lower end of the particle size spectrum, demonstrating unambiguously that these particles originated from nucleation in the atmosphere. The evolution of the particle population associated with each event could be followed by more than 12 hours in time, indicating that atmospheric aerosol formation was taking place in a large spatial scale (some hundreds of kilometres). Particles formed during the last two events grew relatively fast in size, producing a significant number of particles larger than 50 nm in diameter by the morning hours on the DY 121 (Figure 12). Although some temporal fluctuations in the size distribution of
ultrafine particles were detected, it is quite clear that atmospheric aerosol formation was responsible for the great majority of these particles during the whole measurement period.

A cloud entered the site soon after the midnight on the DY 121 and persisted for about 10 hours. The presence of cloud can be seen from Figure 12 as a sudden disappearance of the accumulation and part of the Aitken mode. In the absence of rain, such a disappearance can be interpreted as the activation of these particles into cloud droplets (Paper V). In this case, during the cloudy period about 80% of accumulation mode particles and >60% of particles in the size range 50–100 nm had been activated into cloud droplets. The total number concentration of cloud droplets was estimated to be slightly over 300 cm$^{-3}$, of which about two thirds originated from ultrafine particles. We may thus conclude that particles formed in the atmosphere during the DY 120 contributed significantly to the overall cloud droplet population in this case.

Figure 12. An aggregate of particle number size distributions measured continuously in Pallas (upper-altitude Sammaltunturi site) starting from noon on April 29, 2003. Red crosses display the instances when particles formed originally in the atmosphere reached our measurement size range. During the period between the red bars, the measurement site was inside a cloud and the measured particle number size distribution is representative of that of cloud interstitial particles.
In addition to continental forest sites, large-scale aerosol formation events capable of producing cloud condensation nuclei have been observed in many other clean or moderately polluted environments, including coastal areas (O’Dowd et al., 2002), upper free troposphere (Singh et al., 2002), cloud outflow regions (Twohy et al., 2002), and aged continental plumes (Naughton et al. 2004). An estimate on indirect forcing attributable to atmospheric new particle formation at Pallas region is presented in detail in paper VI. As a result, although direct aerosol forcing is likely to dominate over the indirect one in heavily-polluted regions of the Earth (Kaufman et al., 2002), the forcing resulting from atmospheric aerosol formation might account for a significant fraction of both indirect and total aerosol forcing in the global atmosphere.

5 Review of the papers

The papers included in this thesis are mostly based on aerosol number size distribution (7–500 nm) measurements and analysis of them. Measurements are mainly carried out at two stations, Sammallunturi and Matorova, at Pallas area. Pallas is in a subarctic area in Northern Finland close to the northern limit of boreal forest zone. In papers II and III size distribution measurements at Värriö station, 250 km apart from Pallas, are also used. The papers take a look at particle concentration and new particle formation in Northern Finland. Estimations on geographical scale of new particle formation are made. New particle formation is then connected to formation of cloud condensation nuclei (CCN), and the aerosol-cloud interactions at Pallas are studied. Finally, observational evidence of link between new particle formation and cloud droplet activation is presented.

Paper I presents the first results of particle number size distribution measurements conducted at Pallas. Particle number concentrations and new particle formation events are studied at two stations six kilometres apart from each other. The stations are at different altitudes and surroundings, Sammallunturi is above the tree line on the top of a fjeld and Matorova is surrounded by forest. Differences caused by the different locations are searched, but the variability between the sites is small. Annual and diurnal variation of particle concentration is presented, also in modal respect. Distinct new particle formation events are compared between the sites and with some previous measurements.

Paper II presents a set of new particle formation events observed at Pallas and Värriö sites 250 km apart from each other. A comparison between the sites is made and, based on this, an
estimation of the geographical scale of new particle formation is presented. Trajectories, trace gas concentrations and weather related parameters were investigated in order to explain the observed differences between the two sites. Possible effects of the Kola Peninsula industrial area on new particle formation are presented.

**Paper III** investigates two years of new particle formation events observed at Pallas and Värriö. The comparison is made only for the events that occur in the same air mass on both stations (i.e. the same trajectory path crosses both sites). A division between marine and continental air masses is made and their effects on new particle formation are studied. Seasonal variation of new particle formation parameters is presented. An effort is made to model new particle formation in Värriö based on size distributions measured in Pallas site. These modelling results are presented as two case studies.

**Paper IV** relates new particle formation to production of potential CCN. New particle formation followed by a particle growth of sufficiently long time allows investigating CCN resulting from new particle formation. In total 19 cases are investigated and changes in the CCN number concentration caused by new particle formation are presented. The contribution of gaseous sulphuric acid resulting from SO$_2$ oxidation on the particle growth on Pallas site is also estimated.

**Paper V** deals with aerosol-cloud interactions. A slightly different approach compared to traditional methods is presented by measuring simultaneously the cloud interstitial particle size spectrum and a nearby out-of-cloud particle spectrum. This approach allows us to determine the activated fraction over the whole sub-micron size range (basically down to 7 nm). The number concentration of activated particles, the activated fraction and the diameter corresponding to 50 % activation efficiency are presented for 39 cloud events. The presented values are categorized in terms of the level of pollution. Clear evidence of the important role of ultrafine particles (<100 nm) in cloud formation was found.

**Paper VI** provides direct observational evidence that aerosol particles formed in the atmosphere from gaseous precursors eventually participate into cloud droplet activation, based on continuous aerosol measurements in a remote continental location. By combining the measurement data with theoretical calculations, this paper further demonstrates that the albedo of clouds may be significantly influenced by atmospheric aerosol formation, and that this process needs to be taken into account when estimating the indirect climatic effects of aerosols in the global atmosphere.
6 Conclusions

In total, close to four years of continuous aerosol particle size distribution measurements was analyzed in this thesis. The measurements have been carried out at Pallas and Värrö in Northern Finland. Measurements at Pallas have been made at two different heights (340 m and 560 m above sea level) and surroundings for two years to find out the effects of local scale conditions on new particle formation and growth, as well as on the size distribution of aerosol particles.

Most of the new particle formation events occurred on April and May. Elevation or surrounding of the measuring site did not have noticeable effect on new particle formation and growth. Formation events were always observed at both Pallas stations with maximum a time difference of 30 minutes between starting times, which could not be explained directly with wind direction and speed. Solar radiation was observed to be one of the key factors needed for particle formation. According to trajectory analysis, all the events were associated with marine/polar air masses originating from the Northern Atlantic or the Arctic Ocean. The surrounding forest at Matorova did not have significant effect on the variation of physical properties of aerosol particles. In general, the results between the Pallas stations were surprisingly similar taking into account the different elevations and surroundings. The results indicate that the detected particle formation events are caused by larger than local scale conditions.

A clear seasonal variation was found in the starting time of the events, following the annual pattern of sun rise. No clear seasonal pattern in the formation rate of 7 nm particles was observed. The particle growth rate showed a clear seasonal variation, the values being highest in summer. These results are in agreement with earlier studies at other sites.

Differences in western (marine) and eastern (continental) air masses were studied. In eastern air masses almost all events were observed to start earlier at the eastern station Värrö, whereas in western air masses most of the events were observed to start earlier at the western station Pallas. This demonstrates that particle formation in a certain air mass type depends not only on the diurnal variation of the parameters causing the phenomenon (such as photochemistry) but also on some properties carried by the air mass itself. In marine air masses, the formation and growth rates were close to equal in both sites on average. During western air masses, emissions are primarily biogenic, therefore Värrö (deeper in the boreal forest than Pallas) has higher source rates of condensable vapour. However, higher vapour source rates combined with higher condensation sinks compensate each other. This might explain why no difference in particle
formation rates and growth rates were observed between Pallas and Värriö in these western air masses. On average, continental air masses had higher formation and growth rates compared to marine air masses in both sites. This suggests that in these more polluted continental air masses there is more nucleating and condensable vapour available for particle formation and growth.

The geographical scale of new particle formation was studied at the two northern sites 250 kilometres apart from each other. On 45% of the days, particle formation was observed only at one of the two stations. Most of these cases could be explained by air masses of different origin, or by the presence of rain/fog at the other station. It also appears likely that some of the events observed only at Värriö were caused by nucleation involving H$_2$SO$_4$ from SO$_2$ oxidation in plumes originating from the Kola industry. On the particle formation events observed at both sites on the same day, the average particle formation and growth rates were very similar at both sites. Overall these simultaneous events indicate that in clean and pollutant free air masses the geographical extent of a nucleation event is at least 250 kilometres which is the distance between Pallas and Värriö stations.

New particle formation resulting to CCN formation was studied at Pallas. Several important findings were made and can be summarised as follows: 1) all CCN formation events took place in originally quite clean air coming from the Northern Atlantic/Arctic Ocean, 2) gaseous sulphuric acid resulting from SO$_2$ oxidation gave only a minor contribution to the production of “potential” CCN, clearly other vapours are needed, 3) the number of new “potential” CCN formed varied significantly between different events and correlated weakly with the number of new particles formed during the same event, and 4) the concentrations of the smallest “potential” CCN were affected substantially more by new particle formation than the concentrations of larger “potential” CCN.

Cloud droplet activation of aerosol particles was studied at Pallas, considered as a clean background site. Statistically, a relation between the number of pre-existing particles ($N_{tot}$), the number of activated particles ($A_{tot}$) and the diameter of 50% activation efficiency ($D_{50}$) was found. Larger values of $N_{tot}$ were associated with larger $A_{tot}$, larger $D_{50}$ and lower percent of activated particles. The values of $D_{50}$ were on average 80 nm and varied from 50 to 128 nm. The average activation percent during the cloud events was 47% and varied from 9 to 86%. The air mass origin had an effect on droplet activation due to its effect on the aerosol concentration. The cleaner air masses from Northern Atlantic or the Arctic Ocean had on average 15 nm lower $D_{50}$ than the more polluted air masses from the south and the east. In the accumulation mode, 87% of
the particles were activated on average. Even in the Aitken mode the activation was found notable, since on average 30 % of the Aitken mode particles were activated. Aitken mode particles were observed to cover up to 55 % of the total number of activated particles (i.e. number of formed droplets), 22 % on average. Finally, our example in section 4.3 provided direct observational link between aerosol formation and the participation of these same aerosol particles into cloud droplet activation. New particle formation events followed by a cloud encounter were observed and part of the newly formed particles was observed to act as cloud condensation nuclei.

This study indicates that atmospheric new particle formation has to be taken seriously when estimating the climatic effect of aerosol particles. New particle formation can occur simultaneously over large geographical scales. Evidence on large numbers of newly-formed particles growing to CCN sizes was obtained. Particles down to about 50 nm in diameter are observed to activate into cloud droplets and therefore the role of ultrafine particles (<100 nm) in this activation seems to be very important, at least in these clean or moderately polluted environments. In addition to continental forest sites, large-scale aerosol formation events capable of producing cloud condensation nuclei have been observed in many other clean or moderately polluted environments (Kulmala et al., 2004a) and even in a highly polluted region (Laaksonen et al., 2005). As a result, although direct aerosol forcing is likely to dominate over the indirect one in heavily-polluted regions of the Earth (Kaufman et al., 2002), the forcing resulting from atmospheric aerosol formation might account for a significant fraction of both indirect and total aerosol forcing in the global atmosphere.

Future research on this topic should include better observational characterization of the frequency and magnitude of aerosol formation events leading to cloud condensation nuclei production in different tropospheric environments and, with help of improved theoretical frameworks and parameterizations, a detailed treatment of this phenomenon in climate simulations.

As this study shows, continuous measurements are very effective and therefore, more continuous measurements are needed along with intensive field campaigns in different environments to get more information on new particle formation and its geographical extent as well as the chemistry and physics included in cloud droplet activation. This is needed, especially in the ultrafine particle size range that is relevant to clouds forming in clean or moderately polluted environments. More and reliable measurements in different environments are required to provide a larger amount and more reliable data for climate models.
References


