REPORT SERIES IN AEROSOL SCIENCE
N:o 100 (2008)

Aerosol Particle Formation: Meteorological and Synoptic Processes behind the Event

Larisa Sogacheva

Division of Atmospheric Sciences and Geophysics
Department of Physics
Faculty of Science
University of Helsinki
Helsinki, Finland

Academic dissertation

To be presented with the permission of the Faculty of Science of the University of Helsinki, for public criticism in auditorium E204, Gustaf Hällströmin katu 2, on December 15th, 2008, at 12 o'clock

Helsinki 2008

Author's address: Department of Physics
P.O.Box 64
FI-00014 University of Helsinki
larisa.sogacheva@helsinki.fi

Supervisor: Professor Markku Kulmala, Ph.D.
Department of Physics
University of Helsinki

Reviewers: Docent Hannele Korhonen, Ph.D.
Department of Physics
University of Kuopio

Docent Tatu Anttila, Ph.D.
Climate Change Unit, Atmospheric Modelling and Observations
Finnish Meteorological Institute

Opponent: Senior Scientist Wolfram Birmili, Ph.D.
Research group leader at the Leibniz Institute for Tropospheric Research, Leipzig, Germany

ISBN 978-952-5027-98-3
ISSN 0784-3496
Helsinki 2008
Yliopistopaino

ISBN 978-952-5027-99-0 (E-thesis version)
http://ethesis.helsinki.fi/
Helsinki 2008
Helsingin yliopiston verkkojulkaisut

Larisa Sogacheva
University of Helsinki, 2008

Abstract

Aerosol particles in the atmosphere are known to significantly influence ecosystems, to change air quality and to exert negative health effects. Atmospheric aerosols influence climate through cooling of the atmosphere and the underlying surface by scattering of sunlight, through warming of the atmosphere by absorbing sun light and thermal radiation emitted by the Earth surface and through their acting as cloud condensation nuclei. Aerosols are emitted from both natural and anthropogenic sources. Depending on their size, they can be transported over significant distances, while undergoing considerable changes in their composition and physical properties. Their lifetime in the atmosphere varies from a few hours to a week.

New particle formation is a result of gas-to-particle conversion. Once formed, atmospheric aerosol particles may grow due to condensation or coagulation, or be removed by deposition processes. In this thesis we describe analyses of air masses, meteorological parameters and synoptic situations to reveal conditions favourable for new particle formation in the atmosphere. We studied the concentration of ultrafine particles in different types of air masses, and the role of atmospheric fronts and cloudiness in the formation of atmospheric aerosol particles.

The dominant role of Arctic and Polar air masses causing new particle formation was clearly observed at Hyytiälä, Southern Finland, during all seasons, as well as at other measurement stations in Scandinavia. In all seasons and on multi-year average, Arctic and North Atlantic areas were the sources of nucleation mode particles. In contrast, concentrations of accumulation mode particles and condensation sink values in Hyytiälä were highest in continental air masses, arriving at Hyytiälä from Eastern Europe and Central Russia.

The most favourable situation for new particle formation during all seasons was cold air advection after cold-front passages. Such a period could last a few days until the next front reached Hyytiälä. The frequency of aerosol particle formation relates to the frequency of low-cloud-amount days in Hyytiälä. Cloudiness of less than 5 octas is one of the factors favouring new particle formation. Cloudiness above 4 octas appears to be an important factor that prevents particle growth, due to the decrease of solar radiation, which is one of the important meteorological parameters in atmospheric particle formation and growth.

Keywords: Atmospheric aerosols, particle formation, air mass, atmospheric front, cloudiness

Acknowledgements

The research for this thesis has been carried out at the Division of Atmospheric Sciences and Geophysics, University of Helsinki. I wish to express my gratitude to my supervisor, Prof. Markku Kulmala, for introducing me to aerosol science, for providing ideas, views and constant support for my work. His experience and deep scientific knowledge has been of great help in my professional growth. The topic of my research in-between meteorology and aerosol science, suggested by Prof. Kulmala, provided me a nice opportunity for feeling the synergy between these fields. I sincerely thank Markku for creating such a nice atmosphere in the department, where people working together are open to discussion and communication.

My thanks are extended to all my former and current colleagues at the Division of Atmospheric Sciences and Geophysics for an easy-going and supportive atmosphere, for their patience while introducing me to the basics of aerosol science during the first years of my studies, for their interest in my research and willingness to discuss questions appearing along my work. In particular I want to mention Dr. Ilona Riipinen, Dr. Miikka Dal Maso, Dr. Üllar Rannik, Mr. Heikki Junninen, Dr. Anca Hienola, Mrs. Tiia Grönholm, Dr. Michael Boy and Mrs. Johanna Lauros.

I would like to acknowledge Prof. D.M.Schultz for the nice and emotional discussions we had, for his patience in reading and correcting my views in a way it would be understandable for readers.

Especially, I express my gratitude to Prof. Gerrit de Leeuw. He has been my second supervisor during the last years, and I hope that our collaboration will be even more fruitful during the coming years. I am grateful to Mrs. Anu-Maija Sundström and Dr. Pekka Kolmonen, from whom I got an excellent experience of group working, sharing progress, and troubles, together.

I wish to thank all the co-authors of the Papers included in this thesis for their fruitful cooperation and discussion. I thank Dr. Hannele Korhonen from the University of Kuopio, and Dr. Tatu Anttila from the Finnish Meteorological Institute, for reviewing this thesis and for their comments and suggestions towards improving the manuscript.

I would like to thank Risto Makkonen and Jonni Larjomaa for assisting me with computer problems. Ulla Antila was always helping me with different kinds of practical matters during my work.

I would like to take this opportunity to express my gratitude to my family, my parents and my sister Tania, who have been supporting me in all my undertakings and passions. My gratitude goes also to my school friends and school teachers, who continue to be my irreplaceable support in my social life. My geography teacher Andrey V. Monahov taught me not only how to keep smiling during an endless rain, how to choose the right course while sailing, but also, which is most important, how to repair the yacht by my own hands and to sew a sail needed. All that helped me a lot to keep the course I have chosen for my life, regardless the strength of the wind.

Finally, I thank my husband Andrey and my lovely children Anna and Pavel for all the joy they have brought to my life.

Larisa M. Sogacheva (Tatarinova)

List of Publications

This thesis consists of an introductory review, followed by five research articles published in peer-reviewed journals. The papers are reproduced with the kind permission of the journals concerned.

I **Sogacheva, L**., Dal Maso, M., Kerminen, V.-M., and Kulmala, M. 2005. Probability of nucleation events and aerosol particle concentration in different air mass types arriving at Hyytiälä, Southern Finland, based on back trajectory analysis. *Boreal Env. Res.* **10**, 479-491.

II Dal Maso, M., **Sogacheva, L**., Aalto, P.P., Riipinen, I., Komppula, M., Tunved, P., Korhonen, L., Suur-Uski, V., Hirsikko, A., Kurten T., Kerminen, V.-M., Lihavainen, H., Viisanen, Y., Hansson, H.-C., and Kulmala, M. 2007. Aerosol size distribution measurements at four Nordic field stations: identification, analysis and trajectory analysis of new particle formation busts. *Tellus* **59**B, 350-361.

III **Sogacheva, L.,** Hamed, A., Facchini, M. C., Kulmala, M., and Laaksonen, A. 2007. Relation of air mass history to nucleation events in Po Valley, Italy, using back trajectories analysis. *Atmos. Chem. Phys.*, **7**, 839-853.

IV Lyubovtseva, Yu., **Sogacheva, L**., Dal Maso, M., Bonn, B., Keronen, P., and Kulmala, M. 2005. Seasonal variations of trace gases, meteorological parameters, and formation of aerosols in boreal forests. *Boreal Env. Res.* **10**, 493-510.

V **Sogacheva L.**, Saukkonen, L., Dal Maso, M., Nilsson, E.-D., Schultz, D.M., de Leeuw, G., and Kulmala, M. 2008. New aerosol particle formation in different synoptic situations at Hyytiälä, Southern Finland. *Tellus*, **60**B, 485–494.

CONTENTS

1 Introduction

An aerosol can be defined as a dispersion of solid and liquid particles suspended in gas. Atmospheric aerosol particles affect a great variety of processes. They influence natural ecosystems (Lickens *et al*., 1996; Niyogi *et al*., 2004), human health (Davidson *et al*., 2005) and climate (Ramanathan *et al*., 2001; Lohmann and Feichter, 2005). Aerosol particles influence climate by two distinct mechanisms: through the direct reflection of solar radiation, and through the increase in cloud reflectivity, when they act as cloud condensation and ice nuclei, thereby modifying the microphysics, the radiative properties, and the lifetime of clouds (Andreae and Rosenfeld, 2008). Particles that do not absorb light, such as sulfate aerosols, produce a cooling effect, especially in industrial regions (Charlson *et al*., 1991). Soot particles and other light-absorbing aerosols have a warming rather than cooling effect. Even small amounts of absorption may significantly alter the net radiative forcing by particles (Haywood and Shine, 1995; Baumgardner *et al.,* 2004). The Intergovernmental Panel of Climate Change (IPCC 2007, http://www.ipcc-wg2.org/) has reported that, in spite of the increasing knowledge on atmospheric aerosols during the recent decades, uncertainties in the estimation of direct and indirect aerosol effects on global climate still exist. It is essential to reduce the uncertainty in the aerosol climate forcing to make better prognoses of future climate change.

New particle formation (NPF) can be observed as a sudden increase in the number concentration of particles with a diameter smaller than 25 nm (Dal Maso *et al*., 2005). NPF consists of a complicated set of processes that include the production of nanometer-size clusters from gaseous vapours, the growth of these clusters to detectable sizes, and their simultaneous removal by coagulation with the pre-existing aerosol particle population (Kulmala and Kerminen, 2008).

NPF is a frequently observed phenomenon. Observations made during the last two decades demonstrate clearly that new aerosol particle formation and growth occur almost all over the world at varying altitudes (in the free troposphere, close to the tropopause, in the planetary boundary layer) and environments including, for instance, subarctic Lapland and boreal forest, urban areas, industrial and agriculture regions, and coastal environments around the globe (see Kulmala *et al*., 2004c, and references therein). Measurements are performed in both relatively clean environments and industrial regions, thus the anthropogenic influence on environment and climate can be estimated.

The first step of new particle formation, in which substance changes its phase, is nucleation. Once the initial step has occurred, the nuclei of the new phase tend to grow rather rapidly (Seinfeld and Pandis, 1998). Because of the physical and chemical complexity of the atmosphere, it is often a difficult task to identify the most relevant processes causing nucleation. However, if a wide range of measurements is carried out for a long period of time in one location, it may be possible to detect subtle, previously unknown factors contributing to atmospheric particle formation events.

Aitken (1891, 1892, and 1897) first reported evidence for new particle formation in the atmosphere. During the last two decades the interest for the investigation of different mechanisms behind the formation of atmospheric aerosols has been increased considerably. Significant progress has been made in the area of theoretical studies as well as in producing accurate measurements of atmospheric particles. However, the detailed mechanisms still remain unknown for different environments. Although sulfuric acid is one of the most likely candidates thought to be responsible for the formation of the nanometer-sized particles, sulfur chemistry does not always sustain enough sulfuric acid in the atmosphere to explain more then a small fraction of the observed particle-size growth rate.

Many different theories have been proposed to explain nucleation. Hypothetical nucleation theories work in different environmental conditions (H. Korhonen, personal communications). Binary nucleation theory (Kulmala and Laaksonen, 1990) explains particle formation in the free troposphere and in industrial plumes with very high sulphuric acid concentration. Ternary theory (Korhonen *et al*., 1999; Napari *et al*., 2002; Merikanto *et al*., 2007) produces too many particles but predicts the new particle formation correctly in urban environments. Activation (e.g., Kulmala *et al*., 2004b, 2006) and kinetic (McMurry and Friedlander, 1979) theories are based on observations. Theories involving Creege intermediates (Kurtén *et al*. 2007, Bonn *et al.,* 2008) are speculative at this stage as these intermediates cannot be measured. The theoretical progress includes the quantification of the role of dipole-charge interaction in ion clustering thermodynamics and development of ion-mediated nucleation models (Yu *et al.,* 2007, and references therein). Boy *et al.* (2007) investigated different nucleation theories to explain new particle formation within the meteorological boundary layer by a model MALTA and concluded that contribution of ion-induced nucleation contribution is limited to a few percent in the lower troposphere; ternary and binary nucleation with present understanding over- and underestimate measured atmospheric nucleation rates. However, although different nucleation theories exist, the nucleation mechanisms which lead to NPF are poorly known at present.

Gaydos et al. (2005) successfully explained NPF in Pittsburg using a ternary (ammonia-sulfuric acid-water) nucleation model. Spracklen et al. (2008) assumed that sulfuric acid controls the nucleation rate and showed, using a global chemistry transport model, that particle formation is an important source of cloud condensation nuclei. However in Hyytiälä, according to Riipinen et al. (2007), the empirical nucleation coefficients between the formation rate and sulfuric acid concentration differ by several orders of magnitude. Formation rate relates to the sulfuric acid concentration through a power-law relation with exponents typically ranging from 1 to 2. No correlation was observed between measured ammonia concentration and new particle formation in Hyytiälä. To explain the observed growth, other compounds are required in relatively clean environments. Oxidation products of volatile organic compounds are one of the candidates for the growth of freshly formed particles (Bonn and Moortgat, 2002; Bonn *et al.,* 2008; Laaksonen et al., 2008).

During the process of nucleation and growth of atmospheric particles, aerosol dynamics, atmospheric chemistry and meteorology need to be considered (Kulmala *et al*., 2004c). The importance of atmospheric chemistry as well as meteorological conditions for particle formation and growth has been demonstrated in several studies (Weber *et al*., 1995; Birmili and Wiedensohler, 2000; O'Dowd *et al*., 2002a; Bonn and Moortgat, 2002; Kulmala *et al*., 2004b). Nilsson and Kulmala (1998) proposed a parameterization for the mixing-enhanced nucleation rate. The influence of atmospheric waves, such as Kelvin-Helmholtz instabilities, on NPF was investigated by Bigg (1997), Nyeki *et al*. (1999) and Nilsson *et al*. (2000a). The effects of synoptic weather and planetary boundary layer (PBL) evolution, e.g., adiabatic cooling, turbulence, entrainment and convection on aerosol formation were analyzed in different environments: in the marine boundary layer by Russell *et al*. (1998), Coe *et al*. (2000), Pirjola *et al*. (2000) and O'Dowd *et al.* (2002b); in the continental boundary layer by Nilsson *et al*. (2000b), Aalto *et al*. (2001), Buzorius *et al.* (2001), Nilsson *et al*. (2001a,b), Buzorius *et al*. (2003), Stratmann *et al.* (2003), Uhrner *et al*. (2003), Boy *et al*. (2004) and Siebert *et al.* (2004); in the free and upper troposphere by Schröder and Ström (1997), Clarke *et al*. (1999), de Reus *et al.* (1999), Hermann *et al*. (2003) and Khosrawi and Konopka(2003). Boy and Kulmala (2002) demonstrated that favourable conditions for particle formation include low atmospheric water content, low pre-existing particle concentration and high solar radiation. Hellmuth (2004) performed columnar modeling of nucleation burst evolution in the convective boundary layer, and summarized the main results as follows: NPF was always connected with Arctic and Polar air advecting over the site, leading to the formation of a stable nocturnal boundary layer, followed by a rapid formation and growth of a turbulent convective mixed layer, closely followed by NPF.

Within this multitude of theories for new particle formation, several important factors affect nucleation, such as the chemical composition of the atmosphere, water content, and the amount of solar radiation. By investigating these effects we hope to learn more about the processes that trigger new particle formation in the atmosphere.

2 Aim of the Study

The role of atmospheric air masses and synoptic weather has been considered in previous studies during field campaigns with limited duration but not for long time series of measurements. In our studies we tried to answer the following questions: In which types of air masses does NPF occur most frequently throughout the year? What are the properties of air masses which favour NPF? Is the presence of small newly formed particles associated with preferential air mass trajectories, and is the presence of large aerosol surface area only a result of the advection of anthropogenic pollution? How does synoptic weather and cloudiness influence aerosol particle formation in the atmosphere?

To answer the questions raised above we investigated

- the main transport directions of air masses to Hyytiälä, Southern Finland
- the most likely origin of air masses favourable for particle formation in Hyytiälä and the source area of particles of different size modes
- the role of air mass origin in aerosol particle formation at four Nordic measurement stations
- the role of air mass origin in aerosol particle formation at San Pietro Capofiume (SPC) in the Po Valley, Italy, as well as the main source areas of SO_2 measured at SPC
- the meteorological conditions during event and non-event days in Hyytiälä, Southern Finland using the 8-years dataset
- synoptic weather and cloudiness during aerosol particle formation in Hyytiälä

3 Background

3.1 Atmospheric aerosols

Atmospheric aerosol particles originate from various sources: for example, resuspended soil particles, smoke from power plants, sea salt particles from ocean spray, etc. All these particles vary greatly in their ability to affect visibility, climate and human health.

There are two types of fine particles in the atmosphere: primary and secondary. Both primary and secondary particles can be from natural and anthropogenic sources. Primary particulate material derives directly from the entrainment of material from the Earth's surface and from the ocean or from volcanic eruptions; it is also produced by house holds, industrial and traffic emissions. Natural sources of aerosol are, for example, wind blown desert dust, forest fires, soil erosion, and sea spray. Anthropogenic sources include the primary emission resulting from combustion processes. The latter also produce gases that can be converted into particles. The secondary formation of new aerosol particles in the atmosphere, i.e. from gas phase precursors, is important in terms of controlling the background aerosol population, which significantly impacts on climate. Secondary processes leading to the production of ultra-fine particles by nucleation are still poorly understood. The coastal zone is one of the strongest natural sources of secondary aerosol particles, driven by the release of biogenic vapours, which, after undergoing photochemical reactions, lead to the massive production of nucleation mode aerosols (Flanagan *et al.,* 2005). The secondary particulate material can be produced by gas-phase oxidation, including ozonolysis (Shilling *et al*., 2008).

On a global scale, the aerosol can be schematically divided into three main types: marine, continental background (Patterson *et al*., 1980), and urban aerosol (Hussein *et al.,* 2004). By chemical composition, the first two types contain mainly materials from the nearby surface sources, somewhat modified by the coagulation of particles of different origin and by condensation products resulting from gas-phase reactions. Urban aerosol is another type of aerosol, which is formed under the influence of human and industrial activities. The chemical composition of aerosols varies considerably in time and space, but also with particle size (e.g. Pugatscheva *et al*., 2007; Zhang *et al*., 2005).

The main characterizing parameters of atmospheric particles are their size and composition. Both of these parameters can vary greatly in different environments (Figure 1). Nucleation mode particles are the smallest, with a diameter less than about 30 nm. They are produced by gas-to-particle conversion, which occurs in the atmosphere, as well as by combustion processes (Tobias *et al*., 2001; Burtscher, 2005). Nucleation mode particles can be formed by homogeneous nucleation of vapours. They may further grow when gases with low saturation vapour pressure condense on them. The concentration of the nucleation mode particles varies strongly with the strengths of sources and sinks. As observed in Hyytiälä, Southern Finland (Nilsson *et al.,* 2001a; Sogacheva *et al.,* 2005), several stations located in Northern Europe (Tunved *et al*., 2005; Dal Maso *et al.,* 2007),

and the continental background site Melpitz in Eastern Germany (Engler *et al.*, 2006) freshly nucleated particles may rapidly form in clean air masses and grow to larger sizes, whereas in air masses containing larger pre-existing aerosols coagulation and condensation remove them.

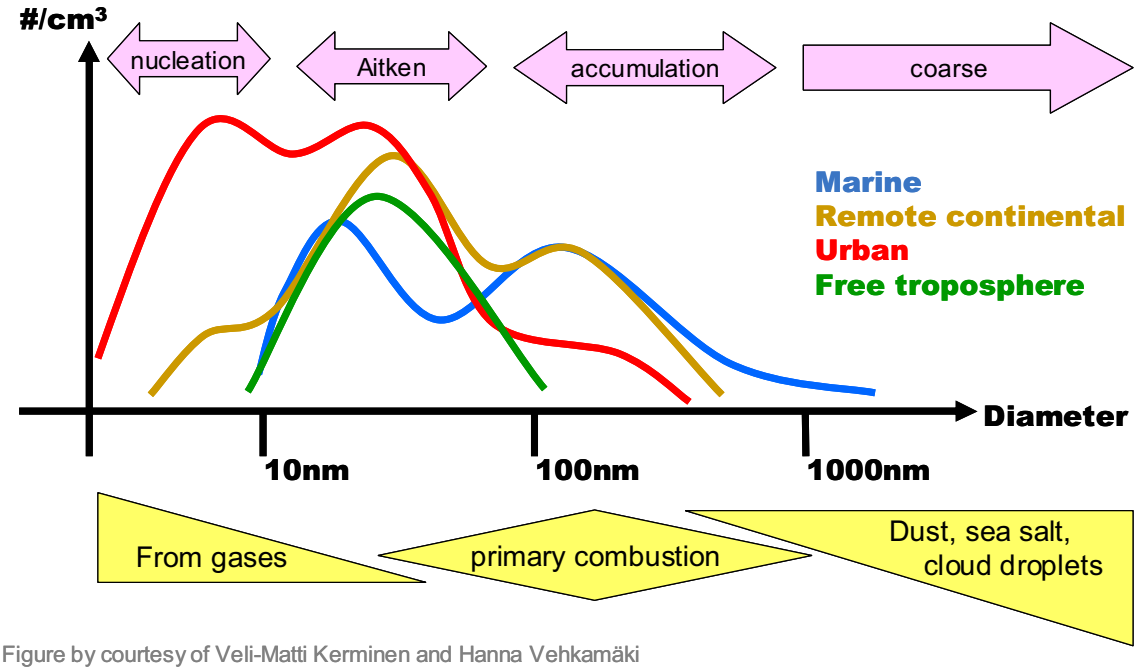

Figure by courtesy of Veli-Matti Kerminen and Hanna Vehkamäki

Figure 1. Aerosol particle size distribution.

Aitken mode particles (30-100 nm) and accumulation mode particles (100 nm-1 µm) are an important source for cloud condensation nuclei (CCN) (Anttila and Kerminen, 2007). Aerosol particles in the accumulation mode are generally produced by primary emissions and sulfate formation in clouds. The other possible sources are coagulation of smaller particles and heterogeneous condensation of vapours onto existing aerosol particles. The accumulation mode particles can be activated to cloud drops and gain considerable mass due to oxidation of sulphur (Komppula, 2005). For particles that act as cloud condensation nuclei, scavenging by precipitation is an important removal mechanism. As a group, fine mode particles (d<1µm) are acidic and contain mostly sulfates, ammonium compounds, hydrocarbons, elemental carbon (soot), toxic metals, and water in the atmosphere (Hinds, 1999).

Coarse particles have diameters larger than about 1 µm. They have different sources and chemical composition compared to the fine (<1 µm) fraction. Coarse particles are wind blown dust, sea spray, and mechanically generated anthropogenic particles such as those from agriculture and surface mining. Coarse particles are basic and contain most of the crustal materials and their oxides, such as silicon, iron, calcium, and aluminum, as well as

sea salt and vegetation debris (Hinds, 1999). Due to their high deposition rate through interception, impaction and sedimentation, the concentration of particles in the coarse fraction varies strongly with the wind speed, the structure of the ground and the distance to large sources (Warneck, 1988).

Several processes influence the formation and the growth of the particles; the most common ones are nucleation, condensation and coagulation. Nucleation is considered to be the first and critical step in NPF. Homogeneous nucleation is one of the possible candidates leading to the formation of thermodynamically stable clusters (TSC) in the absence of existing seed nuclei. Depending on the availability of pre-existing particles and/or condensable vapours, Kulmala *et al*. (2000) hypothesized two possibilities for the growth of TSC to detectable size: (a) when the concentration of pre-existing particles is low, TSCs can effectively grow by self-coagulation when the nucleation rate is $>10^3$ cm^{-3} s^{-1} (Anttila *et al.,* 2004) and (b) in the presence of a large source of available condensable vapours TSCs can effectively grow to detectable sizes and even to Aitken mode sizes by condensation. Homogeneous nucleation is typically unable to produce the nucleation rate needed for new particle formation in atmospheric conditions (Spracklen *et al*., 2005a, 2005b), nevertheless sulfuric acid can be used, together with an empirical activation factor, as an indicator of nucleation rate (Spracklen *et al.,* 2008). In such conditions, the heterogeneous nucleation, which is a process of particle formation and growth in the presence of condensation nuclei or ions, is the key process to start the formation of new clusters (Lazaridis *et al*., 1992).

Condensation growth and coagulation loss are competing processes for nucleation mode particles. The more effective the condensation growth, the larger fraction of nucleated particles can survive to detectable sizes (Kulmala *et al.,* 2000). Condensation is the main mechanism that explains the growth of atmospheric aerosol particles. The condensation sink (CS) is a parameter that indicates how rapidly condensable vapour molecules will condense on the existing aerosol (Dal Maso *et al.,* 2002). The coagulation loss is the removal rate of particles with a certain radius due to the collision and sticking of the particles of any size, when a particle of bigger size and often of different chemical properties is formed.

The aerosol particles are subject to various removal mechanisms: among the most important ones is deposition, which removes the aerosol mass from the atmosphere. Deposition of the atmospheric particles takes place via both dry and wet processes. Wet deposition includes precipitation scavenging in which particles are deposited in rain, snow, fog, cloud water and mist when these intercept the surface (Flossman *et al*., 1985; Flossman, 2002; Foret *et al.,* 2006; Tost *et al*., 2007). According to Loosmore and Cederwall (2004) wet deposition played an important role in the simulated fate and transport, removing as much as 70% of the released aerosol. Dry deposition is the direct transfer of particles to the ground, through sedimentation, impaction or diffusion. Dry deposition is considered more effective for coarse particles and elements such as iron and manganese, whereas wet deposition generally is more effective for fine particles and elements such as cadmium, lead and nickel (Brimblecombe, 1996). Wet deposition is more effective for sulfate and nitrate (Ro *et al*., 1988; Furiness *et al*., 1998).

The residence time, or life time, of atmospheric aerosol particles is a function of various removal processes, such as dry deposition by impaction, diffusion and sedimentation, resuspension as well as wet deposition by rain drops. It has important implications with regard to the transport and distribution of the substances associated with particular matter. The residence time of aerosols depends on their size (Jaenicke, 2008). The atmospheric life time of particles varies from hours to several days. Due to their low total deposition rate, the Aitken and accumulation mode particles have longer residence times in the atmosphere compared to nucleation mode particles. Because of their large size, the coarse particles readily settle out from the atmosphere by gravitation or impact on surfaces, so their lifetime in the atmosphere is only a few hours or days. Recently Tunved *et al.* (2005) estimated the life- or turnover- time of particles of different sizes analyzing the particle transport between two Scandinavian measurement stations. They found that during northerly transport the shortest turnover times are 1.6 - 1.7 days for particles in the nucleation mode and around 2.4 days for particles in the Aitken mode. Nilsson and Rannik (2001) observed a turnover time around 3.5 days for the Aitken mode in the central Arctic on the basis of eddy correlation aerosol deposition. Thus, for the nucleation mode it is not correct to talk about source regions at distances of several thousands kilometers from the recipient. However, the air mass origin and trajectory determine the common properties (temperature, water content, chemical composition, precursor sources, etc.) of the air that may affect new particle formation.

3.2 Major weather systems

Weather most often results from temperature differences between one place and another. Surface temperature differences in turn cause pressure differences. The changes of pressure over continents, and practically the changes of pressure gradients between land and water, determine the direction of the air currents (Battan, 1978). The mechanism maintaining the general circulation of the atmosphere is the large scale pattern of wind and pressure that persists throughout the year or recurs seasonally (Barry and Chorley, 2003). The resulting horizontal pressure gradient accelerates the air from high to low pressure, creating wind, and Earth's rotation causes curvature of the flow via the Coriolis effect.

Wind patterns and precipitation in the Arctic are governed by the low-pressure systems that form over the North Atlantic and the Bering Sea (Figure 2), bringing warm, moist air northward. These weather systems, the Icelandic and Aleutian lows, gather moisture over open water and dump it as precipitation when the air is forced to rise. Another important consequence of the pressure systems location is that after the passage of a low-pressure system, the Azores High may ride over Western Europe, resulting in transport of polluted air from Europe along the southern flank of the ridge.

There is a general consensus that a difference between winter and summer conditions exists. Air-transport patterns are highly dependent on season and on the position of the major weather systems (Figure 2).

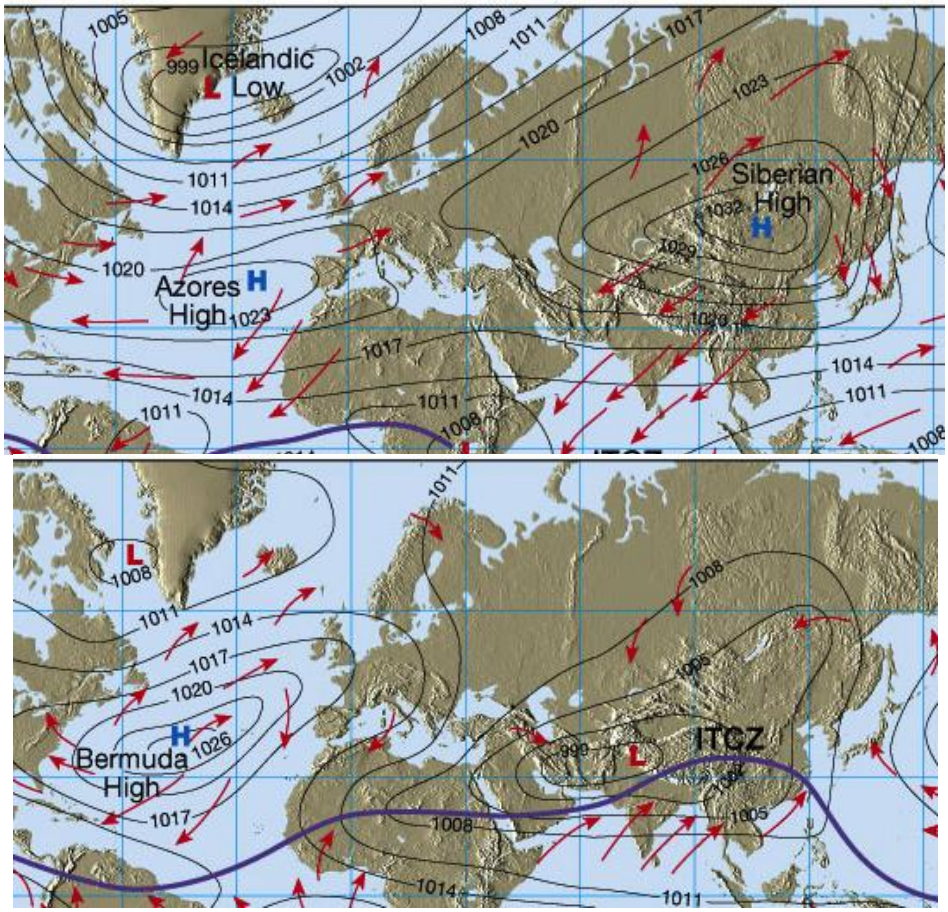

Figure 2. Major weather systems (Islandic Low, Azores High, Siberian High, Bermuda High pressure systems are illustrated by isobars and main wind directions are presented by red arrows) and the intertropical convergence zone (ITCZ) in January (upper panel) and July (lower panel). (From: Lutgens, Frederick K.; Tarbuck, Edward J. *The Atmosphere: an Introduction to Meteorology, 8th Edition*, 2001. Reprinted from permission of Pearson Education Inc., Upper Saddle River, NJ).

In winter and spring, an intense high-pressure system over Siberia pushes the Arctic front far to the south, so that important polluted areas of Eurasia are actually within the Arctic air mass, the lower one-to-two kilometers of which can move contaminants across the pole. Low wind speeds and temperature inversions caused by the cold winter weather, allow contaminants to accumulate in the atmosphere (Charlson and Heintzenberg, 1995). In the warm seasons, the Icelandic and Aleutian lows weaken considerably (Musk, 1988). The atmospheric removal processes are much more efficient in summer than in winter; concentrations of anthropogenic pollutants are lower in summer. Summer is also warmer,

allowing for cloud formation and for drizzling rain that removes contaminants from the air before they are carried away. Moreover, sunlight during the summer months allows for photochemical degradation of some contaminants and their conversion into particles.

3.3 Air masses

Several studies have been carried out to investigate whether the origin of the air influences aerosol particle formation. What is an air mass? Why do air masses differ in their properties? To answer these questions we go deeper into the air mass definition and classification.

An air mass is a large body of air of relatively similar temperature and moisture content characteristics covering thousands of square kilometers (Stull, 1988). Each type of air mass is associated with typical thermodynamical properties. Three factors determine the nature and degree of uniformity of air mass characteristics: (1) the nature of the source area where the air mass obtains its original qualities; (2) the direction of movement and changes that occur as an air mass moves over long distances; and (3) the age of an air mass (Barry and Chorley, 2003). An air mass develops its characteristic properties by remaining over a particular region for period long enough to allow its vertical distribution of temperature and moisture to reach equilibrium with underlying surface (Battan, 1978). A source region can have one of four temperature attributes: equatorial, subtropical, polar or arctic. Air masses are also classified as being either continental (c) or maritime (m) in terms of moisture characteristics determined from extent of continental/marine influence. Combining these two categories, several possibilities are commonly found: maritime Subtropical (mS), continental Subtropical (cS), maritime Polar (mP), continental Polar (cP), continental Arctic (cA) and marine Arctic (mA). The diagram in Figure 3 describes the source regions and common patterns of movement for the various types of air masses associated with Northern Europe. During the period of interest (2003-2005) equatorial air masses never reached the Hyytiälä measurement station.

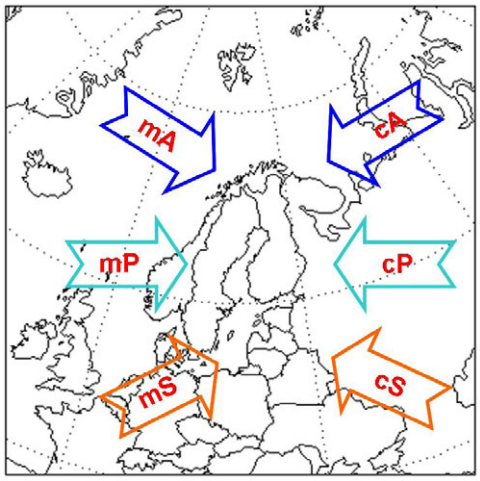

Figure 3. Main types of air masses observed in Scandinavia (marine Arctic (mA), continental Arctic (cA), maritime Polar (mP), continental Polar (cP), maritime Subtropical (mS), continental Subtropical (cS))

As an air mass moves away from its source region, it is affected by different heat and moisture exchanges with the ground surface and by dynamic processes in the atmosphere (Stuhl, 1988). Each type of air mass can be regarded as transformable into another, reflecting the influence from sea or continents on energy content (heat and water vapour), but also on trace gases and suspended particulate matter. When transformation occurs, the air mass will gradually adopt typical properties concerning temperature and water content as well as emissions coming from the underlying surface.

3.4 Atmospheric Fronts

Frequently, two air masses, especially in the mid latitudes, develop a sharp boundary or interface, where the temperature difference between them becomes intensified. Such an area of intensification is called a frontal zone or a front (McIlveen, 1992). The boundary between the warm and cold air masses always slopes upwards over the cold air. This is due to the fact that cold air is much denser than warm air. The sloping of warm air over cold air leads to a forced uplifting (frontal lifting) of the warm air if one air mass is moving toward the other. In turn, the uplifting causes condensation due to decreasing temperature; condensation leads to cloud formation and precipitation along the frontal boundary.

The weather conditions observed at cold fronts (Figure 4, upper panel) are variable, depending upon stability of the warm sector air and the vertical motion relative to the frontal zone (Barry and Chorley, 2003). Cold fronts move between 15 to 50 kilometers per hour, this is more then twice as fast as a warm front, since cold air is denser than warm air and rapidly replaces the warm air. In the Northern Hemisphere a cold front moves in a southeast to east direction due to pressure gradients and the Coriolis effect (Stull, 1988). Before the cold front passage the temperature is relatively high, the pressure falls steadily, the prevailing wind direction is south (S) to south-west (SW). With the passage of the cold front, the wind veers sharply, turning to north-west (NW), the pressure begins to rise and temperature falls. The formation of clouds and precipitation at the frontal zone is caused by frontal lifting. High altitude Cirrus (*Ci*) clouds are found well in advance of the front. Above the surface location of the cold front, high altitude Cirrostratus (*Cs*) and middle altitude Altocumulus (*Ac*) are common. Precipitation is normally found just behind the front where frontal lifting has caused the development of towering Cumulus (Cu) and Cumulonimbus (*Cb*) clouds. Cold fronts may feature narrow bands of thunderstorms and severe weather, and may on occasion be preceded by squall lines. The weather usually clears quickly after a frontal passage. Symbolically, a cold front is represented by a solid (blue) line with triangles along the front pointing towards the warmer air and in the direction of movement (the example of a synoptic map see in section 6.3, Figure 6, left panel).

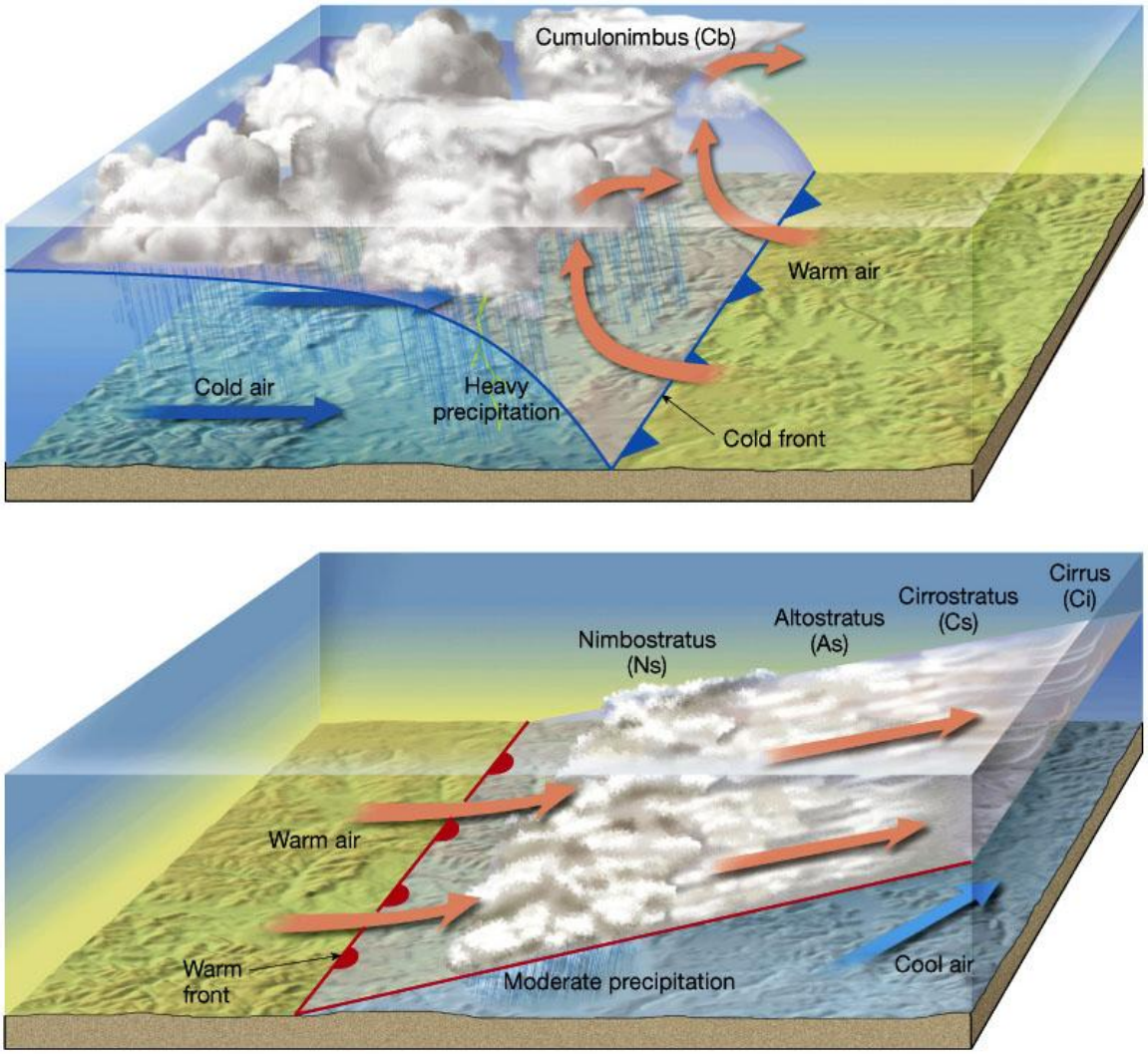

Figure 4. Schematic structure of cold (upper panel) and warm (lower panel) atmospheric fronts. (From: Lutgens, Frederick K.; Tarbuck, Edward J. *The Atmosphere: an Introduction to Meteorology, 8th Edition*, 2001. Reprinted from permission of Pearson Education Inc., Upper Saddle River, NJ).

A warm front is the transition zone in the atmosphere where an advancing warm, moist air mass replaces a retreating cold air mass. On a synoptic map (as an example see section 6.3, Figure 6, left panel) a warm front is drawn as a solid (red) line with half-circles. The position of the half-circles shows the direction of frontal movement. Warm fronts move about 10 km h^{-1} in a north-east (NE) direction. This is less than half the speed of a cold front, because cold air is more dense and harder to remove from the earth's surface. The formation of clouds and precipitation ahead of the frontal zone is caused by gradual frontal lifting (Figure 4, bottom panel). High altitude *Ci, Cs* and middle altitude *As* clouds are found well in advance of the front. About 600 kilometers ahead of the front, Nimbostratus (*Ns*) clouds occur. These clouds produce precipitation in the form of snow or rain.

Between the *Ns* clouds and the surface location of the warm front, low altitude Stratus (*St*) clouds are found.

Before the warm front passage the prevailing wind direction is south or south-east. The temperature is relatively low and the pressure is usually dropping. The frontal boundary has a very gentle slope of the order or 0.5-1°, so the cloud systems associated with the upper portion of the front herald its approach some twelve hours or more before the arrival of the surface front. Different types of *Ci* and *St* can be observed. Depending on season light-to-moderate rain, snow, sleet, or drizzle happen. During the warm front passage, winds are variable, temperature increases after the frontal passage. Pressure levels off, then slightly rises, followed by a decrease. Scattered Stratocumulus (*Sc*) may exist in the clear sky. Precipitation occurs rarely in the form of light rain or showers. At the passage of the warm front the wind veers, the temperature rises and a pressure drop is observed.

Being more mobile as compared to a warm front, the cold front rotates around the intensifying low pressure center and catches the warm front. This forms an occluded front, which is the boundary that separates the new cold air mass (to the west) from the older cool air mass already in place north of the warm front. Two types of occluded fronts are generally recognized (McIlveen, 1992). A cold type occluded front occurs when the air behind the front is colder than the air ahead of the front. When the air behind the front is warmer than the air ahead of the front a warm type occluded front is produced. Symbolically, an occluded front is represented on a weather map by a solid line with alternating triangles and circles pointing the direction in which the front is moving (Figure 6, left panel). On colored weather maps, an occluded front is drawn with a solid purple line.

During an occlusion front passage the prevailing wind direction turns from south-south-east to west-north-west. The temperature changes are not as noticeable as during the cold/warm passages. Pressure usually falls before the front and increases after the front passage. *Ci* and *Cs* usually disappear after the occlusion front, whereas *Ns* still exists. The intensity of precipitation varies from light to heavy around the frontal area.

While the meteorological conditions along the atmospheric fronts vary significantly, we expect that they influence aerosol particle formation in different ways. One of the tasks in our studies was to analyze synoptic situations that favour new particle formation.

4 Air mass back trajectories

Since the new particle formation process was shown to be influenced by the air mass properties (Nilsson *et al*., 2001a), an important issue in understanding the formation of aerosol particles is to follow the history of the air arriving at the measurement point. Air mass back trajectory analysis, which has been used during the last few decades, is one of the methods for source area definition. The evolution of the aerosols in an air mass can be described as functions of emission sources, time spent over sources, as well as meteorological parameters controlling the evolution of the aerosols.

4.1 Back trajectories application

The importance of meteorological transport process for the interpretation of measurements of trace gas concentrations has been the subject of extensive research. The variation of the concentrations of species in the atmosphere, which is determined by emission sources, transformation and removal processes, can be attributed to two different processes: vertical mixing and horizontal, long-range transport. Back trajectory analysis is instrumental in establishing the spatial domain of sources contributing to the composition of air parcels arriving at selected receptor sites. In connection with the measured concentrations, trajectory statistics provide the potential source regions of the pollutants.

Aerosol data have been analyzed based on air mass trajectories (e.g., Kemp, 1993; Mukai and Suzuki, 1996). Soluble ion chemistry of atmospheric aerosols and SO_2 concentrations over the eastern North Atlantic have been explained by Andreae *et al*. (2000) by using air mass trajectory analysis. Using back trajectory data, Birmili *et al.* (2001) investigated the role of air masses in determining particle size distributions and showed the significantly enhanced number concentration of Aitken and accumulation mode particles during periods of pronounced continentally aged air. Bonasoni *et al.* (2004) studied aerosol-ozone correlations during dust-transport episodes using trajectory statistical analysis.

Vasconcelos *et al.* (1999) described conditional frequency analysis as an investigation of the spatial distribution of back trajectories segment endpoints on a grid superimposed over the region containing the receptor. By means of back trajectories, Siebert *et al.* (1998) studied long-range transport of pollutants from the Po valley to High-Alpine sites. Wotava and Kröger (1999) tested the ability of trajectory statistics to reproduce emission inventories of air pollutants and concluded that general patterns of the spatial emission density distribution can be correctly reproduced.

Moody and Galloway (1988) were the first to consider trajectory coordinates as the clustering variables. Cluster analysis is a multivariate statistical technique increasingly used in air pollution research (Abdalmogith and Harrison, 2005). This method involves splitting the data set into a number of groups which need to be homogeneous and as

distinctly different from each other as possible. Brankov *et al*. (1998) presented a trajectory clustering methodology for interpreting a 5-year time-series of ozone concentrations at Whiteface Mountain, United States. Cape *et al*. (2000) used cluster analysis to interpret ozone concentrations measured at Mace Head, Ireland, and found clear differences among the trajectories for the measured ozone concentration. Buchanan *et al*. (2002) applied the cluster analysis technique to investigate long-range transport of airborne particle concentrations in Edinburgh, Scotland. Tunved *et al.* (2003, 2004) used cluster trajectory analysis of one-year aerosol size distribution data to explain changes in aerosol properties.

In the present study we used trajectory statistics to determine the main transport direction of air masses, calculated probability of new particle formation as a function of air mass history, studied the long-range transport to explain the concentration of nanoparticles at the measurement station, and used the source-receptor technique to reveal the main sources of SO_2 measured concentration.

4.2 Back trajectories accuracy and limitation for use

The accuracy and limitations of trajectory calculations have been investigated by several researchers (Stohl, 1998; Stunder, 1996; Kahl, 1996). These authors concluded that the accuracy of an individual air mass trajectory is ultimately limited by temporal and spatial resolution of meteorological observations, by measurement and analysis errors and by any simplifying assumptions used in the trajectory model. Despite the fact that trajectory models have been employed successfully to investigate complex transport processes such as recirculation of pollutants (Tyson *et al.,* 1996), it is virtually impossible to describe transport phenomena in turbulent flows by calculating single trajectories. Errors associated with a single trajectory are reduced when daily trajectories (Stohl, 1998; Brankov *et al*., 1998) or ensemble trajectories (Scheele and Siegmund, 2001) are considered to characterize the air parcel path.

5 Site description and measurements

Most of the studies presented in thesis have been done using the measurements at the SMEAR II station, which is located in the Hyytiälä Forestry Field Station of the University of Helsinki in Southern Finland (61°51' N, 24°17' E, 180 m a.s.l.). The station was designed to study mass and energy flows in atmosphere-vegetation-soil continuum (Hari and Kulmala, 2005). Around the station, which is located in boreal forest, there is a homogeneous 45-years-old Scots pine stand. The dominant stand height is about 14 m. Rannik (1998) describes the micrometeorology of the site.

Besides the Smear II station, we used in our studies the aerosol size distribution data from Nordic field stations, all situated inside boreal forest. The SMEAR I station at Värrio (67°46' N, 29°35' E, 180 m a.s.l.), situated in a rural area of Lapland. The Aspvreten measurement station(58°46' N, 17°24' E, 25 m a.s.l.) is located near the Swedish coastline in Sörmland, where the anthropogenic influence is considered small (Tunved *et al.,* 2003). The Sammaltunturi Global AtmosphericWatch station Palas (67°58' N 24°07' E, 565 m a.s.l.), operated by the Finnish Meteorological Institute, is situated on top of a field in western Lapland. The Pallas area is in the sub-Arctic region near the northern limit of the boreal forest zone.

The boreal forests play an important role in both climatic regulation and carbon cycling and are characterized by large volatile organic compound (VOC) emissions (Hakola *et al*., 2003). Organic compounds comprise a large fraction of the global aerosol burden and there is growing evidence that naturally emitted terpenes contribute notably to gas-to-particle conversion (Bonn and Moortgat, 2002; Bonn *et al.,* 2007). In the boreal region of Northern Europe, monoterpenes are abundant (Tarvainen *et al*., 2007) with concentrations depending on season, boundary layer conditions and temperature.

The San Pietro Capofiume (SPC) measurement station (44°39' N, 11°37' E, 11 m a.s.l.) is located at about 30 km northeast from the city of Bologna, in the Po Valley, Italy. The Po Valley is densely populated, highly industrialized and known to have a relatively high level of anthropogenic pollution. The local atmospheric circulation features, determined by the dynamic effect of the Alps, often favour the development of critical pollution episodes (Finardi and Pellegrini, 2004).

At all these sites aerosol size distribution are measured using a Differential Mobility Particle Sizer (DMPS), an instrument consisting of a bipolar charger, one or two Differential Mobility Analyzers (DMA) to separate aerosol particles with different sizes and one or two Condensation Particle Counters (CPC) to detect the particles (Aalto et al., 2001). Depending on the setup, the DMPS can measure particle size distributions with a high size resolution from sizes as low as 3 nm. The sampling time per distribution is usually *ca.* 10 min.

6 Methods

6.1 Classification of new particle formation

A particle formation event is registered when a mode of fresh particles appears, manifested as a sudden increase in the number concentrations in the nucleation mode, with sizes less than 25 nm in diameter. The classification method used in the creation of the database is based on the flowchart published by Dal Maso *et al.* (2005), which classifies a day into one of three main groups: event (classes Ia, Ib or II), non-event, and undefined. In this study we applied these rules as follows. To classify as an event a day the following criteria had to be fulfilled: a) particles smaller than 25 nm had to be present; b) these particles had to form a mode that had not existed before; c) this mode had to persist at least 1 hour; d) the particles in the new mode had to grow in size with time. A failure to fulfil the criteria a), b) and c) resulted in a classification to the non-event class and a failure to fulfil criterion a) or d) resulted in a classification to the undefined class. The classification was performed so that the probability of false positives for events was minimized, even if this meant an increased amount of false negatives for events.

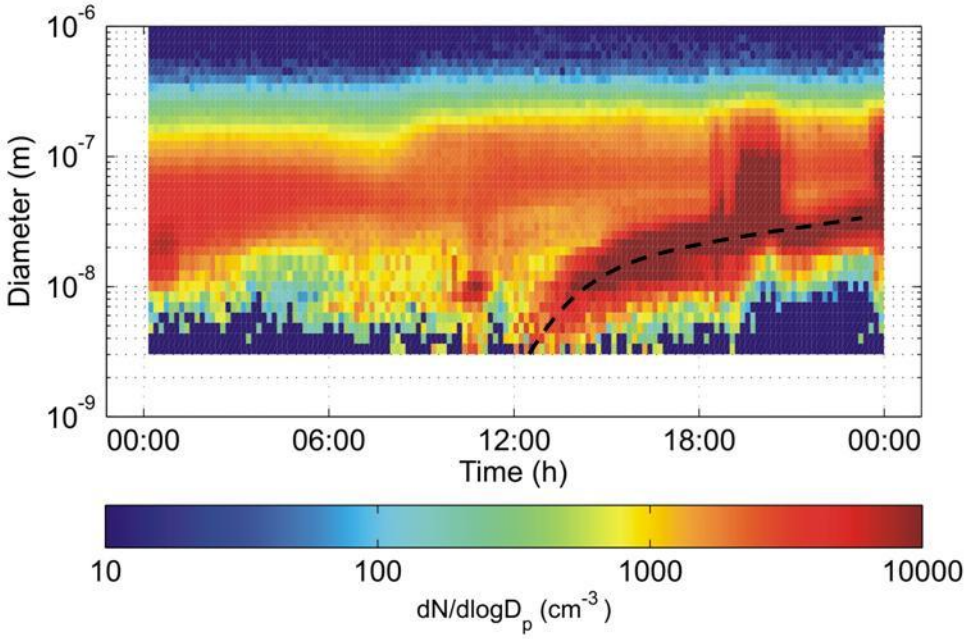

Figure 5. New particle formation event measured in Hyytiälä with a DMPS system on the 24th of April, 2006, at the SMEAR II station at Hyytiälä, Southern Finland (dashed line – median size of the mode formed by the newly formed particles; by fitting that line the approximate growth rate of the particles can be estimated). (Figure by courtesy of M. Dal Maso).

When a day was classified as an event day (Figure 5), it was subsequently classified according to the possibility of further analysis. If it was possible to obtain the formation and growth rate of new particles, the day was classified as a class I event. For some days, while it was clear that the day was an event day, it was not possible to obtain these characteristics, due to, for example, strong fluctuations in the particle concentration. Such days were classified as class II events. Class Ia events are event days during which the new mode is clearly distinguishable from the pre-existing particle population for the whole duration of the event, making these days good candidates for modeling case studies (Korhonen *et al*., 2004, Boy *et al.,* 2006). On these days, the total number concentration is dominated by the new particles. The rest of the class I events were classified as class Ib; it contains events during which the growing mode of new particles is at some time obscured by or superimposed to the pre-existing distribution.

6.2 Back trajectories calculation

To analyze the source and transport pathways of air masses, a trajectory analysis was made. The back trajectories were calculated using the HYSPLIT_4 model, developed by NOAA/ARL (Draxler and Hess 1998). HYSPLIT_4 is a single particle Lagrangian trajectory dispersion model. The model runs were made using the Global FNL meteorological archive with a spatial resolution of 191x191km. The FNL meteorological dataset was produced by the US National Center for Environment Prediction (NCEP). Air parcel back trajectories were calculated typically 96 hours backwards in time. However, some trajectories were missing or shorter than 96 hours in duration, since the FNL archive data has some gaps when output meteorological data are not available. Current literature suggest that the error accompanying HYSPLIT-generated trajectories can be estimated to be anywhere from 15% to 30% of the travel distance (Stohl 1998; Draxler and Hess 2004). In our studies we consider the sources of aerosol particles on a regional scale, so the accuracy of trajectory calculations is sufficient to reveal differences in the particle concentration for different types of air masses. The second benefit was the long time series of the measurements and hence the high number of the trajectories used in calculations.

6.3 Air mass analysis

To classify the type of the air masses arriving at Hyytiälä and the passages of the atmospheric fronts over Southern Finland, synoptic maps of the surface (Figure 6, left) and 850-hPa levels from the Berliner Wetterkarte maps (http://www.berliner-wetterkarte.de/) from the German Weather Service (Deutscher Wetterdienst, DWD) were examined together with the satellite images (Figure 6, right) from the Advanced Very High Resolution Radiometer (AVHRR). The AVHRR is an imager (http://www2.ncdc.noaa.gov/docs/klm/html/c3/sec3-1.htm) that can be used for determining cloud cover.

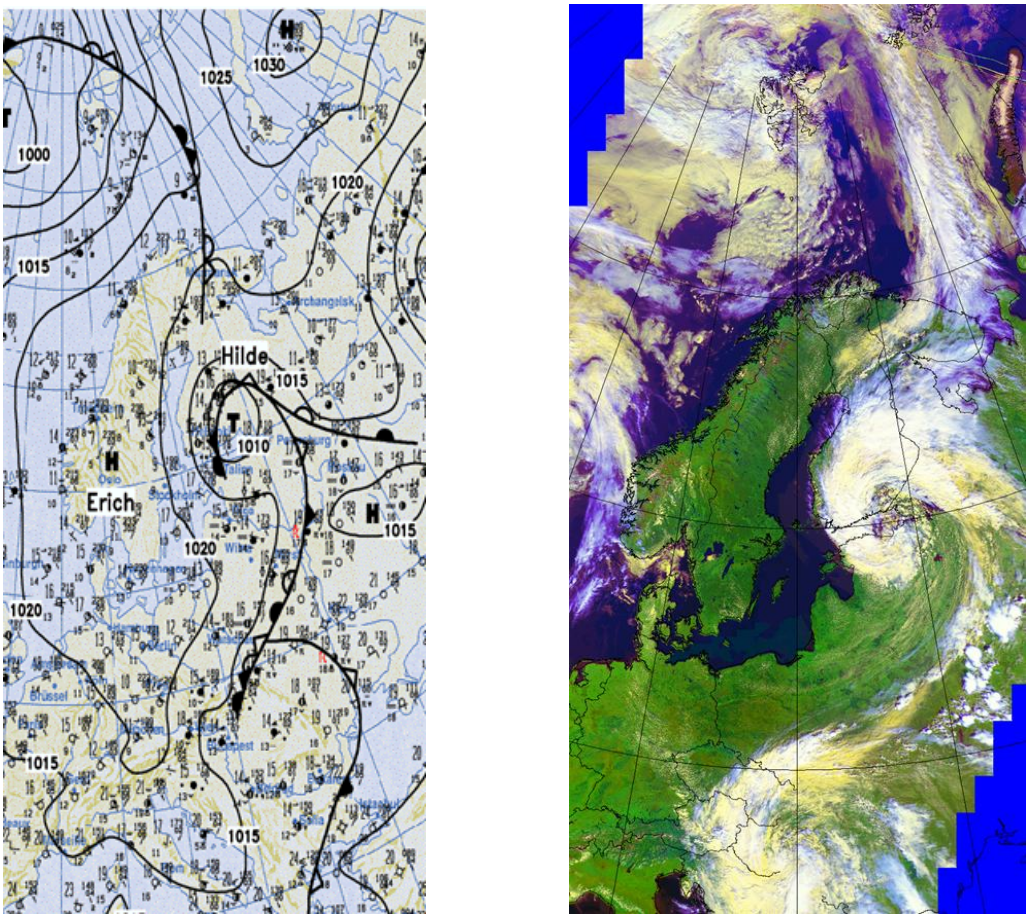

Figure 6. Example for Berliner Wetterkarte (synoptic map, left) and AVHRR satellite image (right) over Central Europe and Scandinavia, for 29th of July, 2004.

Synoptic weather maps with a time resolution of 12 h were examined for the occurrence of fronts and for air mass classification in the lower troposphere (Bissolli and Dittmann, 2001). We used the air mass classification made daily at Berliner Wetterkarte for 0000 UTC (local time at Hyytiälä is UTC plus 2 h, but 3 h during daylight savings time in the summer). The air mass classification recognizes Arctic, Polar and Subtropical air masses, each divided into marine, continental, and transition from marine to continental (those which lose moisture while traveling over the continent.) (Beilage zur Berliner Wetterkarte, http://www.met.fu-berlin-de/~manfred/luftmassen.html).

6.4 Cloudiness

Cloudiness over Southern Finland was analyzed from the AVHRR imagery in one of two time intervals. On event days, cloudiness was estimated for approximately 4 h around the event starting time, whereas on non-event days cloudiness was estimated in a time window 0800–1400 Local Time, which covers most of the starting times of particle formation events at Hyytiälä (Dal Maso *et al.,* 2005). Thus, the days were classified according to the sky cover (in octas) into the following groups: clear sky (0 octas), near-clear sky (1–2 octas), isolated clouds (3–4 octas), broken clouds (5–6 octas), nearly unbroken cloudiness (7–8 octas), and overcast (8 octas, unbroken cloudiness). Hereinafter, we call days with cloudiness higher than 4 octas as 'high-cloud-amount' days and cloudiness less than 5 octas 'low-cloud-amount' days.

7 Review of Papers and Author's Contribution

The motivation and approach of each article as well as the author's contribution are described below. Sections 7 and 8 detail the main results and conclusions from these articles.

Paper I

In Paper I we investigated the probability of new ultrafine particle formation as well as the spatial sources of the nucleation, Aitken and accumulation mode particles in Hyytiälä, Southern Finland, by means of air mass back trajectories analysis.
In this paper I calculated and analyzed the back trajectories and wrote most of the text.

Paper II

In paper II we analyzed aerosol size distributions measured at the Hyytiälä, Värriö and Pallas stations in Finland and at the Swedish station Aspvreten, over a period of several years. We identified occurrences of new particle formation events and obtained characteristics for the events from the size distribution data and analyzed the directions from which air masses leading to new particle formation arrived.
In this paper I carried out the back trajectories calculation as well as analysis, and contributed to part of writing.

Paper III

In Paper III we studied the transport of air masses to San Pietro Capofiume (SPC) in the Po Valley, Italy, by means of back trajectory analysis. Our main aim was to investigate whether air masses originate from different regions on days with and without new particle formation events, using three years of continuously recorded data at SPC. We analyzed meteorological conditions along the back trajectories of air parcels for event and non-event days. By means of source-receptor analysis we investigated the influence of distant sources of SO_2 to SO_2 concentrations measured at SPC.
In this paper I calculated the back trajectories, analyzed them and wrote most of the paper.

Paper IV

In Paper IV we analyzed aerosol particle number concentration and size distribution data together with meteorological and gas concentrations data recorded at Hyytiälä to investigate the possible links between physical, chemical, and meteorological parameters for event and non-event days for different seasons. The seasonal patterns of average trace gas concentrations (O_3, H_2O, NO_x, SO_2), UVA radiation, temperature, relative humidity and condensation sinks were established and their influence on the seasonal dependence of the formation and growth rates of nucleation mode particles was evaluated.
I did most of calculations (except for the effect of ozone on chemical lifetime) and contributed to part of writing.

Paper V

In Paper V we examined the meteorological and synoptic conditions favourable for new particle formation in Hyytiälä. Synoptic weather maps and satellite images over Southern Finland were analyzed for the period 2003-2005, focusing mainly on air mass types, atmospheric frontal passages, and cloudiness.
I did most of the analysis of synoptic weather maps and satellite images, all the calculations, and also wrote most of the paper.

8 Main Results

8.1 Air mass transport directions favourable for new particle formation

8.1.1 Hyytiälä, Southern Finland

Several previous studies have investigated the effects of air masses and synoptic weather on aerosol particle formation at Hyytiälä, Southern Finland. During the BIOFOR 3 campaign (11 March–30 April 1999), Nilsson *et al*. (2001a) reported that nucleation was favored in Arctic, and, to some extent, Polar air masses. The aim of our study was to use back trajectory analysis to investigate the main air mass transport direction to Hyytiälä and to indentify source areas of air masses which bring to Hyytiälä conditions during which new particle formation occurred most often during the period 1997-2003.

Prevailing air mass transport directions for Hyytiälä, Southern Finland, are from south-west (SW) to north-west (NW), whereas transport from north (N) and north-east (NE) is of secondary importance (**Paper I**). Seasonal differences in prevailing flow directions exist but they are minor. In spring, the frequency of SW to NW directions increases. In summer, western flows prevail. In autumn, the frequency distribution of flow directions is smoother compared to other seasons. Since the horizontal pressure gradient is smaller in summer, the speed of air flows is lower and air masses travel shorter distances. Hence the influence of the local sources increases.

The dominant role of Arctic and Polar air masses causing new-particle formation is clearly seen during all seasons (**Paper I**). On average nucleation events occur more often in clean marine air originating from North Atlantic and Arctic areas (Figure 7).

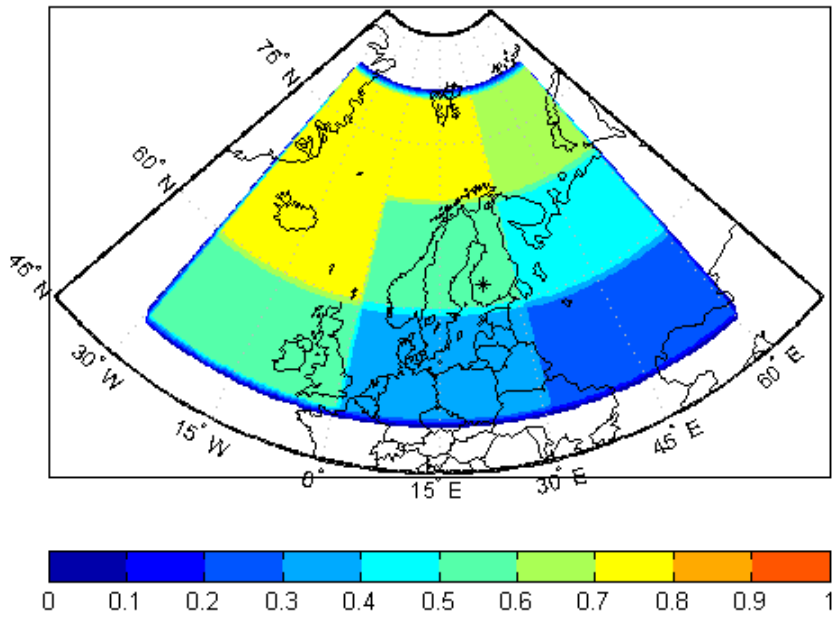

Figure 7. Distribution of nucleation event probabilities during the years 1997-2003. The color in a grid cell describes the probability of a nucleation event at the receptor location (*, Hyytiälä) for corresponding trajectory direction.

In typically more polluted continental air masses, originating over Central and Eastern Europe, new-particle formation was very rarely observed. More explicitly, the role of Arctic and Polar air masses in the nucleation processes was more clearly observed in spring when the frequency of new particle formation reached its maximum. The results allow us to conclude that a lower temperature, low amount of anthropogenic pollution, and in some cases lower humidity, are the necessary factors associated with new particle formation events in Southern Finland.

Clear differences were observed in sources of particles of different sizes (**Paper I**). In all seasons (Figure 8) Arctic and North Atlantic areas were the sources of nucleation mode particles. The influence of that region on new particle formation reached a maximum in spring (**Paper I**) when highest concentrations for all modes are observed in Hyytiälä. Being an industrial region, the south-west of Russia and central Europe were the main sources of accumulation mode particles during all seasons. The formation and/or production of accumulation mode particles over those regions may be connected with high concentrations of anthropogenic combustion products, including those from fossil fuel combustion.

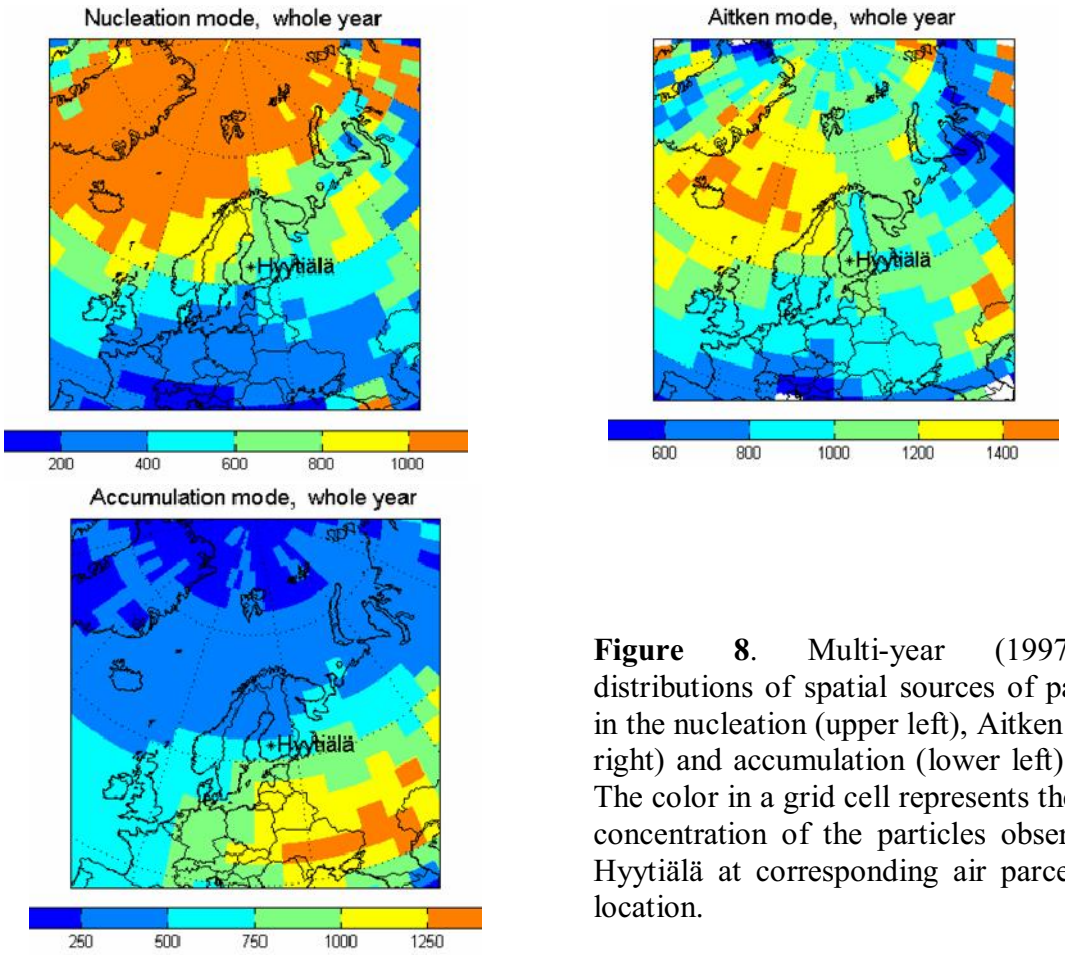

Figure 8. Multi-year (1997-2003) distributions of spatial sources of particles in the nucleation (upper left), Aitken (upper right) and accumulation (lower left) mode. The color in a grid cell represents the mean concentration of the particles observed at Hyytiälä at corresponding air parcel back location.

Air masses with high concentrations of Aitken and accumulation mode particles are characterized by high condensation and coagulation sinks, which decrease the lifetime of vapours participating in the new particle formation process and remove newly formed ultrafine particles (Kulmala *et al*., 2004a, Kulmala *et al*., 2005). Our analysis shows that the highest condensation sinks measured in Hyytiälä are observed in the continental air masses arriving at Hyytiälä from Eastern Europe and Central Russia (Figure 9). The lowest condensation sinks correspond to the NW air mass transport direction (**Paper I**). This may be a key factor in the observed tendency of northerly, cleaner air masses favouring new particle formation events.

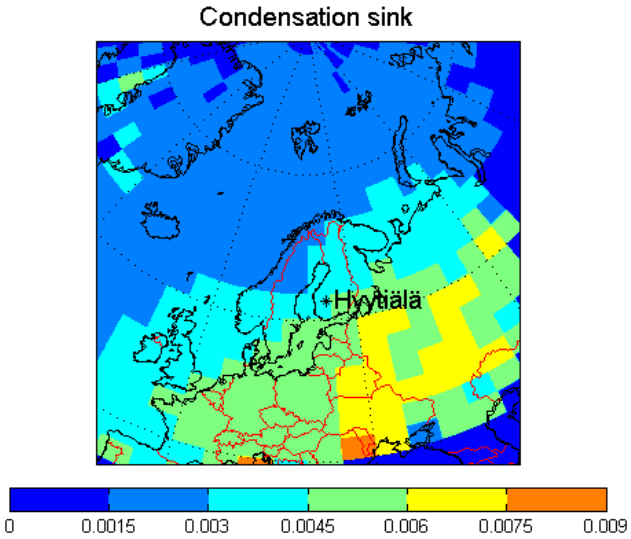

Figure 9. Spatial dependence of the condensation sink observed in Hyytiälä on the air mass history (colour represents the condensation sink, s^{-1}).

8.1.2 Scandinavia

Being a large-scale phenomenon, air masses determine, according to their origin, the air quality and conditions over large areas. However the properties of air masses are changing continuously according to the properties of the underlying surface. Strong surface sources (both natural and anthropogenic) may change its properties considerably.

To confirm or to reject the conclusion about the key role of the air mass originating over the North Atlantic and the Arctic in new particle formation over Scandinavia, Dal Maso *et al.* (2005) carried out an analysis of air mass back trajectories with respect to new particle formation at four Nordic stations, Hyytiälä (61°51'N, 24°17'E), Värriö (67°46'N, 29°35'E), Pallas (67°58'N, 24°07'E), and Aspvreten (58°26'N, 16°24'E), all situated within the boreal forest.

To investigate the area from which the air leading to new particle formation originates, the calculated back trajectories were analyzed with respect to NPF days. As the analyses of each trajectory individually was impractical for the large number of days, we used the azimuth of the air parcel 24 hours before the arrival at the measuring site as a parameter describing the air mass source direction (**Paper II**).

Figure 10 shows the relative distribution of air mass azimuths on event and non-event days for all four stations. From these it is clearly visible that air masses on event days are predominantly arriving from northwestly directions. If we draw an axis from northwest–southeast, almost all event day trajectories arrived from the NW side of this axis. The source of the air masses leading to new particle formation was established to be towards the North Atlantic area for all stations. Southern air transport rarely leads to new particle formation at any of the stations.

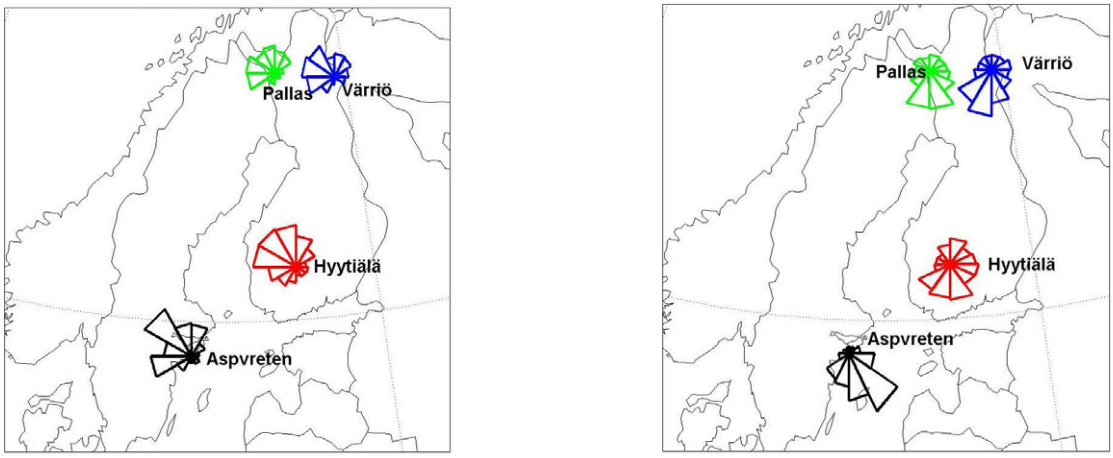

Figure 10. The azimuth of the air masses 24 h prior to arriving at the measurement stations on event (left) and non-event (right) days.

Undefined days showed most variation in air mass transport direction between the stations (**Paper II**). In Pallas and Värriö, the distribution of the transport direction of undefined days was quite uniform, with no direction standing out. In contrast, the distribution at Hyytiälä is dominated by trajectories arriving from directions between west and south.

Air mass back trajectory analysis also allowed to explain the changes in condensation sink observed at the Nordic stations (**Paper II**) with respect to the locations of the main pollution sources. The clear difference in the condensation sink (CS) averages between the north-west sector and the southeast-southwest sector was that the CS in air arriving

from north-west, which on average is much less polluted compared to Central and Eastern Europe, is lower by factor of 2.5–3 at all stations.

Summarizing, using long-term measurements over a geographically large area we were able to establish that new atmospheric particle formation in Scandinavia, presented in our study by four measurement stations, takes place preferably during air mass transport from the north-west direction.

8.1.3 Po Valley, Italy

Having investigated the role of air masses in a clean area of boreal forest in Scandinavia, where biogenic VOC emissions are involved in new particle formation during the growing season (Laaksonen *et al.,* 2008), we aimed to investigate the role of the air masses at the San Pietro Capofiume (44°39'N, 11°37'E) measurement station in Italy.

The station is located in the Po Valley, the largest industrial, trading and agricultural area in Italy with a high population density (for more details see Hamed *et al*., 2005). Hamed *et al.* (2005) found that new particle formation events happen more often at higher SO_2 concentration, compared to non-event days. To investigate the origin and history of air masses during the particle formation process and to identify the main sources of SO_2 transported to the SPC, we performed air mass back trajectories analysis.

The Po Valley axis is prevalently oriented west-east: this maximizes the shading effect of the mountains on the bottom of the valley, enhancing differences in radiation, flow and turbulence over mountain slopes. The atmospheric circulation of the Po Valley is characterized by the strong modification of air flow due to high mountains (Alps and Apennines) that surround the valley on three sides. The dynamic effects of mountains also have a major impact on regional and local airflow patterns that impact the climate of adjacent regions.

Air mass transport from westerly to north-easterly directions dominates in all seasons (**Paper III**). Due to low pressure zone activity, horizontal transport is much stronger in winter compared to summer. The velocity of air parcels decreases towards the arrival point due to the increasing influence of the surface roughness.

Nucleation events at SPC occur more frequently in air masses arriving form Central Europe, whereas event frequency is much lower in the air transported from both southern directions and the Atlantic Ocean (Figure 11a). Trajectories often pass over the polluted (SO_2) Slovenia region and over the Veneto emission sources, east from the station, whereas in Scandinavia aerosol particle formation happens mostly in clean Arctic air masses (**Paper I, Paper II**).

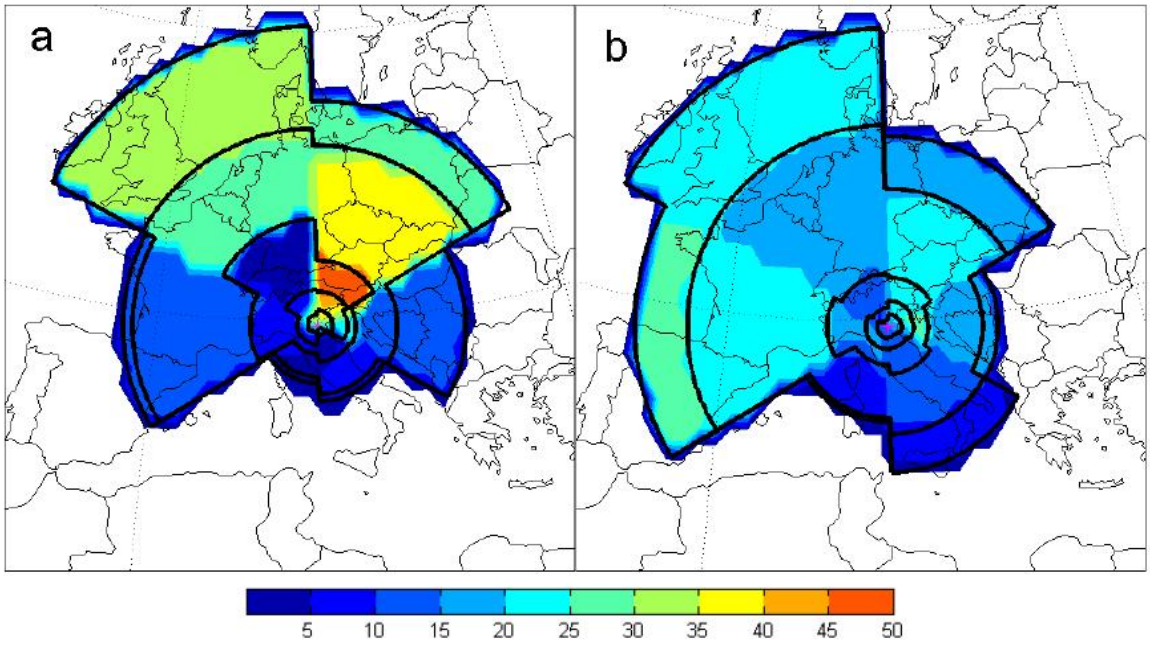

Figure 11. Frequency (%, see color scale) of the location of an air parcel in different direction sectors between the reference back time steps (contour lines for −12, −24, −48, −72, −96 hours back from the arrival time of the air parcel) for **(a)** event and **(b)** non-event for the whole period of measurements.

Meteorological parameters along the back trajectories differ considerably for event and non-event days (**Paper III**). With rare exceptions, mixed layer depth is higher along the event trajectories. However, such parameters as temperature and relative humidity, which along with origin are determined by the elevation of the air parcel, have opposite differences between event and non-event trajectories. We found that, on average, event trajectories undergo stronger subsidence than non-event trajectories during the last 12 h before the arrival at SPC; the amplitude of the increase in the vertical velocity for event class 1 trajectories is highest. This behavior of air parcel trajectories for event days is similar to the one observed at Hyytiälä by Sogacheva *et al.* (2004). Lower precipitation and lower relative and absolute humidity are also typical for event days, as was found for Hyytiälä, Southern Finland (**Paper IV**). However the temperature on NPF event days was higher than on non-event days.

Source regions in the Po Valley seem to be more important in their contribution to the SO_2 concentrations at SPC than the emission sources in Central Europe. The contribution of strong emission sources over Central and Eastern Europe is non-negligible (**Paper III**).

8.2. Meteorological conditions during new particle formation at Hyytiälä

Air masses, along with atmospheric circulation and synoptic processes, determine the meteorological conditions. Meteorological conditions during new particle formation at Hyytiälä have been analyzed in several studies but in most cases for short periods of interest (Mäkelä *et al*, 2000, Nilsson *et al*., 2001a, 2001b, Buzorious *et al*., 2001, 2003, Boy and Kulmala, 2002, Suni *et al*., 2003). In **Paper IV** we analyzed observations at the Hyytiälä background station during 1997-2003 to investigate the possible links between physical, chemical, and meteorological parameters for event and non-event days for different seasons.

In winter, all events were observed at low temperatures ($-5^{o}C>T>-20^{o}C$), high RH (RH>80%) and maximal observed annual concentrations of SO_2 and NO_x. The probability for the nucleation mode in winter to appear is closely related to the substantial increase in O_3 (up to 30%) and UVA radiation, providing an increase in production of OH, which is one of the possible candidates involved in the formation of nonvolatile vapours (Kroll and Seinfeld, 2008). The CS in winter is minimal and the trends of RH and CS are uncorrelated.

In spring, which is the season of the highest frequency of the particle formation process (Dal Maso *et al*, 2005), new particle formation was observed in conditions of maximum temperature at day time, high ozone and UVA levels, and very low relative humidity (RH<40%) and low values of CS ($2\text{-}4\times10^{-3}$ s^{-1}) (**Paper IV**). Diurnal behavior of CS and RH are shown to be well correlated. High correlation in daily patterns of CS and RH means that changing the surface area of the preexisting aerosol (CS) can be connected mainly with changes to RH.

In summer, VOCs start to trigger the formation of the particles. Maximal growth rate of nucleation mode was observed in summer at high temperature, high UVA radiation, maximum concentrations of monoterpenes, hydroxyl and water vapour, and with minimum concentrations of NO_x and SO_2 (**Paper IV**). The CS is maximum in summer. The daily patterns of CS and RH are well correlated. The daily dynamics of O_3 is determined by its production in the boundary layer.

In autumn a second maximum in the frequency of formation events was often observed. For the event days UVA and temperature were higher as compared to non-event days (**Paper IV**) . The ozone concentration increased by 40% during new particle formation. CS had two minimas in event days, one was around 3a.m., another more pronounced minimum was observed around the mean starting time of NPF event (9 a.m. for Hyytiälä, see Dal Maso *et al.,* 2005). It is important to note that the diurnal pattern of CS did not coincide with the daily pattern of RH. Maximum decrease of CS was observed at 5 – 9 a.m. when RH remained constant. It is worth to note that in autumn during event days SO_2 concentration reached a maximum during the morning hours indicating a downward transport from the free troposphere, followed by a decrease during the day.

8.3. Synoptic situation and cloudiness conditions at Hyytiälä during the new particle formation episodes

The role of atmospheric air masses and synoptic weather on NPF in Hyytiälä has been considered in previous studies for periods of field campaigns but not for the long time series of measurements. We investigated the connections between synoptic weather conditions, cloudiness and new particle formation events for a 3-years period. We paid special attention to the analysis of cloudiness aiming to explain the frequency of new particle formation by the frequency of the days with different cloud cover.

The occurrence of Arctic, Polar and Subtropical air masses at Hyytiälä has a clear annual cycle (**Paper V**). Arctic air masses are observed at Hyytiälä more often in winter, reaching a maximum in December, but sustaining high frequency well into the spring. This might be one of the explanations (together with increasing solar radiation) of the well pronounced peak in the frequency of nucleation events observed at Hyytiälä in late spring. The occurrence of Arctic air masses at Hyytiälä decreases strongly toward the summer and reaches its minimum in July.

During about half of the days, Polar air masses prevail at Hyytiälä, with a maximum occurrence in summer, when the Icelandic and Aleutian Lows weaken considerably (Figure 2), the Azores High is getting stronger and shifted slightly to the north, and Arctic air seldom reaches Hyytiälä. Subtropical air has its highest frequency in midsummer. Marine air masses prevail at Hyytiälä throughout the year, except for April (**Paper V**). Continental air is distributed evenly throughout the year. Transition air masses are more frequent in winter and are not observed in June and July.

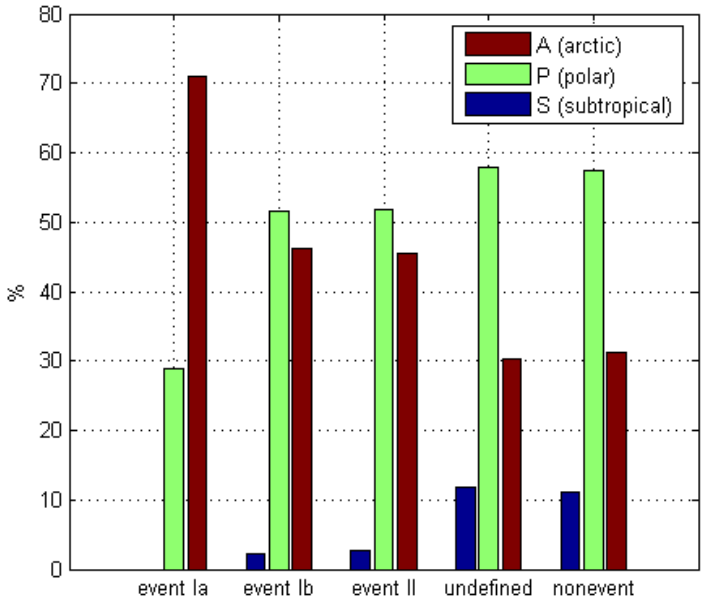

Figure 12. Fraction of the different types of the air masses (Arctic, Polar, Subtropical) during event days (subclasses Ia, Ib, II), undefined and non-event days.

The strongest new particle formation (event class Ia) occurs most often in Arctic air (Figure 12). The fractions of Arctic and Polar air in class Ib and class II events are comparable with a small dominance of Polar air masses; in a few cases, event classes Ib and II have occurred in Subtropical air masses. The distribution of air mass frequency is similar for undefined and non-event days and occurs in the ratio 6:3:1 for Arctic, Polar and Subtropical air, respectively. No significant differences have been revealed in the character of the air mass (marine, continental, transition) with respect to new particle formation except for class Ia, for which the fraction of transition air masses is higher compared to other days.

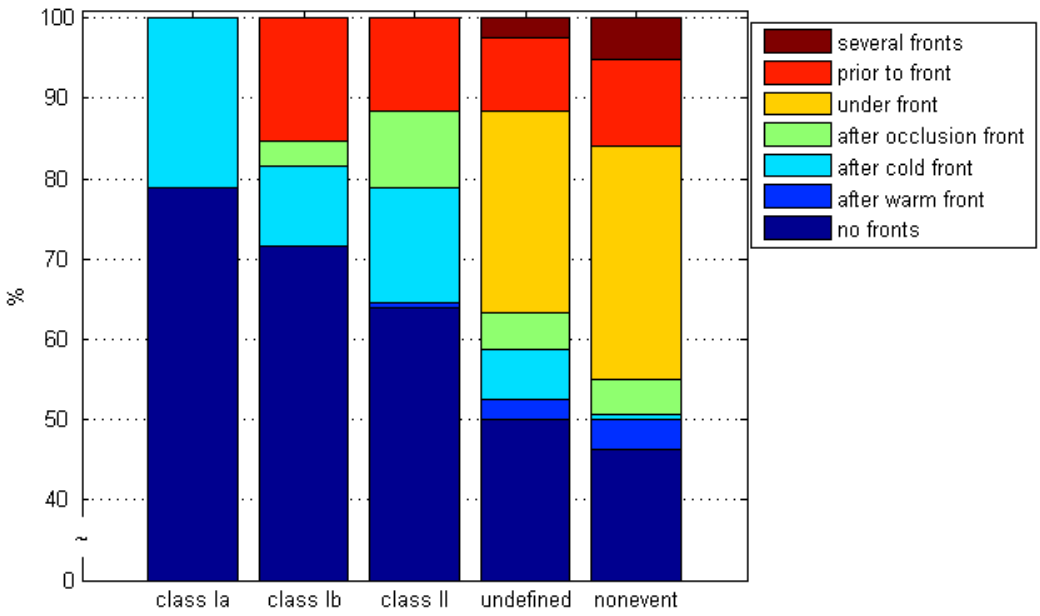

Figure 13. Fraction of the different types of the synoptic situations classified according to atmospheric frontal passages for event days (class Ia, class Ib, class II), undefined and non-event days.

Frontal passages have been observed more frequently in January, gradually decreasing in spring (**Paper V**). The most favourable situation for new particle formation is after cold-frontal passages, and the following days until the next front reaches Hyytiälä (Figure 13). New particle formation event classes Ib and II occur before fronts as well; however, in these cases, the growth of the particles slows down as the frontal cloudiness develops and particle growth is smaller comparing to that during class Ia events. Several frontal passages and slow-moving fronts are observed at Hyytiälä often during undefined and non-event days.

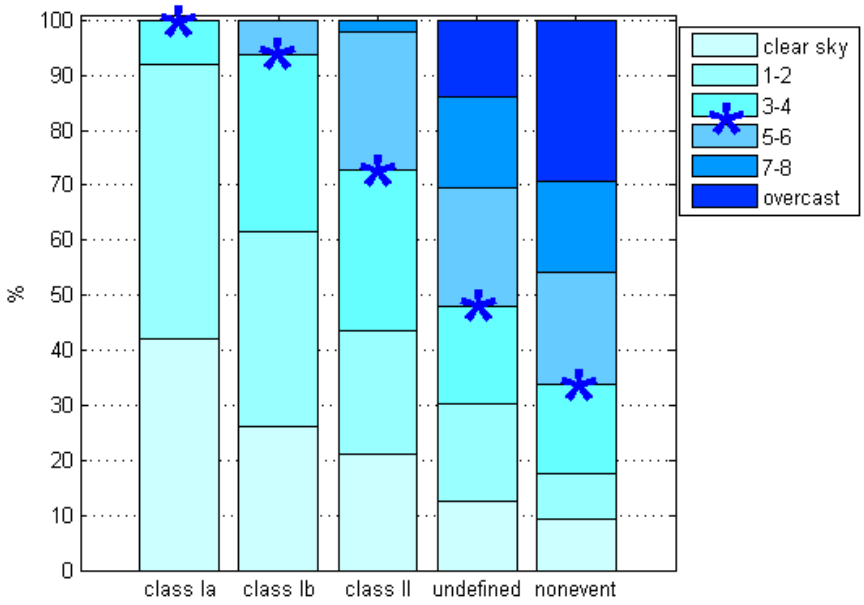

Figure 14. Cloudiness (color, in octas) for event days (class Ia, class Ib, class II), undefined and non-event days. Blue star is a border between 3–4 octas (isolated clouds) and 5–6 octas (broken clouds) cloudiness.

92% of class Ia events are registered at Hyytiälä under clear skies or near clear sky (1–2 octas) conditions (Figure 14); cloudiness of 3-4 octas is observed during the remainder (8%) of class Ia events. The fraction of days with 3–4 octas cloudiness becomes rather significant in class Ib events (32 %); class II events rarely occur at 7–8 octas cloudiness. We consider cloudiness less than 5 octas as one of the preferable conditions for new particle formation; however, at higher cloud amounts, class Ib and class II events may occur. Cloudiness above 4 octas appears to be an important factor for slowing down an aerosol formation event.

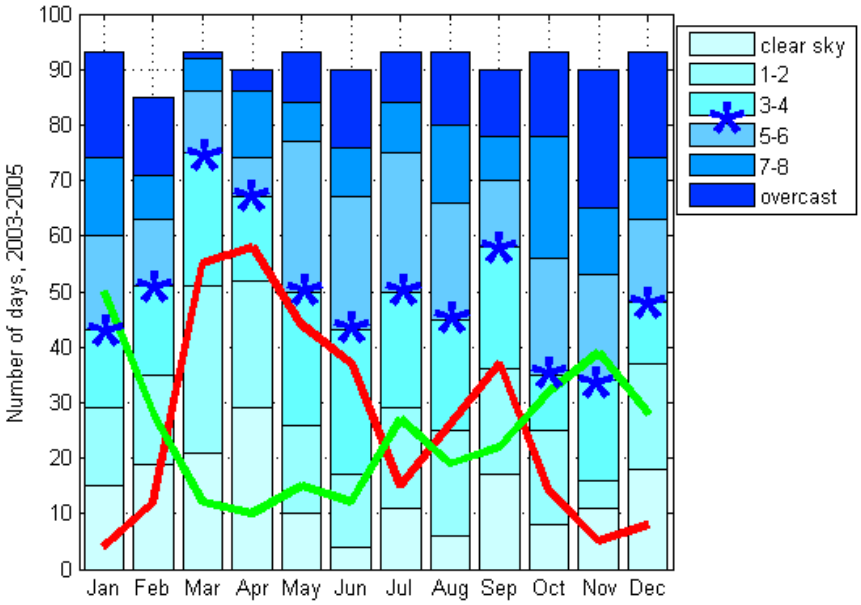

Figure 15 Annual distribution of cloudiness (colorbar, in octas), event (red line) and non-event (green line) days at Hyytiälä. Blue star is a border between 3–4 octas (isolated clouds) and 5–6 octas (broken clouds) cloudiness.

The monthly cumulative number of nucleation events for 2003–2005 corresponds well to the cumulative number of days with cloudiness 3–4 octas and lower for the same period apart from July (Figure 15). As mentioned above, the concentration of VOC in summer plays a significant role in NPF. March–April and September maxima in new particle formation coincide with the high number of days with low cloud fraction in the corresponding months.

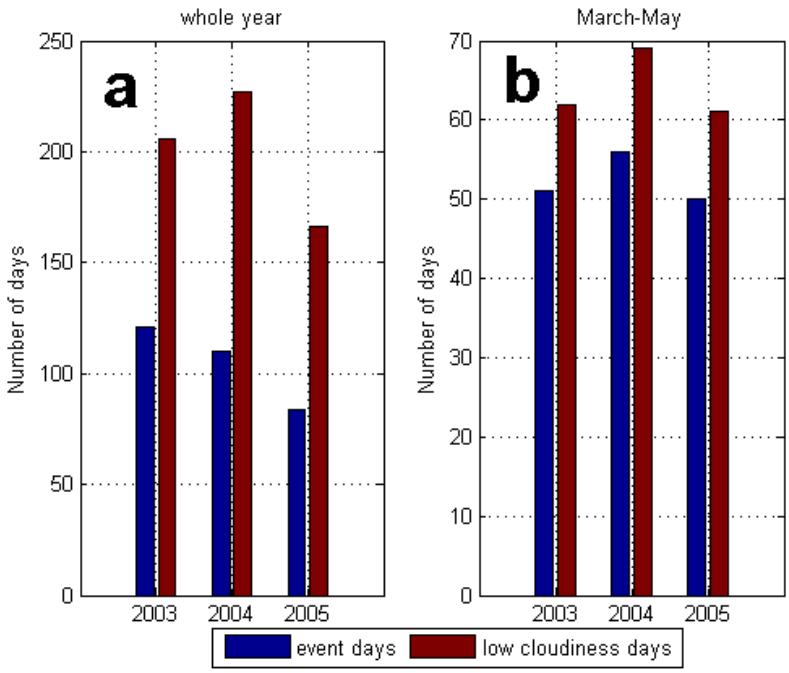

Figure 16. Yearly number of event days and number of low-cloud-amount days for years 2003, 2004 and 2005 (a), and the same for months March–May (b).

Year-to-year differences in the frequency of new particle formation have a trend similar to that for the number of low-cloud-amount days. The smaller number of nucleation events in year 2005 with respect to year 2003 can be explained by the lower occurrence of the clear sky and low-cloud-amount days (Figure 16a). The high number of low-cloud-amount days in the winter, when particle formation occurrence is low, for the year 2004 explains the discrepancy in our assumption on the yearly correspondence of the number of nucleation days to the number of low cloudiness days. During the period of the most frequent new particle formation (March–May), the peak in the number of nucleation days in year 2004 corresponds well to the peak in the number of low-cloud-amount days (Figure 16b).

9 Conclusions

In this thesis we describe the analyses of air masses and meteorological parameters at four Nordic stations (Hyytiälä, Värriö, Pallas and Aspvreten) located in boreal forest, and at San Pietro Capofiume station located in the Po Valley, Italy, as well as synoptic situations and cloudiness at Hyytiälä, Southern Finland, to identify conditions which are favourable for new particle formation in atmosphere.

Using back trajectory analysis, we show that the prevailing air mass transport directions for Hyytiälä, Southern Finland, are from south-west to north-west, whereas north and north-east transport is less frequent. Seasonal differences in prevailing flow directions have been identified, but they are minor. In winter, long-range transport of air mass to Hyytiälä takes place more often than in summer.

The dominant role of Arctic and Polar air masses facilitating the formation of new particle is clearly seen during all seasons and on multi-year average at Hyytiälä, as well as at other Nordic stations. Air masses transported from Central Europe and Russia rarely lead to new particle formation at any of the stations. The highest number concentrations of accumulation mode particles and highest condensation sink values measured in Hyytiälä are observed in continental air masses arriving at Hyytiälä from Eastern Europe and Central Russia.

Contrary to Scandinavia, presented in our studies by four Nordic stations, nucleation events in Po Valley, at San Pietro Capofiume, occur more frequently in air masses arriving form Central Europe, than in air transported from either the Atlantic Ocean or southern directions. Meteorological parameters along back trajectories arriving at SPC differ considerably for event and non-event days. With rare exceptions, mixed layer depth is higher along the event trajectories.

The most favourable situation for new particle formation during all seasons is cold air advection after cold-frontal passages, and the days following it until the next front reaches Hyytiälä. We demonstrated that in Hyytiälä cloudiness less than 5 octas is one of the favourable conditions for class Ia event of new particle formation; however, at higher cloud amounts, NPF may occur as well. Cloudiness above 4 octas appears to be an important factor for turning an aerosol formation event into class Ib and class II, and preventing Ia class events. The frequency of new particle formation is related to the frequency of low-cloud-amount days in Hyytiälä.

References

Aalto, P., Hämeri, K., Becker, E., Weber, R., Salm, J., Mäkelä, J. M., Hoell, C., O'Dowd, C. D., Karlsson, H., Hansson, H.- C., Väkevä, M., Koponen, I. K., Buzorius, G., and Kulmala, M. 2001. Physical characterization of aerosol particles during nucleation events. *Tellus*, **53B**, 344–358.

Abdalmogith, S. S., and Harrrison, R.M. 2005. The use of trajectory cluster analysis to examine the long-range transport of secondary inorganic aerosol in the UK. *Atmos. Env.*, **39**, 6686–6695.

Aitken, J. A. 1891. On dust, fog and clouds. *Nature* **23**, 195–197.

Aitken, J. A. 1892. On the number of dust particles in the atmosphere of certain places in Great Britain and on the Continent, with remarks on the relation between the amount of dust and meteorological phenomena. *Trans. Roy.Soc.,* **37**, 416-417.

Aitken, J. A. 1897. On some nuclei of cloudy condensation. *Trans. Roy.Soc.* **39**, 15–25.

Albrecht, B.A. 1989. Aerosols, cloud microphysics, and fractional cloudiness. *Science*, **245**, 1227-1230.

Andreae, M.O., Elbert, W., Gabriel, R., Johnson, D.W., Osborne, S., and Wood, R. 2000. Soluble ion chemistry of the atmospheric aerosol and SO2 concentration over the eastern North Atlantic during ACE-2. *Tellus*, **52B**, 1066-1087.

Andreae, M.O., and Rosenfeld, D. 2008. Aerosol-cloud-precipitation interactions. Part 1. The nature and sources of cloud-active aerosols. *Earth-Science Rev.,* **89**, 13-41.

Anttila, T., Kerminen, V.-M., Kulmala, M., Laaksonen, A., and O'Dowd, C.D. 2004. Modelling the formation of organic particles in the atmosphere. *Atmos. Chem. Phys.*, **4**, 1071-1083.

Anttila, T., and Kerminen, V.-M. 2007. On the contribution of Aitken mode particles to cloud droplet populations at continental background areas – a parametric sensitivity study. *Atmos. Chem. Phys.*, **7**, 4625-4637.

Battan, L.J. 1978. Fundaments of meteorology. Prentice-Hall, 322p.

Barry, R.G., and Chorley, R.J. 2003. Atmosphere, Weather and Climate. 8th edition. Routledge, 11 New Fetter Lane, London. 422p.

Baumgardner, D., G. Kok, and G. Raga. 2004. Warming of the Arctic lower stratosphere by light absorbing particles. *Geophys. Res. Lett.*, **31**, L06117, doi:10.1029/2003GL018883.

Bigg, E. K. 1997. A mechanism for the formation of new particles in the atmosphere. *Atmos. Res.*, **43**, 129–137.

Birmili, W., Wiedensohler, A., Heintzenberg, J., and Lehmann, K. 2001. Atmospheric particle number size distribution in central Europe: Statistical relations to air masses and meteorology. *J. Geophys. Res.*, **106**, 32005–32018.

Bissolli, P., and Dittmann, E. 2001. The objective weather type classification of the German Weather Service and its possibilities of application to environmental and meteorological investigations. *Met. Zeitschrift*, **10**, 4, 253-260.

Bonasoni, P., Cristofanelli, P., Calzolari, F., Bonafe, U., Evangilisti, E., Stohl, A., Sajani, S. Z., van Dingenen, R., Colombo, T., and Balkansky, Y. 2004. Aerosol-ozone correlations during dust transport episodes. *Atm. Chem. Phys.*, **4**, 1201-1215.

Bonn, B., Hirsikko, A., Hakola, H., Kurtén, T., Laakso, L., Boy, M., Dal Maso, M.,

Mäkelä, J. M., and Kulmala, M. 2007 Ambient sesquiterpene concentration and its link to air ion measurements. *Atmos. Chem. Phys.,* **7**, 2893-2916.

Bonn, B., M. Kulmala, I. Riipinen, S.-L. Sihto, and T. M. Ruuskanen. (2008). How biogenic terpenes govern the correlation between sulfuric acid concentrations and new particle formation. *J. Geophys. Res*., **113**, D12209, doi:10.1029/2007JD009327.

Boy M., , Bonn, B., Kazil, J., Lovejoy, N., Turnipseed, A., Jim Greenberg, J., Thomas Karl, T., Mauldin, L., Kusciuch, E., Smith, J., Kelly Barsanti, K., Guenther, A., Wehner, B., Hellmuth, O., Siebert, H., Bauer, S., Wiedensohler, A., and Kulmala, M. 2007. Relevance of several nucleation theories in different environments. Nucleation and Atmospheic aerosols. Eds. O'Dowd, C.D., Wagner, P.E. Springer, 87-91.

Boy, M., Hellmuth, O., Korhonen, H., Nilsson, D., ReVelle, D., Turmipseed, A., Arnold, F., and Kulmala, M. 2006. MALTE – model to predict new aerosol formation in the lower troposphere. *Atmos. Chem. Phys*., **6**, 4499-4517.

Boy, M., Kazil, J., Lovejoy, E.R., Guenther, A., and Kulmala, M. 2008. Relevance of ion-induced nucleation of sulphuric acid and water in the lower troposphere over the boreal forest at northern laliludes. *Atmos. Research*, in press.

Boy M. and Kulmala M. 2002. Nucleation events on the continental boundary layer: influence of physical and meteorological parameters. *Atmos. Chem. Phys.*, **2**, 1-16.

Boy, M., Kulmala, M., Ruuskanen, T. M., Pihlatie, M., Reissell, A., Aalto, P. P., Keronen, P., Dal Maso, M., Hellen, H., Hakola, H., Jansson, R., Hanke, M., and Arnold, F. 2005. Sulphuric acid closure and contribution to nucleation mode particle growth. *Atmos. Chem. Phys*., **5**, 863–878.

Bonn B. and Moortgat G.K. 2002. New particle formation during α-and β-pinene oxidation by O_3, OH, and NO_3, and the influence of water vapour: particle size distribution studies. *Atmos. Chem. Phys.*, **2,** 183-196.

Boy, M., Petäjä, T., Dal Maso, M., Rannik, Ü., Rinne, J., Aalto, P., Laaksonen, A., Vaattovaara, P., Joutsensaari, J., Hoffmann, T., Warnke, J., Apostolaki, M., Stephanou, E. G., Tsapakis, M., Kouvarakis, A., Pio, C., Carvalho, A., R¨ompp, A., Moortgat, G., Spirig, C., Guenther, A., Greenberg, J., Ciccioli, P., and Kulmala, M. 2004. Overview of the field measurement campaign in Hyytiälä, August 2001 in the framework of the EU project OSOA. *Atmos. Chem. Phys*., **4**, 657–678.

Brankov, E., Rao, S.T., and Porter, P.S. 1998. A trajectory-clustering-correlation methodology for examining the long-range transport of air pollutants. *Atmos. Env*., **32**, 1525–1534.

Brimblecombe, P. 1996. Air Composition and Chemistry. Cambridge University Press, Cambridge, 253p.

Buchanan, C.M., Beverland, I.J., and Heal, M.R. 2002. The influence of weather-type and long-range transport on air particle concentrations in Edinburgh, UK. *Atmos. Env*., **36**, 5343–5354.

Burtscher, H. 2005. Physical characterization of particulate emissions from diesel engines: a review. *J. Aerosol Sci*, **36**, 896–932.

Buzorius, G., Rannik, Ü., Aalto, P., Dal Maso, M., Nilsson, E. D., Lehtinen, K. E. J., and Kulmala, M. 2003. On particle formation prediction in continental boreal forest

using micrometeorological parameters, *J. Geophys. Res*., **108** (D13), 4377, doi:10.1029/2002JD002850.

Buzorius, G., Rannik, Ü., Nilsson, D., and Kulmala, M. 2001. Vertical fluxes and micrometeorology during aerosol particle formation events. *Tellus*, **53B**, 394–405.

Cape, J.N., Methven, J., and Hudson, L.E., 2000. The use of trajectory cluster analysis to interpret trace gas measurements at Mace Head, Ireland. *Atmos. Env*., **34**, 3651–3663.

Charlson, R.J., Langner, J., Rodhe, H., Leovy, C.B., and Warren, S.G. 1991. Perturbation of the northern hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols. *Tellus,* **43AB** , 152–163.

Charlson, R. J., and J. Heintzenberg, eds.1995. *Aerosol Forcing of Climate*, Wiley, New York, 416 pp.

Clarke, A. D., Eisele, F., Kapustin, V. N., Moore, K., Tanner, D., Mauldin, L., Litchy, M., Lienert, B., Carroll, M. A., and Albercook, G. 1999. Nucleation in the equatorial free troposphere: Favorable environments during PEM-Tropics. *J. Geophys. Res*., **104** (D5), 5735–5744.

Coe, H.,Williams, P. I., McFiggans, G., Gallagher, M.W., Beswick, K. M., Bower, K. N., and Choularton, T. W. 2000. Behavior of ultrafine particles in continental and marine air masses at a rural site in the United Kingdom, *J. Geophys. Res*., **105** (D22), 26 891– 26 905.

Dal Maso, M. , Kulmala, M. , Mäkelä, J. M. , Aalto, P. and O'Dowd, C. D. 2002. Condensation and coagulation sinks and the formation of nucleation mode particles in coastal and boreal forest boundary layers. *J. Geophys. Res.*, 107, 8097, doi:10.1029/2001JD001053.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P.P. and Lehtinen, K.E.J. 2005. Formation and growth of fresh atmospheric aerosols: Eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland. *Boreal Environ. Res.*, **10**, 323-336.

Dal Maso, M., Sogacheva, L., Aalto, P.P., Riipinen, I., Komppula, M., Tunved, P., Korhonen, L., Suur-Uski, V., Hirsikko, A., Kurtén, T., Kerminen, V.-M., Lihavainen, H., Viisanen, Y., Hansson, H.-C. and Kulmala, M. 2007. Aerosol size distribution measurements at four Nordic field stations: identification, analysis and trajectory analysis of the new particle formation bursts. *Tellus B*, **59**(3), 350-361.

de Reus, M., Ström, J., Hoor, P., Lelieveld, J., and Schiller, C. 1999. Particle production in the lowermost stratosphere by convective lifting of the tropopause, *J. Geophys. Res*., **104** (D19), 23 935–23 940.

Draxler R.R and Hess G.D. 1998. An Overview of the HYSPLIT_4 Modeling System for Trajectories, Dispersion and Deposition. *Australian Meteorological Magazine*, **47**, 295-308.

Draxler R.R and Hess G.D. 2004. Description of the HYSPLIT_4 Modeling System. *NOAA Technical Memorandum ERL ARL-224*.

Engler, C., Rose, D., Wehner, B., Wiedensohler, A., Brüggemann, E., Gnauk, T., Spindler, G., Tuch, T., and Birmili, W. 2007. Size distribution of non-volatile particle residuals (Dp<800nm) at a rural site in Germany and relation to air mass origin. *Atmos. Chem. Phys*., **7**, 5785-5802.

Finardi, S., and Pellegrini, U. 2004. Systematic analysis of meteorological conditions causing severe urban air pollution episodes in the central Po Valley, 9th Int. Conf. on Harmonization within Atmospheric Dispersion Modeling for Regulatory Purposes, 250-254.

Flanagan, R,. Geever, M., and O'Dowd, C.D. 2005. Direct measurements of new-particle fluxes in the coastal environment. *Environ. Chem.,* **2**, 256-259.

Flossman, A.I. 2002. The effect of the impaction scavenging efficiency on the wet deposition by a convective warm cloud. Tellus 45B, 34-39.

Flossman, A.I., Hall, W.D., and Pruppacher, H.R. 1985. A theoretical study of the wet removal of atmospheric pollutants. Part 1: The redistribution of aerosol particles captured through nucleation and impaction scavenging by growing cloud drops. J. Atmos Sci., **42**, 582-606.

Foret, G., G. Bergametti, F. Dulac, and L. Menut. 2006. An optimized particle size bin scheme for modeling mineral dust aerosol. *J. Geophys. Res.*, **111**, D17310, doi:10.1029/2005JD006797.

Furiness, C., Smoth, L., Ran, L., and Cowling, E. 1998. Comparison of emissions of nitrogen and sulfur oxides to deposition of nitrate and sulfate in the USA by state in 1990. International Nitrogen Conference, **102**, 313-320.

Gaydos, T. M., C. O. Stanier, and S. N. Pandis. 2005. Modeling of in situ ultrafine atmospheric particle formation in the eastern United States, *J. Geophys. Res*., **110**, D07S12, doi:10.1029/2004JD004683.

Hakola, H., Tarvainen, V., Laurilla, T., Hiltunen, V., Helen, H., and Keronen, P. 2003. Seasonal variation of VOC concentration above a boreal coniferous forest. *Atmos. Environ.* **37**, 1623 – 1634.

Hamed, A., Joutsensaari, J., Mikkonen, S., Sogacheva, L., Dal Maso, M., Kulmala, M., Cavalli, F., Fuzzi, S., Facchini, M. C., Decesari, S., Mircea, M., Lehtinen, K. E. J., and Laaksonen, A. 2007. Nucleation and growth of new particles in Po Valley, Italy. *Atmos. Chem. Phys*., **7**, 355–376.

Hari, P., and Kulmala, M. 2005. Station for Measuring Ecosystem-Atmosphere Relations (SMEAR II). *Boreal Env. Res*. **10**, 315–322.

Haywood, J.M., and Shine, K.P. 1995. The effect of anthropogenic sulfate and soot aerosol on the clear sky planetary radiation budget, *Geophys. Res. Let.,* **22**, 603–606.

Hermann, M., Heintzenberg, J., Wiedensohler, A., Zahn, A., Heinrich, G., and Brenninkmeijer, C. A. M. 2003. Meridional distributions of aerosol particle number concentrations in the upper and lower stratosphere obtained by civil aircraft for regular investigation of the atmosphere based on an instrument container (CARIBIC) flights. *J. Geophys. Res*., **108** (D3), 4114, doi:10.1029/2001JD001077.

Hinds, W. 1999. Aerosol Technology. 2nd edition. John Wiley & Sons, Inc.

Hussein, T., Puustinen, A., Aalto, P. P., Mäkelä, J. M., Hämeri, K., and Kulmala, M. 2004. Urban aerosol number size distributions, *Atmos. Chem. Phys.,* **4**, 391-411.

Jaenicke, R. 2008. Is atmospheric aerosol an aerosol? – A look at the sources and variability. *Faraday Discuss.,* **137**, 235-243.

Kahl, J.D.W. 1996. On the prediction of trajectory model error. *Atmos. Environ*., **30**, 2945–2957.

Kemp, K., 1993. A multi-point receptor model for long-range transport over southern Scandinavia. *Atmos. Environ.*, **27A**, 823–830.

Khosrawi, F. and Konopka, P. 2003. Enhanced particle formation and growth due to mixing processes in the tropopause region. *Atmos. Environ.*, **37**, 903–910.

Komppula, M. 2005. New particle formation and its connection with cloud droplet population in a remote continental site in Northern Finland. *Report series in aerosol science*, **75**.

Korhonen, P., Kulmala, M., Laaksonen, A., Viisanen, Y., McGraw, R., and Seinfeld, J. H. 1999. Ternary nucleation of H2SO4, NH3, and H2O in the atmosphere. *J. Geophys. Res.*, **104** (D21), 26 349–26 353.

Korhonen, H., Lehtinen, K.E.J., and Kulmala, M. 2004. Multicomponent aerosol dynamics model UHMA: model development and validation. *Atmos. Chem. Phys.* **4**, 757-771.

Kroll, J.H., and Seinfeld, J.H. 2008. Chemistry of secondary organic aerosol: Formation and evolution of low-volatility organics in the atmosphere. *Atm. Env.*, **42**, 3593–3624.

Kulmala, M., and Kerminen, V.-M. 2008. On the formation and growth of atmospheric nanoparticles. *Atmos. Res.*, doi:10.1016/j.atmosres.2008.01.005

Kulmala, M., Kerminen, V.-M., Anttila, T., Laaksonen, A., and O'Dowd, C. D. 2004a. Organic aerosol formation via sulphate cluster activation. *J. Geophys. Res.*, **109**, D04205, doi:10.1029/2003JD003961.

Kulmala, M., Laakso, L., Lehtinen, K. E. J., Riipinen, I., Dal Maso, M., Anttila, T., Kerminen, V.-M., Hörrak, U., Vana, M., and Tammet, H. 2004b. Initial steps of aerosol growth. *Atmos. Chem. Phys.*, **4**, 2553–2560.

Kulmala, M., and Laaksonen, A. 1990. Binary nucleation of water-sulfuric acid system: Comparison of classical theories with different H_2SO_4 saturation vapor pressures. *J. Chem. and Phys.*, **93**, 696–701.

Kulmala, M., Lehtinen, K. E. J., and Laaksonen, A. 2006. Cluster activation theory as an explanation of the linear dependence between formation rate of 3 nm particles and sulphuric acid concentration. *Atmos. Chem. Phys.*, **6**, 787–793.

Kulmala, M., Pirjola, L., and Mäkela, J. M. 2000. Stable sulphate clusters as a source of new atmospheric particles. *Nature*, **404**, 66–69.

Kulmala M., Petajä T., Monkkönen P., Kopponen I., Dal Maso M., Aalto P., Lehtinen K.E.J. and Kerminen V.-M. 2005. On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments. *Atmos. Chem. Phys.*, **5**, 409-416.

Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W., and McMurry, P. H. 2004c. Formation and growth rates of ultrafine atmospheric particles: A review of observations. *J. Aerosol Sci.*, **35**, 143–176.

Kurtén, T., Bonn, B., Vehkamäki, H., and Kulmala, M. 2007. Computational Studies of the Reactions between Biogenic Stabilized Criegee Intermediates and Sulphyric Acid. *J. Phys. Chem. A* **, 111***,* 3394-3401.

Laaksonen, A., Kulmala, M., O'Dowd, C. D., Joutsensaari, J., Vaattovaara, P., Mikkonen, S., Lehtinen, K. E. J., Sogacheva, L., Dal Maso, M., Aalto, P., Petäjä, T., Sogachev, A., Yoon, Y. J., Lihavainen, H., Nilsson, D., Facchini, M. C., Cavalli, F., Fuzzi, S., Hoffmann, T., Arnold, F., Hanke, M., Sellegri, K., Umann, B., Junkermann, W., Coe, H., Allan, J. D., Alfarra, M. R., Worsnop, D. R.,

Riekkola, M. -L., Hyötyläinen, T., and Viisanen, Y. 2008. The role of VOC oxidation products in continental new particle formation. *Atmos. Chem. Phys.*, **8**, 2657-2665.

Lazaridis, M., Kulmala, M., and Gorbunov, B. Z. 1992. Binary heterogneous nulceation at a nonuniform surface, *J. Aerosol Sci.,* **23**, 457–466.

Lickens, G.E., Driscoll, C.T., and Buso, D.C. 1996. Long-term effects of acid rain: response and recovery of forest ecosystem. *Science*, **272**, 244-246.

Lohman, U., and Feichter, J. 2005. Gobal indirect aerosol effect: a review. *Atmos. Chem. Phys.*, **5**, 715-737.

Loosmore, G.A., and Cederwall, R.T. 2004. Precipitation scavenging of atmospheric aerosols for emergency response application: testing and updating model with new real-time data. *Atmos. Environ.*, **38**, 993-1003.

Lyubovtseva Yu., Sogacheva L., Dal Maso M., Bonn B., Keronen P. and Kulmala M. 2005. Seasonal variations of trace gases, meteorological parameters, and formation of aerosols in boreal forests. *Boreal Env. Res.*, **10**, 493-510.

Lutgens, F. K., and Tarbuck, E. J. The Atmosphere: an Introduction to Meteorology. 2001. Prentice Hall, 8th edition.

McIlveen, J.E.R. 1992. Fundamentsof weather and climate. Chapman & Hall, 498p.

McMurry, P.H., and S.K. Friedlander. 1979. New particle formation in the presence of an aerosol, *Atmos. Environ.*, **13**, 1635-1651.

Merikanto, J., Napari, I., Vehkamäki, H., Anttila, T., and Kulmala, M. 2007. New parameterization of sulfuric acid-ammonia-water ternary nucleation rates at tropospheric conditions. *J. Geophys. Res.*, **112**, D15207, doi:10.1029/2006JD007977.

Moody, J.L. and Galloway, J.N. 1988. Quantifying the relationship between atmospheric transport and the chemical composition of precipitation on Bermuda. *Tellus*, **40B**, 463–479.

Mukai, H. and Suzuki, M. 1996. Using air trajectories to analyze the seasonal variation of aerosols transported to the Oki islands. *Atmos. Env.*, **30**, 3917–3934.

Musk, L.F. 1988. Weather systems. Cambrige University press, Cambrige, 160pp.

Mäkelä J.M., Dal Maso M., Pirjola L., Keronen P., Laakso L., Kulmala M. and Laaksonen, A. 2000. Characteristics of the atmospheric particle formation events observed at a boreal forest site in Southern Finland. *Boreal Env. Res.*, **5**, 299-313.

Napari, I., Noppel, M., Vehkamäki, H., and Kulmala, M. 2002. Parameterization of ternary nucleation rates for H_2SO_4-NH_3-H_2O vapors. *J. Geophys. Res.*, **107**, D19, 4381, doi:10.1029/2002JD002132.

Nilsson, E. D., and Kulmala, M. 1998. The potential for atmospheric mixing processes to enhance the binary nucleation rate, *J. Geophys. Res.*, **103** (D1), 1381–1389.

Nilsson E.D., Paatero J., and Boy, M. 2001a. Effect of air masses and synoptic weather on aerosol formation in the continental boundary layer. *Tellus*, **53B**, 462-478.

Nilsson, E.D., and Rannik, Ü. 2001. Turbulent Aerosol Fluxes over the Arctic Ocean, part 1, dry deposition over sea and pack ice. *J. Geophys. Res.*, **106**, 32125-32138.

Nilsson, E. D., Rannik, Ü., Buzorius, G., Kulmala, M. and O'Dowd, C. 2001b. Effects of the continental boundary layer evolution, convection, turbulence and entrainment on aerosol formation. *Tellus*, **53B**, 441-461.

Niyogi, D., Chang, H.-I., Saxena, V. K., Holt, T., Alapaty, K., Booker, F., Chen, F., Davis, K.J., Holben, B., Matsui, T., Meyers, T., Oechel, W.C., Pielke, R. A., Wells, R., Wilson, K., and Xue, Y. 2004. Direct observations of the effects of aerosol loading on net ecosystem CO_2 exchanges over different landscapes. *Geophys. Res. Lett.*, **31**, L20506, doi:10.1029/2004GL020915.

Nyeki, S., Kalberer, M., Lugauer, M., Weingartner, E., Petzold, A., Schröder, F., Colbeck, I., and Baltensperger, U. 1999. Condensation Nuclei (CN) and ultrafine CN in the free troposphere to 12 km: A case study over the Jungfraujoch high-alpine research station. *Geophys. Res. Let.*, **14**, 2195–2198.

O'Dowd, C. D., Aalto, P., Hämeri, K., Kulmala, M., and Hoffmann, T. 2002a. Atmospheric particles from organic vapours. *Nature*, **416**, 497–498.

O'Dowd, C. D., Hämeri, K., Mäkelä, J. M., Pirjola, L., Kulmala, M., Jennings, S. G., Berresheim, H., Hansson, H.-C., de Leeuw, G., Kunz, G. J., Allen, A. G., Hewitt, C. N., Jackson, A., Viisanen, Y., and Hoffmann, T. 2002b. A dedicated study of new particle formation and fate in the coastal environment (PARFORCE): Overview of objectives and achievements, *J. Geophys. Res.*, **107** (D19), 8108, doi:10.1029/2001JD000555.

Patterson, E. M., Kiang, C. S., Delany, A. C., Wartburg, A. F., Leslie, A. C. D., and Huebert, B. J.. 1980. Global Measurements of Aerosols in Remote Continental and Marine Regions: Concentrations, Size Distributions, and Optical Properties, *J. Geophys. Res.*, **85**, 7361–7376.

Pirjola, L., O'Dowd, C. D., Brooks, I. M., and Kulmala, M. 2000. Can new particle formation occur in the clean marine boundary layer? *J. Geophys. Res.*, **105** (D21), 26531–26546.

Pugatchova, A., Kikas, Ü., Prussel, M., Reinart, A., Tamm, E., and Ulevicius, V. 2007. Relation of Aerosol Microphysical Properties and Chemical Composition of Aerosol in the Baltic Sea Region. *Nucleation and Atmospheric Aerosols*, C.D. O'Dowd and P.E. Wagner (eds.), Springer, 711–715.

Ramanathan, V., Crutzen, P.J., Kiehel., J.T., and Rosenfeld, D. 2001. Aerosol, climate and hydrological cycle. *Science*, **294**, 2119-2124.

Rannik, Ü. 1998. On the surface layer similarity at a complex forest site. *J. Geophys. Res.*, **103**, 8685–8697.

Riipinen, I., Sihto, S.-L., Kulmala, M., Arnold, F., Dal Maso, M., Birmili, W., Saarnio, K., Teinilä, K., Kerminen, V.-M., Laaksonen, A. and Lehtinen, K.E.J., 2007. Connections between atmospheric sulphuric acid and new particle formation during QUEST III - IV campaigns in Hyytiälä and Heidelberg. *Atmos. Chem. Phys.*, **7**, 1899-1914, SRef-ID: 1680-7324/acp/2007-7-1899.

Ro, C.U., Tang, A.J.S., Chan, W.H., Kirk, R.W., Reid, N.W., and Lusis, M.A. 1988. Wet and dry deposition of sulphur and nitrogen compound in Ontario. *Atmos. Environ.*, **22**, 2763-2772.

Russell, L. M., Lenschow, D. H., Laursen, K. K., Krummel, P. B., Siems, S. T., Bandy, A. R., Thornton, D. C., and Bates, T. S. 1998. Bidirectional mixing in an ACE 1 marine boundary layer overlain by a second turbulent layer, *J. Geophys. Res.*, **103** (D13), 16 411–16 432.

Ryzhkov, A. B. and Ariya, P. A. 2006. The importance of water clusters (H2O)n (n=2..4) in the reaction of Criegee intermediate with water in the atmosphere. *Chem. Phys. Lett*. **419**, 479-485.

Schröder, F. and Ström, J. 1997. Aircraft measurements of sub micrometer aerosol particles (> 7 nm) in the midlatitude free troposphere and tropopause region. *Atmos. Res*., **44**, 333–356.

Scheele, M.P., and Siegmund, P.C. 2001. Estimating errors in trajectory forecasts using ensemble predition. *J. Appl. Meteorol*., **40**, 1223-1232.

Seinfeld, J.H., and Pandis, S.N. 1998. Atmospheric chemistry and physics. John Wiley & Sons, Inc. 1326pp.

Shilling, J. E., Chen, Q., King, S. M., Rosenoern, T., Kroll, J. H., Worsnop, D. R., McKinney, K. A., and Martin, S. T. 2008. Particle mass yield in secondary organic aerosol formed by the dark ozonolysis of λ-pinene. *Atmos. Chem, Phys*., **8**, 2073-2088. SRef-ID: 1680-7324/acp/2008-8-2073

Siebert, H., Stratmann, F., and Wehner, B. 2004. First observations of increased ultrafine particle number concentrations near the inversion of a continental planetary boundary layer and its relation to ground-based measurements, *Geophys. Res. Lett*., **31**, L09102,doi:10.1029/2003GL019086.

Siebert P., Kromp-Kolb H., Kasper A., Kalina M., Puxbaum H., Jost D.T., Schwikowski M., and Baltensperger, U. 1998. Transport of polluted boundary layer from the Po valley to High-Alpine sites. *Atmos. Environ.* **32**, 3953-3965.

Sogacheva, L., Rannik, Ü., and Kulmala, M. 2004. Preliminary analysis of air mass properties accompanying the nucleation events in Southern Finland. *Report Series in Aerosol Science,* **68**, 291-296.

Sogacheva, L., Dal Maso, M., Kerminen, V.-M., and Kulmala, M. 2005. Probability of nucleation events and aerosol particle concentration in different air mass types arriving at Hyytiälä, southern Finland, based on back trajectory analysis. *Boreal Env. Res*., **10**, 479-491.

Spracklen, D., Pringle, K., Carslaw, K., Chipperfield, M., and Mann, G.W. 2005a. A global off-line model of size-resolved aerosol processes. I. Model development and prediction of aerosol properties. *Atmos. Chem. Phys*., **5**, 2227-2252.

Spracklen, D., Pringle, K., Carslaw, K., Chipperfield, M., and Mann, G.W. 2005b. A global off-line model of size-resolved aerosol processes. II. Identification of key uncertainties. *Atmos. Chem. Phys*., **5**, 3233-3250.

Spracklen, D. V., Carslaw, K.S., Kulmala, M., Kerminen, V.-M., Sihto, S.-L., Riipinen, I., Mericanto, J., Mann, G.W., Chipperfield, M.P., Wiedensholer, A., Birmili, W., and Lihavainen, H. 2008. Contribution of particle formation to global cloud condensation nuclei concentrations, *Geophys. Res. Lett.*, **35**, L06808, doi:10.1029/2007GL033038.

Stohl, A., 1998. Computation, accuracy and applications of trajectories-a review and bibliography. *Atmos. Environ*., **32**, 947–966.

Stratmann, F., Siebert, H., Spindler, G., Wehner, B., Althausen, D., Heintzenberg, J., Hellmuth, O., Rinke, R., Schmieder, U., Seidel, C., Tuch, T., Uhrner, U., Wiedensohler, A., Wandinger, U., Wendisch, M., Schell, D., and Stohl, A. 2003. New-particle formation events in a continental boundary layer: First results from the SATURN experiment. *Atmos. Chem. Phys*., **3**, 1445–1459.

Stull R.B., 1988. An introduction to Boundary Layer Meteorology. Dordrecht/Boston/London, Kluver academic publishers, 670p.

Stunder, B.J.B. 1996. An assessment of the quality of forecast trajectories. *J Appl. Meteor.*, **35**, 1319–1331.

Suni T., Breninger F., Vesala T., Markkanen T., Hari P., Mäkelä A., Ilvesniemi H., Hänninen H., Nikinmaa E., Huttula T., Laurila T., Aurela M., Grelle A., Linddroth A., Arneth A., Shibistiva O., and Lloyd, J. 2003. Air temperature triggers of the recovery of evergreen boreal forest photosynthesis in spring. *Glob. Change Biology*, **9**, 1410-1426.

Tarvainen, V., Hakola, H., Rinne, J., Hellén, H., and Haapanala, S. 2007. Towards a comprehensive emission enventory of terpeniods from boreal ecosystems. *Tellus*, **59B**, 526-534.

Tobias, H.J., Beving, D.E., Ziemann, P.J., Sakurai, H., Zuk, M., McMurry, P.H., Zarling, D., Waytulonis, R., and Kittelson, D.B. 2001. Chemical Analysis of Diesel Engine Nanoparticles Using a Nano-DMA/Thermal Desorption Particle Beam Mass Spectrometer. *Environ. Sci. Technol*., **35**, 2233-2243.

Tost, P. Jöckel, A. Kerkweg, A. Pozzer, R. Sander, and J. Lelieveld. 2007. Global cloud and precipitation chemistry and wet deposition: tropospheric model simulations with ECHAM5/MESSy1. *Atmos. Chem. Phys*., **7**, 2733-2757.

Tunved, P., Hansson, H.-C., Kulmala, M., Aalto, P., Viisanen, Y., Karlsson, H., Kristenson, A., Swietliki, E., Dal Maso, M., Ström, J. and Komppula, M. 2003. One year boundary layer aerosol size distribution data from five Nordic background stations. *Atmos. Chem. Phys.* **3**, 2783–2833.

Tunved P., Nilsson E.D., Hansson H.-C., Ström J. 2005. Aerosol characteristics of air masses in northern Europe: Influences of location, transport, sinks, and sources. *J. Geophys. Res.*, **110**, D07201, doi:10.1029/2004JD0005085.

Tunved, P., Ström, J. and Hansson, H.C. 2004. An investigation of processes controlling the evolution of the boundary layer aerosol size distribution properties at the Swedish background station Aspvreten. *Atmos. Chem. Phys.* **4**, 2581–2592.

Tyson, P.D., Garstang, M., Swap, R., 1996. Large-scale recirculation of air over southern Africa. *J Applied Meteorology* **35**, 2218–2236.

Uhrner, U., Birmili, W., Stratmann, F., Wilck, M., Ackermann, I. J., and Berresheim, H. 2003. Particle formation at a continental background site: Comparison of model results with observations. *Atmos. Chem. Phys*., **3**, 347–359.

Vasconcelos, L. A. de P., Kahl, J.D.W., Liu, D., Macias, E.S., and White, W.H. 1999. Spatial resolution of a transport inversion technique. *J. Geophys. Res*. **101**, D14, 19337–19342.

Warnek, P. 1988. Chemistry of the Natural Atmosphere. Academic Press, San Diego, 770p.

Weber, R. J., McMurry, P. H., Eisele, F. L., and Tanner, J. 1995. Measurement of expected nucleation precursor species and 3–500-nm diameter particles at Mauna Loa observatory, Hawaii. *J Atm. Sci*., **52**, 2242–2257.

Wotava, G., and Kröger, H. 1999. Testing the ability of trajectory statistics to reproduce emissions inventories of air pollutants in cases of negligible measurement and transport errors. *Atmos. Environ.*, **33**, 3037-3043.

Yu, F., Wang, Z., and Turco, R.P. Ion-mediated nucleation as an important source of global tropospheric aerosols. Nucleation and Atmospheic aerosols. Eds. O'Dowd, C.D., Wagner, P.E. Springer, 938-942.

Zhang, Q., Worsnop, D.R., Canagaratna, M.R., and Jimenez, J.L. 2005. Hydrocarbon-like and oxygenated organic aerosols in Pittsburgh: insights into sources and processes of organic aerosols. *Atmos. Chem. Phys*., **5**, 3289–3311.