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ABSTRACT

Aerosol formation and subsequent particle growth in ambient air have been frequently observed at a boreal forest site (SMEAR II station) in Southern Finland. The EU funded project BIOFOR (Biogenic aerosol formation in the boreal forest) has focused on: (a) determination of formation mechanisms of aerosol particles in the boreal forest site; (b) verification of emissions of secondary organic aerosols from the boreal forest site; and (c) quantification of the amount of condensable vapours produced in photochemical reactions of biogenic volatile organic compounds (BVOC) leading to aerosol formation. The approach of the project was to combine the continuous measurements with a number of intensive field studies. These field studies were organised in three periods, two of which were during the most intense particle production season and one during a non-event season. Although the exact formation route for 3 nm particles remains unclear, the results can be summarised as follows: Nucleation was always connected to Arctic or Polar air advecting over the site, giving conditions for a stable nocturnal boundary layer followed by a rapid formation and growth of a turbulent convective mixed layer closely followed by formation of new particles. The nucleation seems to occur in the mixed layer or entrainment zone. However two more prerequisites seem to be necessary. A certain threshold of high enough sulphuric acid and ammonia concentrations is probably needed as the number of newly formed particles was correlated with the product of the sulphuric acid production and the ammonia concentrations. No such correlation was found with the oxidation products of terpenes. The condensation sink, i.e., effective particle area, is probably of importance as no nucleation was observed at high values of the condensation sink. From measurement of the hygroscopic properties of the nucleation particles it was found that inorganic compounds and hygroscopic organic compounds contributed both to the particle growth during daytime while at night time organic

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compounds dominated. Emissions rates for several gaseous compounds was determined. Using four independent ways to estimate the amount of the condensable vapour needed for observed growth of aerosol particles we get an estimate of $2–10 \times 10^7$ vapour molecules cm^{-3}. The estimations for source rate give $7.5–11 \times 10^4$ cm^{-3} s^{-1}. These results lead to the following conclusions: The most probable formation mechanism is ternary nucleation (water–sulphuric acid–ammonia). After nucleation, growth into observable sizes ($\geqslant 3$ nm) is required before new particles appear. The major part of this growth is probably due to condensation of organic vapours. However, there is lack of direct proof of this phenomenon because the composition of $1–5$ nm size particles is extremely difficult to determine using the present state-of-art instrumentation

1. Introduction

It is widely recognised that the increasing atmospheric concentrations of greenhouse gases such as carbon dioxide and methane can potentially drive a significant warming process of the earth's climate. However, a topic of more recent attention is the possibility that increased atmospheric concentrations of aerosol particles might drive a significant radiative forcing process of the planet (Charlson et al., 1992; Charlson and Wigley, 1994). The secondary aerosols have both natural and anthropogenic origin. Aerosol particles influence the climate by two distinct mechanisms: the direct reflection of solar radiation by aerosol particles, and the indirect increase in cloud reflectivity caused by enhanced numbers of cloud condensation nuclei. IPCC (1996) has reported that uncertainties in the estimation of direct and indirect aerosol effects on global climate are big. These uncertainties arise largely from the limited information on the spatial and temporal distribution of aerosols and clouds.

For these reasons, particle formation and growth in the atmosphere have recently received growing experimental and theoretical interest. Therefore, instrumental techniques for measuring concentrations of freshly formed particle have been developed, and particles with diameter of about 3 nm can be detected. These small particles have been found in a large variety of environments: in the free troposphere (Clarke, 1992; Schröder and Ström, 1997; Raes et al., 1997), in the marine boundary layer (Covert et al., 1992; Hoppel et al., 1994), in the vicinity of evaporating clouds (Hegg et al., 1991), in Arctic and Antarctic areas (Wiedensohler et al., 1996; Pirjola et al., 1998; O'Dowd et al., 1997), in suburban Helsinki (Väkevä et al., 2000), in coastal areas of the North

Atlantic (O'Dowd et al., 1998), in industrialised agricultural regions in Germany (Birmili and Wiedensohler, 2000), in a mountain site in southern Germany (Birmili et al., 2000) and in rural UK (Coe et al., 2000).

Starting during the mid-nineties, aerosol formation and growth events have been observed also in forested areas e.g., over boreal forest in Finland (Mäkelä et al., 1997, 2000a; Kulmala et al., 1998), and in other type of forests in Portugal (Kavouras et al., 1998), Greece (Kavouras et al., 1999), Canada (Leaitch et al., 1999), and in USA (Marti et al., 1997). In all these cases particle formation and growth events took place in remote forested areas, where the release of highly reactive volatile organic carbons (VOCs) from trees followed by a rapid oxidation to low volatile products, has to be considered as a potential source for nucleating vapours.

During the processes of formation and growth of atmospheric aerosols the aerosol dynamics, atmospheric chemistry and meteorology form a coupled system. The importance of atmospheric chemistry (Pirjola and Kulmala, 1998; Pirjola 1999) as well as meteorological conditions (Nilsson and Kulmala, 1998; Nilsson et al., 2000; Väkevä et al., 2000) on particle formation and growth have been demonstrated under tropospheric conditions. Although ternary nucleation of water-ammonia-sulphuric acid vapours (Korhonen et al., 1999; Kulmala et al., 2000b) has shown to be able to explain atmospheric nucleation — i.e., formation of ~ 1 nm particles — in many cases (Kulmala et al., 2000a), the exact routes for formation of 3 nm particles are still unclear, because besides nucleation, also the growth from 1 nm size to 3 nm size is needed.

The compounds participating in formation and growth of aerosol particles and cloud droplets

originate from anthropogenic or natural sources. The coupling between the biosphere and the atmosphere forms a complex, interactive and non-linear system where the biosphere affects biogeochemical cycles in several ways. In order to evaluate biogenic aerosol formation and growth, continuous measurements of atmospheric aerosols started in Hyytiälä, Southern Finland in January 1996 (Mäkelä et al., 1997). Since then, a number of formation and growth events have been observed. The EU-funded BIOFOR (Biogenic aerosol formation in the boreal forest) project started in October 1997 and continued until the end of September 1999. The strategy of BIOFOR project was to use intensive measurement campaigns and intensive modelling together with continuous measurements to understand the basic processes leading to the aerosol formation and growth, and to quantify the processes.

The two primary objectives of the BIOFOR project were as follows.

1. To determine formation mechanisms of aerosol particles in the boreal forest site.
2. To verify emissions of secondary organic aerosols from the boreal forest site, and to quantify the amount of condensable vapours produced in photochemical reactions of biogenic volatile organic compounds (BVOC) leading to aerosol formation.

In the present paper, we give an overview of the BIOFOR by summarising the methods used and main results obtained during the project.

2. Description of the experiment

2.1. The SMEAR II station

The BIOFOR experiments took place at SMEAR II Station (Station for Measuring Forest Ecosystem-Atmosphere Relations), Hyytiälä, Southern Finland (61°51′N, 24°17′E, 181 m asl) (Fig. 1). The station represents boreal coniferous forest, which cover 8% of the earth's surface and store about 10% of the total carbon in terrestrial ecosystem. The biggest city near the SMEAR II station is Tampere, which is about 60 km from the measurement site with about 200,000 inhabitants.

The 34-year-old Scots pine (*Pinus sylvestris* L.) dominated stand is homogeneous for about 200 m
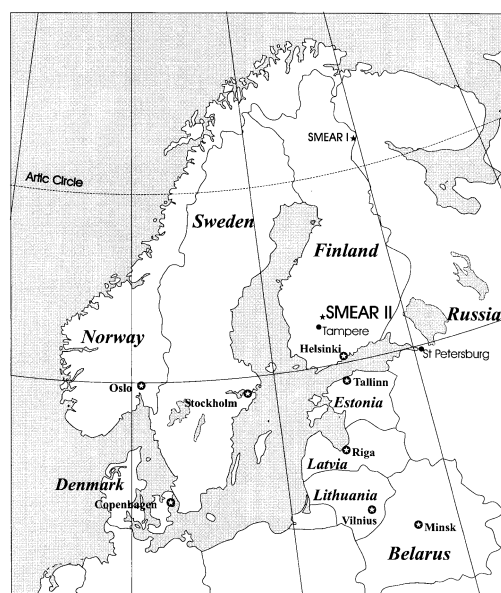

Fig. 1. Map showing the location of the SMEAR II measurement station. In the map some of the major cities are also indicated.

in all directions from the measurement site, extending to the North for about 1.2 km (60° sector). Further from homogeneous stand mainly pine-dominated forests cover extended areas. The terrain is subject to modest height variation. The height of the dominant trees in the stand is 13 m, its zero plane displacement is about 9 m, and roughness length is 1.2 m. The mean diameter at breast height is 13 cm and the total (all-sided) needle area index is 9. The wood biomass is 47 t/ha and the tree density is 2500 per ha. The nitrogen content per total needle area varies from 0.67 to 1.2 g/m² increasing with the height. The nitrogen (mass) concentration is between 1.0 to 1.4%. The dominant stand contains only 1% of species other than Scots pine: downy birch (*Betula pubescens*), grey alder (*Alnus incana*) and aspen (*Populus tremula*). The ground vegetation consists of heather (*Calluna vulgaris*), lingonberry (*Vaccinium vitis-idaea*) and blueberry (*V. myrtillus*). The dominating moss species is *Dicranum undulatum*. The annual mean temperature is 3°C and precipitation is 700 mm. The parent material of the soil is coarse, silty, glacial till and the soil is a haplic podzol.

SMEAR II facility is planned and implemented

to determine material and energy flows in the atmosphere-vegetation-soil continuum at different temporal and spatial scales (Vesala et al., 1998). It can be divided into four operational blocks: I) atmospheric measurements accomplished using a 72 m high mast, II) tree measurements performed using a 15 m high tower, III) soil measurements carried out on two catchment (watershed with weir) areas and IV) aerosol measurements from 2 m above the ground. The station includes advanced set-ups (Vesala et al., 1998) for measurements of aerosol particle size distribution in the size range of 3–500 nm, vertical flux of aerosol particles of diameters down to 10 nm, exchange of trace gases on shoot-scale and soil surface in intervals of 1 min, spatial distribution of irradiance in the vicinity of a shoot by 800 sensors, spatial distribution of irradiance within the canopy with 200 sensors and 2 soil catchment areas (890 and 300 m^2) for the soil water, dissolved ions and organic carbon balances. SMEAR II station is nowadays a platform for number of international measurement campaigns.

2.2. BIOFOR scientific tasks

The BIOFOR project consists of the following scientific tasks:

Task 1: continuous monitoring. Continuous measurements of ultrafine aerosol particle concentrations, their vertical net flux and relevant background data (local meteorology, micrometeorology, vertical profiles of inorganic gases) are obtained in the SMEAR II station. In more detail the continuous measurements are described in Table 1. Task 1 has produced the basic long-term data on aerosol size distributions and their vertical fluxes accompanied by required meteorological data like temperature, relative humidity, solar radiation and the strength of turbulence. In addition, the state of vegetation (level of photosynthesis) and soil (temperature, water content and bacterial and mycorrhizal activity) was determined. In the SMEAR II station the formation and growth of natural, biogenic aerosols can be measured and be connected to the function of trees and soil. The local formation rate of particles and the fate of nascent particles have been identified. Task 1 has included also data evaluation and data delivery.

Task 2: intensive field studies. Three campaigns (14.4.–22.5.1998, 27.7.–21.8.1998 and 15.3.–30.4.1999) on aerosol characteristics (aerosol chemistry and hygroscopic properties) and concentrations of organic and inorganic gases have been performed. The vertical profiles have been determined sampling aerosols from various altitudes using an 18-m tower and a 72-m mast as platforms. The meteorological conditions have been investigated using radiosoundings, sound detection and ranging (SODAR), surface weather observations and back trajectories. Also the vertical profiles of aerosol size distributions and nucleation mode concentrations have been measured. Task 2 has also included the preparation of measurements, calibration of instruments, data evaluation and data delivery. The instrumentation used during campaigns is presented in Table 2. The sites where instruments were used in the intensive field studies are shown in Fig. 2.

Task 3: data evaluation and modelling. The data evaluation, model development and comparison of evaluated data and models have been performed. The modeling work has been divided into four work packages: (a) atmospheric chemistry models, (b) nucleation models (ternary nucleation, ion induced nucleation) (c) Lagrangian model along trajectories with aerosol dynamics and gas phase chemistry, and (d) boundary layer dynamics model with aerosol dynamics and gas phase chemistry. Also the linkage to biological activity as a level of organic emissions is formed.

All data measured during the BIOFOR campaigns are available on the Biofor web pages http://mist.helsinki.fi/Biofor/index.html (ask for usercode and password from the corresponding author). In addition to the numerical data there are also a number of plots produced as a result of the analysis of the data. The data are classified into 9 subgroups: (1) aerosol total number concentration and size distribution measurements in the size range 3–800 nm, (2) aerosol chemistry, (3) aerosol and gas fluxes by eddy covariance and gradient methods, (4) measurements of meteorological parameters and gas concentrations at six different levels from the mast, (5) meteorology of boundary layer and trajectories, (6) concentrations and emissions of BVOC (biological volatile organic compounds), (7) ground level concentrations of inorganic gases, (8) measurements of the

Table 1. *The list of continuous measurements that took place in the SMEAR II station during BIOFOR campaign*

Quantity	Height (m), Location	Instrument	Brand
temperature	67.2, 42.0 and 16.8, mast	Pt-100 sensors	
wind speed	67.2, 42.0 and 16.8, mast	cup anemometer	Vector Instruments A 101 M/L
wind direction	50.4, mast	vane	Vector Instruments W 200 P
RH	23.0, mast	dew point	General Eastern M4
air pressure	2.0, cottage	barometer	Druck DPI260
rain	2.0	tipping bucket counter	Vector Instruments ARG-100
surface wetness	2.0	rain detector	Vaisala DRD 11-A
air stability, wind speed, wind direction	up to 500, institute	sodar (sound detection and ranging)	A Sensitron AB monostatic 2.3 kHz doppler SODAR system
UV-A	15.0, tower	pyranometer	Solar Light SL 501A
UV-B	15.0, tower	pyranometer	Solar Light SL 501A
global radiation	15.0, tower	pyranometer	Astrodata Reemann TP3
reflected radiation	70.0, mast	pyranometer	Astrodata, Reemann TP3
net radiation	70.0, mast	pyranometer	Astrodata Reemann MB1
PAR	15.0, tower	quantum sensor	LiCor LI-190SZ
RPAR	70.0, mast	quantum sensor	LiCor LI-190SZ
NO_x	67.2, 42.0 and 16.8, mast	chemiluminescence	Thermo Environmental TEI 42C TL
O_3	67.2, 42.0 and 16.8, mast	ultraviolet light absorption	Thermo Environmental TEI 49
H_2O (absolute and relative)	67.2, 42.0 and 16.8, mast	infrared light absorption	Hartmann & Braun URAS 4
SO_2	67.2, 42.0 and 16.8, mast	fluorescence	Thermo Environmental TEI 43 BS
CO_2	67.2, 42.0 and 16.8, mast	infrared light absorption	Hartmann & Braun URAS 4
aerosol particle size distribution 3–500 nm	2.0, cottage	electrical mobility	differential mobility particle sizer
aerosol mass 0–2.5 μm and 2.5–10 μm	2.0, cottage	impactor	Dekati PM-10
fluxes (Heat, CO_2, H_2O, particle number)	46.0 and 23.3, mast	eddy covariance, anemometer and infrared light absorption gas analyser, condensation particle counter	Solent 1012R anemometer, LiCor Li-6262 gas analyser and TSI-3010 particle counter
cuvette fluxes (CO_2, H_2O, leaf conductance)	top of the canopy, tree tower	Cuvette	Cuvette, Hartmann & Braun URAS 4

size distribution of wet (ambient) aerosol from 0.5–32 μm at 18 m height, and (9) solar radiation measurements. The detailed descriptions of the instruments used are given on the web pages.

3. Highlights of the results

3.1. *Aerosol concentrations and particle formation events*

At the SMEAR II Station continuous measurements of submicron aerosol number size distribution have been performed since January 1996 (every 10 min). They show approximately 50 days per year with clearly detectable aerosol particle formation events (Mäkelä et al., 2000b). The monthly number of particle formation events during the BIOFOR period (1.10.1997–30.9.1999) is presented in Fig. 3. The most typical time for these events is March–April. Subsequent to the new particle formation, significant particle growth is usually observed. Almost 20% of the events continue sufficiently long to produce particles with diameter over 80 nm. Particles of this size can then become effective cloud condensation nuclei (CCN) especially if they are hygroscopic. Using the particle size distribution data, particle forma-

Table 2. *The instrumentation used in intensive field campaigns during BIOFOR*

| Quantity	Height (m), Location	Instrument	Brand
soundings (p, T, RH, WS, WDIR)	up to 17,000	Rawinsond	Vaisala RS80 and Vaisala DigiCORA
trajectories, 96 h		Trados	Trados
snow depth			
weather maps			
satellite images		NOAA-14	
NH_3	2.0, truck	diffusion scrubber and flow injection (DS-FIA)	
HNO_3 and SO_2	2.0, truck	parallel plate denuder–ion chromatograph (PPD-IC)	
monoterpenes	2.0, tower	Tenax TA, ATD-GC-MS	
carbonyl compounds	2.0, tower	Waters SEP-PAK DNPH-Silica cartridges, HPLC	
light hydrocarbons	2.0, tower	sampling in evacuated electro-polished stainless steel canisters, GC-FID	
monoterpene emissions	2.0, tower	Tenax TA, ATD-GC-MS	
carbonyl compound emissions	2.0, tower	Waters SEP-PAK DNPH-Silica cartridges, HPLC	
organic compunds	64.0, 35.0 and 16.0, mast	Tenax TA + Carbotrap, TDS-GC-MS	
aerosol number concentration	67.0, 18.0 and 2.0, mast	condensation particle counter	TSI-3025 and TSI-3010
aerosol particle size distribution 3–800 nm	67.0, mast	electrical mobility	differential mobility particle sizer
aerosol particle size distribution 10–600 nm	18.0, mast	electrical mobility	differential mobility particle sizer
aerosol particle size distribution 700–20,000 nm	2.0, truck	aerodynamical particle sizer	TSI-3320
wet aerosol particle size distribution	18.0, tower	optical particle counter	PMS CSASP-100 and ASASP-300
ultrafine particle concentration 3–10 nm	2.0, cottage	pulse height analysis ultrafine particle counter	modified TSI-3025
air ions	2.0, tapiola	Gerdien-type ion counters	University of Tartu, Estonia
aerosol hygroscopic properties 10–365 nm	67.0 and 2.0, mast, tapiola	tandem differential mobility particle sizer	TDMA
aerosol CCN properties	2.0, tapiola	cloud condensation nucleus counter	CCNC
aerosol volatility	18.0, mast	optical particle counter	
ice nuclei	2.0, cabin	filter collection + microscopy	
aerosol mass	2.0, truck	tapered element oscillating microbalance (TEOM)	TEOM
organic and elementary carbon	2.0, truck	ambient carbon particulate monitor (Series 5400)	ACPM-5400
event/no event impactor	2.0, forest	DLP-impactor, IC, Mass	Dekati LPI
aerosol main ions	2.0, near cottage	SDI impactor (13-stage), IC	
aerosol main ions	2.0, tree tower	two-stage filter,IC	EMEP
aerosol mass	2.0, cabin	2-stage nuclepore filter	
aerosol mass	2.0, near cottage	PM10 and PM2.5	Dekati impactor
single particle electron microscopy	2.0, cabin	collection to microscope grids, chemical post treatment and transmission electron microscopy	
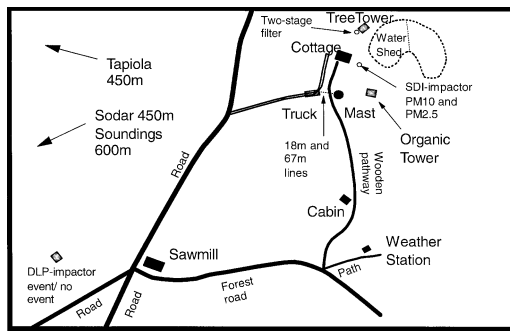
Fig. 2. The map showing location of various instruments during BIOFOR campaign.
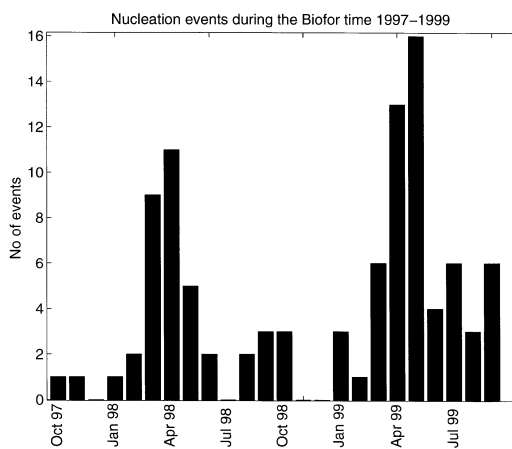

Fig. 3. Histogram showing the monthly number of nucleation events during the Biofor time October 1997–September 1999.

tion events can be classified according to how clear the events are (Mäkelä et al., 2000b). The events were divided into 3 separate classes with a quality number ranging from 1 to 3. The best ones, which showed a clear nucleation mode that was easily distinguishable until it had grown to the Aitken mode size, were classified as class 1. The events with only few nucleated particles, some background, or non-continuous growth characteristics were classified as class 2. In class 3 events, the growing nucleation mode was detectable, but the characteristics related to the formation and growth were very unclear. The classification is subjective, and has to be considered as a guideline for further quantitative analysis. The classification

of the events during intensive campaigns is given in Table 3.

When the particle formation event occurs, the mode of the fresh particles appears into the measurement range. In Fig. 4. aerosol number size distributions measured using Differential Mobility Particle Sizer (DMPS) during a typical nucleation event day are shown. The nucleation mode practically dominates the spectrum with its high number concentration during the nucleation burst. For this event, particle growth from nucleation mode up to accumulation mode is clearly observable. The growth is frequently seen to continue during the following days up to accumulation mode.

Using the DMPS data aerosol the number

Table 3. *The classification of events and the growth rate of newly formed particles during the intensive field campaigns; for more information about classification see Subsection 3.1*

1998		
Date	Class	Growth rate (nm/h)
980414	2	6.3
980415	2	3.3
980416	3	3.3
980425	3	3.3
980511	3	10.5
980513	2	10.5
980517	3	10.5
980520	1	8.6
980521	3	3.3

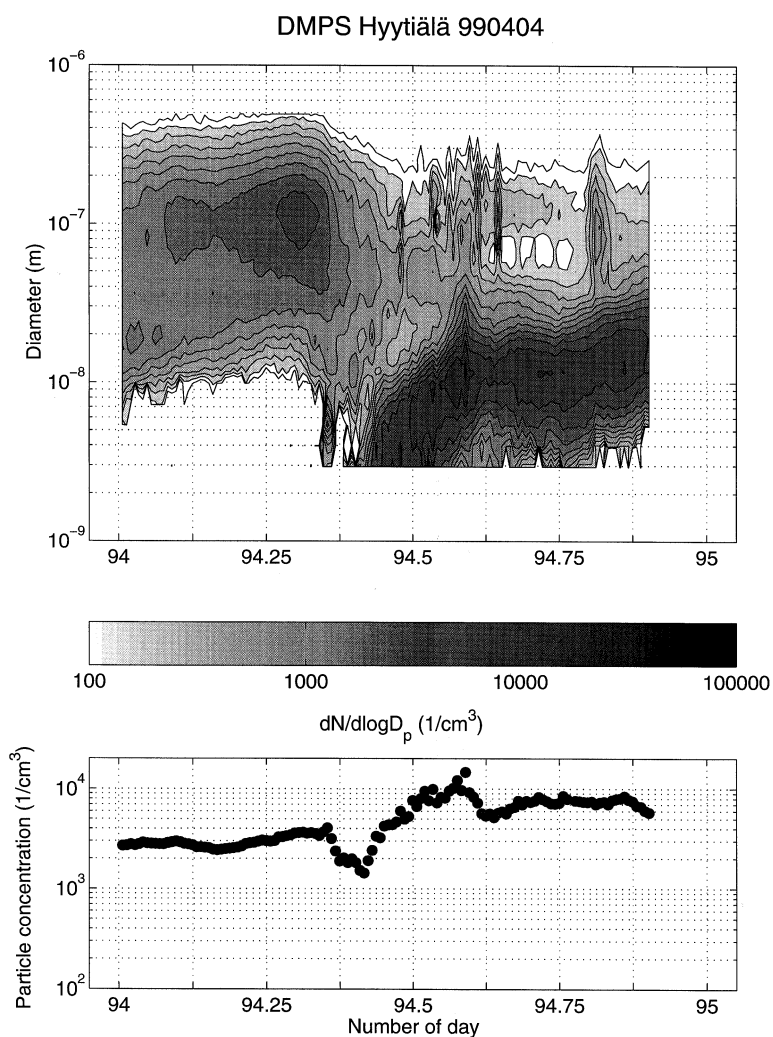| 1999		
Date	Class	Growth rate (nm/h)
990329	3	5.5
990330	1	3.5
990402	1	6.7
990403	2	2.2
990404	1	3.5
990405	1	6.7
990406	1	3.5
990408	2	5.4
990410	3	2.2
990412	1	4.4
990413	2	2.2
990414	1	5.4
990419	1	4.4
990421	3	6.7
990427	2	4.4
990429	3	3.5
990430	3	4.4

DMPS Hyytiälä 990404

Fig. 4. Particle number size distributions measured at 2 m level (long-term measurement) for the nucleation event day 4 April 1999 (top). Also shown the integrated concentration during the same period (below).

concentration of nucleation, Aitken and accumulation modes can be obtained. The daily and monthly averages of these concentrations during the BIOFOR period are given in Fig. 5. The nucleation mode has clear maximum during springtime owing to the nucleation events. During the events, the total particle concentrations increased significantly, typically reaching a concentration of the order of 10,000 cm^{-3} while during non-event periods, the typical concentration is 2000 cm^{-3}. A second maximum of nucleation mode particle concentration is seen during the autumn. The Aitken and accumulation mode particles have maximum that starts early spring lasting till end of summer followed by a winter minimum.

3.2. Synoptic meteorology

Weather maps from the European Meteorological Bulletin and Berliner Wetterkarte and NOAA-14 satellite images in visible and infrared channels were collected to achieve a consistent overview of the day-to-day synoptic

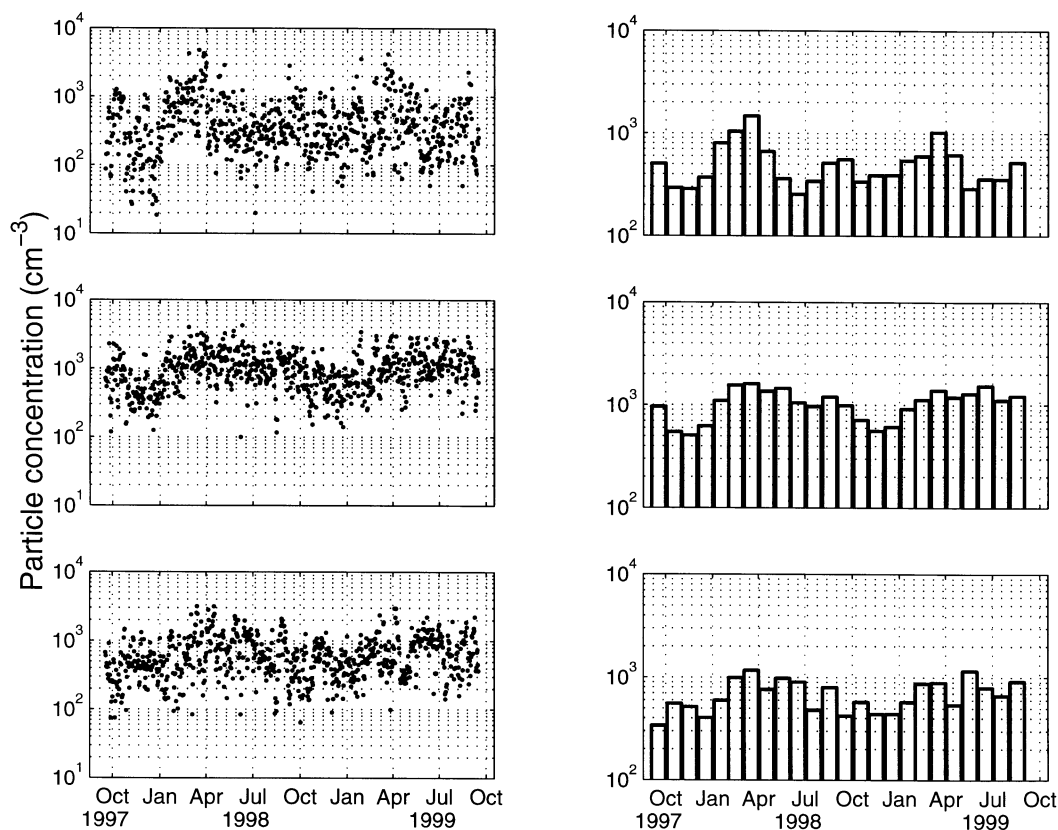*Fig. 5.* The daily (left) and monthly (right) average number concentration of nucleation ($D_p < 25$ nm), Aitken (25 nm $< D_p < 90$ nm) and accumulation ($D_p > 90$ nm) mode particles over the BIOFOR period.

weather patterns during the BIOFOR campaigns and to keep track of air mass movements and positions of fronts and their cloud systems. As an additional support, we have calculated 96-h long back-trajectories with the Lagrangian–Gaussian long-range trajectory and dispersion model TRADOS arriving at Hyytiälä at 5 different pressure levels (1000, 925, 850, 700 and 500 hPa) in 3-h intervals. Trajectories were also calculated based on prognostic pressure and wind fields in order to help decide beforehand the measurements to be made the following day.

Large efforts were made to monitor the boundary layer structure and surface layer fluxes during the BIOFOR field campaigns. Radiosondes were launched on a regular basis by the Finnish Meteorological Institute (FMI) from Jokioinen (179 km southwest of Hyytiälä) at 0 and 12 UTC and from Tikkakoski (93 km northeast of

Hyytiälä) at 6 and 18 UTC. At Hyytiälä soundings were made during BIOFOR 1 and 3 in collaboration with the Finnish Defence Forces. A SODAR system (Sound Detection and Ranging) was used to measure the stability of the air (echo strength) and mean and standard deviation of the horizontal and vertical wind components as well as wind direction up to 500 m. Two eddy covariance systems at 23.3 and 46.0 m levels in the mast in Hyytiälä measured turbulent fluxes in the surface layer. Surface weather observations made at the Kuorevesi/Halli aviation weather station some 20 km east of Hyytiälä were obtained and a data set of daily precipitation and snow cover depth observations at the FMI's Hyytiälä climatological station was compiled.

In terms of synoptic meteorology, particle production occurred in arctic, and to some extent, in Polar air masses, with a preference for air in

transition from marine to continental air masses, and never in sub-Tropical air (Nilsson et al., 2001a). These air masses had an origin north of the BIOFOR experimental location and reached the experimental site through flows from the south-west to north-east. Nucleation was always associated with cold air advection, cold air outbreaks behind cold fronts, and never warm air advection, which related to the typically low cloudiness and large diurnal amplitudes in the continental boundary layer associated with cold air advection and clear skies. However, arctic and polar air together with cold air advection did not lead to nucleation when there was precipitation or when there was a dense cloud cover. The limiting factors were found to be high cloudiness and precipitation, e.g., due to cold front passages and large pre-existing aerosol condensation sink or limited sources of precursor gases. The preference of nucleation to occur in Arctic air masses offers a meteorological explanation to the annual cycle seen in Fig. 3, with a minimum of nucleation events in summer since the Arctic air mass type seldom form in summer. The connection to cold air outbreaks suggests that the maximum in nucleation events during spring and autumn may be explained by the larger latitudinal temperature gradients and higher cyclone activity.

3.3. Boundary layer dynamics and structure

As a result of the weather conditions, nucleation was always associated with the boundary layer evolution during the late morning in a boundary layer transitioning from stable nocturnal conditions to a deep convective well-mixed boundary layer. As the stable nocturnal boundary layer breaks up and a turbulent, convective mixed layer form and grow by vigorous entrainment with from the residual layer, a regular reduction in particle concentration prior to its increase during the production event, is observed due to dilution of the surface layer. The onset of nucleation followed so closely onset of strong turbulence that it strongly suggests that nucleation is associated with either turbulence, convection and/or entrainment (Nilsson et al., 2001b). The delay in observing 3 nm particles after the onset of strong turbulence is between 10 min and 2 h and can be explained by a combination of the turbulent time scale ($\sim$10 min) and the growth time from nucleation

to 3 nm. This, combined with a significantly higher concentration of particles above the canopy compared to within it (Aalto et al., 2001) and a downward turbulent aerosol number flux above the canopy (Buzorius et al., 2001), suggests that the new aerosol particles were formed within the mixed layer or just above it.

Conserved variable mixing diagram analysis indicates that particle production is detected after the surface layer and mixed layer have reached a new mixed-state equilibrium indicating that the production of 3 nm particles occurs within the mixed layer (Nilsson et al., 2001b) and are not entrained from the free troposphere. This analysis does not, however, rule out the entrainment of stable clusters into the new mixed layer where they can grow by condensation processes to detectable sizes over a period of the order of 1 h. When modelling the mixed layer evolution during the morning-noon transition, it appears that nucleation in some cases is connected to the strongest entrainment and growth rates of the mixed layer depth, but, in some cases, nucleation can also start during the turbulent period before this surface layer growth stage.

In summary, it appears that the production region of the new particles is neither in the canopy nor in the free troposphere (but the sources of precursor gases could still be found in either). It remains an open question where in the mixed layer, entrainment zone or residual layer that nucleation occurs. The problem cannot be solved without measurements of the vertical aerosol profile or vertical 1-dimensional aerosol models. In particular, the production region can not be located definitively until instrumentation is available to detect stable clusters of the order of 1 nm.

3.4. Turbulence, vertical fluxes and aerosol profiles

3.4.1. Turbulent fluxes above the forest. Turbulence, vertical fluxes of heat, momentum, carbon dioxide, water vapour, and particles were measured using the eddy covariance method above the forest. Nucleation days occurred during the cold air advection conditions, with prevailing clear sky conditions. Correspondingly, there was more sensible heat transport away from the surface by turbulence compared to other, non-event days (Nilsson et al., 2001b, Buzorius et al., 2001). As a result, buoyancy forces contributed to turbulence

intensity and by the time when nucleation mode particles appeared on event days, turbulence was always increased by buoyancy compared to night-time values.

The highest photosynthetic activity occurs in the summer. Thus, the largest downward CO_2 fluxes, corresponding to CO_2 uptake by the forest, were observed during the summer campaign period. Also the water fluxes were highest in the summer because of increased transpiration. But no clear nucleation events occurred in the summer period.

Most of the nucleation days occurred during the third BIOFOR period. In spring 1999, the photosynthetic activity of the forest started in the end of March and increased throughout April. Although the nucleation days were all sunny, during some of these days preceding low night-time temperatures inhibited the photosynthetic activity of the forest, leading to low CO_2 and water fluxes. Therefore, the CO_2 fluxes above the forest were not correlated with new particle formation events during the given period.

3.4.2. Aerosols fluxes and profiles. During the events, aerosol fluxes determined using an eddy covariance technique are observed to be downwards. Also the measurements made by Condensation Particle Counters (CPC) and DMPS at different heights support this finding. From particle flux data, using the eddy covariance method (Buzorius et al., 1998), usually a small overall downward flux is observed. The downward flux clearly increases during nucleation events, with an exception of the cases when the surface wind was from direction of 220–250° (direction of the Tampere city and the Hyytiälä institute buildings). Then a strong upward particle flux is observed due to local surface-level pollution.

Fig. 6 shows particle fluxes on 4 April 1999, which was one of the clearest event days. The solid lines represent the minimal and maximal fluxes observed during 10 best nucleation days in spring campaign of 1999, see also Buzorius et al. (2001). On 4 April very large absolute fluxes were observed, but there was also large temporal variability. The variation on an hourly time scale is probably due to a mesoscale circulation in convective mixed layer (Nilsson et al., 2001a).

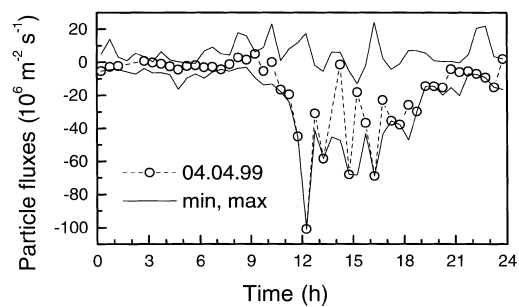The particles were predominantly deposited into the forest during the nucleation events. This

Fig. 6. Aerosol particle ($D_p > 10$ nm) number fluxes during nucleation event day 4 April 1999. The solid lines indicate the minimal and maximal fluxes observed during ten best event days in Spring 1999.

implies higher particle concentrations, and thus sources, above the canopy. The difference between the maximum and minimum fluxes at a given hour is largely because of the temporal variability mentioned above, but also because during different days, events evolved differently in time. In addition, non-stationary particle concentration and random uncertainty of flux estimates might have contributed to the difference. Usually, before the appearance of nucleation mode particles and their corresponding downward flux, a decrease in Aitken and accumulation mode particle concentration occurred when near-surface air was mixed during the mixed layer growth with the air coming from above. This was manifested as upward particle fluxes before the events.

The vertical profile of the size spectra was obtained sampling air from two elevated levels: 18 m and 67 m above ground. The sampling was done using high flow to minimise the particle losses. The event is detected at different levels almost simultaneously and the minor differences make it hard to determine any significant gradient from the particle spectra. This is what is expected in this type of well-mixed boundary layer. The convective time scale was usually around 10 min, that is anything with a lifetime larger than 10 min would be homogeneously mixed through the mixed layer (Nilsson et al., 2001b).

The difficulties in absolute calibration of the DMPS set ups as well as sampling losses in the lines suggested that the gradient of particles will be best determined placing two identical CPC pairs in the mast (18 m and 67 m height). The CPC pairs consisted of the ultrafine CPC (TSI

Inc 3025) for determination of the particles larger than 3 nm in diameter and conventional CPC (TSI Inc. 3010) for particles larger than 10 nm in diameter. The difference of the reading of the CPC's gives an approximate value for the ultrafine mode particle concentrations in the beginning of the burst. The data from the CPC pairs is shown for the event day of 4 April 1999 in Fig. 7. The difference between the CPC

readings from the two levels shows that the ultrafine particles have higher concentrations in higher level during the nucleation burst. This result will support the particle flux data that illustrate a net loss of particles to the canopy; however, it does not necessarily indicate a particle source at the top of the boundary layer or higher altitudes, even though nucleation is more probable in these regions.
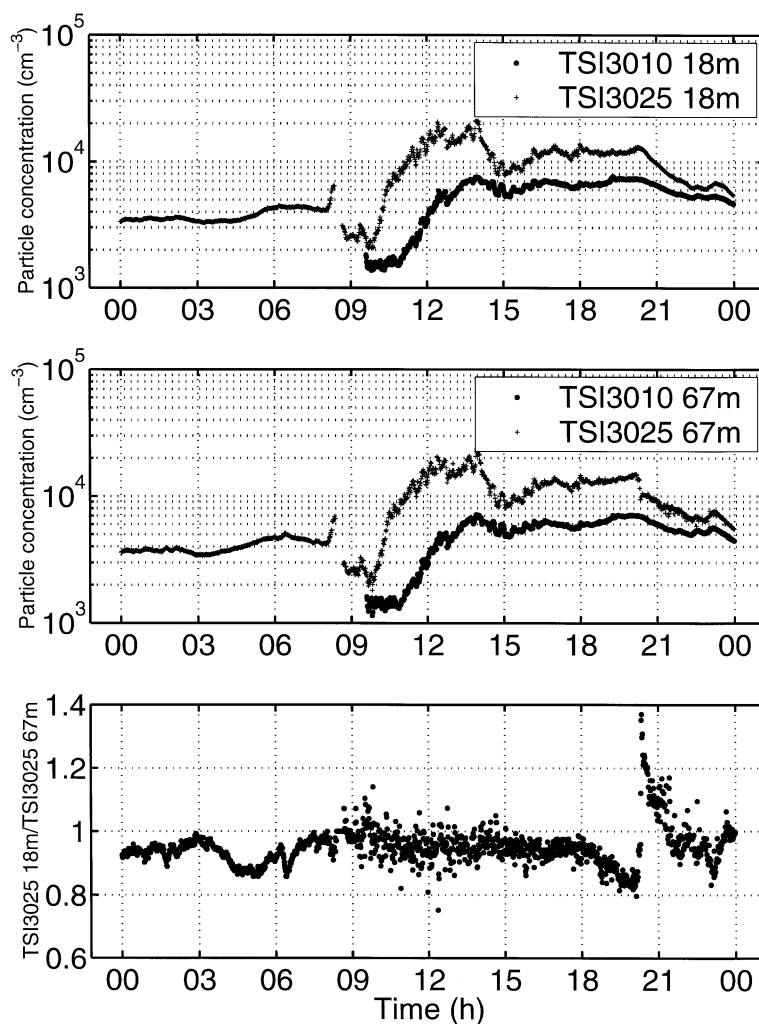

Fig. 7. Total aerosol particle number concentration during 4 April 1999. The concentration of particles larger than 3 nm and larger than 10 nm are shown for 67 m (top) and 18 m (middle) sampling levels. The data is plotted as 1 min averages. The ratio of concentration of particles larger than 3 nm between the two levels is shown at bottom subplot.

3.5. Chemical properties and aerosol composition

3.5.1. Hygroscopic properties of recently formed and evolving aerosols. One of the essential properties related to particle production and growth mechanisms is the composition of the nucleation mode particles. This information is very difficult to obtain due to the small mass of the particles as well as the good time resolution required. The high time and size resolution information on the particle composition was available indirectly as the hygroscopic properties of individual particles (Hämeri et al., 2001). The hygroscopic responses of the nucleation mode particles indicates that the soluble fraction associated with these particles regularly undergo significant changes indicating rapid changes in chemical composition of particles in situ.

The average daily behaviour of the nucleation mode particle soluble fraction during nucleation event days is shown in Fig. 8. The result obtained show clear changes of the composition during the growth process. The soluble fraction is low early morning and starts increasing already before the burst is observed. The maximum value of the soluble fraction is obtained during the growth process in the afternoon. The highest values were obtained for the smallest (10 nm) particles, the median value being close to 0.7. Later in the evening the soluble fraction decreases and has a low value during the night. The high values of the soluble fraction during the nucleation events indicates that the condensing compounds during the

early growth were water-soluble and the particles transformed to become relatively more insoluble later in the evening. It is important also to notice that the diurnal cycle of the soluble fraction was present not only during nucleation event days but also during non-event days. This is clearly indication that other requirements, but high concentrations of condensable gases, are needed to facilitate particle production.

The mass fluxes during the condensation growth were investigated using a model using both soluble and insoluble vapour. As a general result, we found that the mass flux of insoluble vapours are in the same order of magnitude of the mass flux of soluble vapours (Kulmala et al., 2001). The model studies performed during the project by aerosol dynamical model AEROFOR2 (Pirjola and Kulmala, 2001) shows that the sulphuric acid concentration is much less than needed to explain the observed hygroscopic growth. This indicates that a significant fraction of soluble mass can not be explained by sulphuric acid but a soluble organic compound is more likely.

3.5.2. Aerosol chemical composition. In order to be able to obtain more fundamental information about the chemical compounds involved, size segregated aerosol sampling was performed using multi stage low pressure and micro orifice impactors. The analyses for the impactor stages were carried out afterwards with ion chromatography. The impactor samples were taken selectively using two identical impactors. One impactor sampled during the particle formation period, and another one sampled during the times when no particle formation was observed. The division into event and non-event cases was done in situ in the field, based on the on-line DMPS-data (Mäkelä et al. 2001).

The results on the chemical ionic composition of the particles show only small differences between the event and non-event sample sets for most of the compounds such as sulphate and ammonia. Furthermore, no systematic difference between the particulate carboxylic acid concentrations could be found between the two sample sets. In the event samples some ultrafine particulate MSA (methanesulphonic acid) was found to be present. This is a clear consequence of the events showing preference for Polar and Arctic marine air masses. The most significant difference between
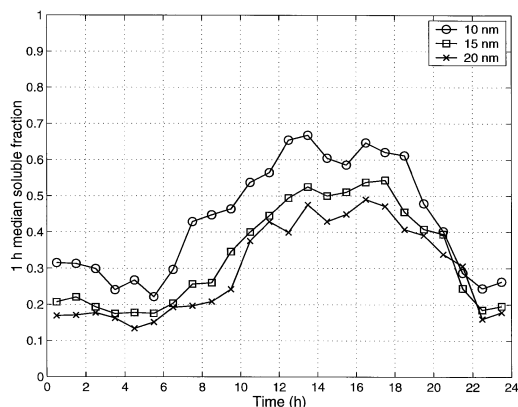

Fig. 8. Average diurnal behaviour of the median nucleation mode particle soluble fraction during the event days of spring 1999 campaign.
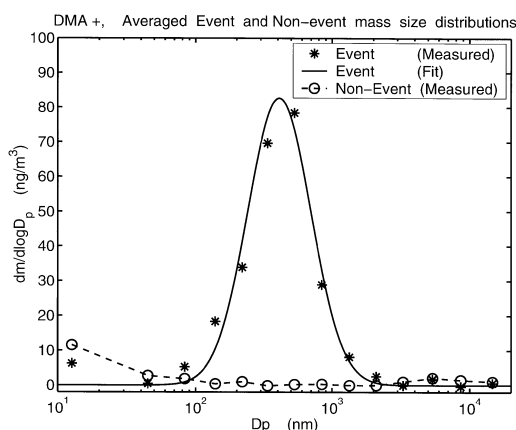
Fig. 9. Average mass size distributions for dimethylamine during event and non-event periods.

the sets was found for dimethylamine (DiMA) (Fig. 9), which seems to be present in the particle phase during the particle formation periods and/or during the subsequent particle growth. Its concentrations during the non-event conditions were much smaller than during the event conditions. During the particle formation periods, an average total concentration of 20–40 ng/m³ of the particulate DiMA was found, whereas during the non-formation periods, particulate DiMA concentrations did not exceed 2–3 ng/m³.

Dimethylamine is a volatile organic base, which behaves rather like atmospheric ammonia. Ambient DiMA has been observed in several locations (e.g., Mosier et al., 1973; Tuazon et al., 1978) but its gas/particle partitioning has been investigated mainly only in marine air (Gibb et al., 1999). Regarding the various sources, DiMA may be assumed to be abundant during most of the time in Hyytiälä, since the amines, as well as NH_3, are known to be end products of the microbial turnover of labile organic matter. Also anthropogenic sources such as industrial activities, feedlot operations, waste incineration and sewage treatment are known to exist.

The relevant question for future investigations is why DiMA is so systematically transformed into the particulate phase only during particle formation periods.

3.5.3. Volatile organic compounds. The diurnal variations of biogenic volatile organic compound (BVOC, mainly monoterpene) concentrations

were measured at four heights (2 m, 17.8 m, 38.4 m and 64.6 m) on selected days during the three BIOFOR campaigns and at 2 m on all days of the spring 1999 campaign. Branch emission measurements were made on selected days.

Janson et al. (2001) found that the leaf emission rates varied from 10^1 to 10^2 ngC gdw^{-1} h^{-1} and showed large variation, as is to be expected under varying conditions of temperature. The standardised rates for the three campaigns lie between 200 and 400 ngC gdw^{-1} h^{-1} (20°C), and are in agreement with rates previously reported for Scots pine. No correlation to event days was found. The main terpenes emitted were α-pinene and Δ^3-carene. In the forest air, they found α-pinene and Δ^3-carene concentrations to account for 48 ± 11% and 23 ± 5% of the ambient monoterpenes (spring 1999). Other terpenes identified were β-pinene, limonene, camphene, myrcene, tricyclene, and occasional traces of sabinene. Isoprene (not emitted by Scots pine) was found in very low concentrations. No significant differences in the relative composition with relation to event and non-event periods were observed.

Concentrations inside the forest ranged from a few hundred pptv to several ppbv, were always highest inside the forest (near the source), and decreased rapidly with height above the forest. Nighttime concentrations were usually higher than daytime, in spite of lower emissions, because of the shallower and more stable nocturnal boundary layer. The vertical profile (Spanke et al., 2001) and diurnal variation at 2 m (Janson et al., 2001) are shown in Fig. 10. With the exception of one short (2–3 h) episode of unusually high terpene concentrations on the morning of 25 April 1998 (which we have not been able to explain) nothing exceptional with regards to branch emissions or concentrations was observed which might explain the frequency of nucleation events during the spring. In fact, there was no correlation between monoterpene concentrations and number concentration of nucleation particles. Also, concentrations were highest during the summer period (as is to be expected because of the higher temperatures), when no well-defined nucleation events were observed. However in general the concentrations were higher on event days, although not with any high degree of confidence.

3.5.4. Inorganic gases. Janson et al. (2001) also found that the SO_2 concentrations ranged from
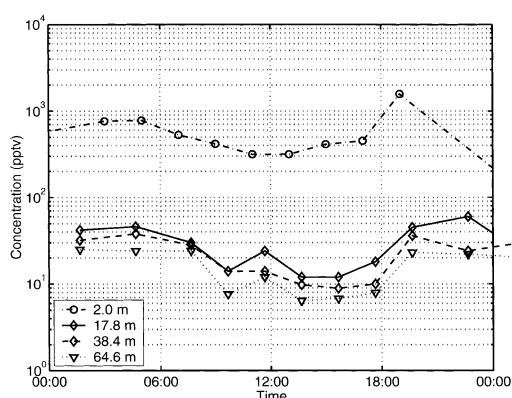
Fig. 10. Diurnal behaviour and vertical gradient of α-pinene concentration measured during 11 August 1998. The lines connecting the data points are for guiding the eye.

less than 5 pptv to 1600 pptv during the spring 1999 campaign and were generally higher during event days as compared to non-event days, indicating that there might be a connection. Average event day concentrations were on the order of 200 to 400 pptv. Summertime concentrations were generally lower, averaging 90 pptv, but showed a larger variation between minimum and maximum, and showed no significant difference between event and non-event days. Ammonia concentrations were also higher on event days during both the summer and spring periods. Summer concentrations ranged from 10 to 430 pptv and averaged 52 pptv, while springtime concentrations ranged from 5 to 400 pptv and averaged 49 pptv.

In contrast to SO_2 and NH_3, HNO_3 showed higher concentrations during the non-event days, not the event days. Average daytime concentrations were then 200 pptv, while they were about 100 pptv during event days and during the summer 1998 campaign. At the low values for HNO_3 and NH_3 observed here, the gas-particle partitioning equilibrium between $HNO_{3(g)}$, $NH_{3(g)}$ and $NH_3NO_{3(p)}$ is of little interest.

4. Discussion

4.1. Production of particles and condensable vapours

Kulmala et al. (2001) developed an analytical tool taking advantage of only the measured aero-sol spectral evolution as a function of time, to derive formation and growth properties of nucleation mode aerosols. This method, when used with hygroscopic growth factors, can also estimate basic composition properties of these recently-formed particles. From the diameter growth-rate derived from size spectra, aerosol condensation and coagulation sinks can be calculated (Kulmala et al., 2001). Using this growth-rate and condensation sink, the concentration of condensable vapours and their source rate can be estimated. Then, combining the coagulation sink together with measured number concentrations and apparent source rates of 3 nm, 1 nm particle nucleation rates and concentration can be estimated. To estimate nucleation rates and vapour concentration source rates producing new particle bursts over the Boreal forest regions, three cases from the BIOFOR project were examined using this analytical tool. In this environment, the nucleation mode growth-rate was observed to be 2–3 nm h^{-1}, which required a condensable vapour concentration of $2.5{-}10 \times 10^7$ cm^{-3} and a vapour source rate of approximately $7.5{-}11 \times 10^4$ cm^{-3} s^{-1} to be sustained. The formation rate of 3 nm particles was ≈ 1 particle cm^{-3} s^{-1} in all three cases. The estimated formation rate of 1 nm particles was $10 - 100$ particles cm^{-3} s^{-1}, while their concentration was estimated to be between 10,000 and 100,000 particles cm^{-3}. Using hygroscopicity data and mass flux expressions, the mass flux of insoluble vapour is estimated to be of the same order of magnitude as that of soluble vapour, with a soluble to insoluble vapour flux ratio ranging from 0.7 to 1.4 during these nucleation events.

The variation of growth rate and condensation sink (for concepts see Kulmala et al. 2001) is presented in Table 3 and Fig. 11. The growth rate of nucleation mode particles varies between 2 nm/h to 10 nm/h. The vapour depletion due to the condensation sink is seen to be somewhat higher on non-event days. There was one class 3 event during which the sink term was also high. However, during that day the vapour concentration was high enough (growth rate 4 nm/h) to produce 3-nm particles and to obtain detectable particle growth. According to our results, a large condensation sink can prevent the particle production event from occurring, but a small sink does not necessarily indicate that 3 nm particle production events will happen.
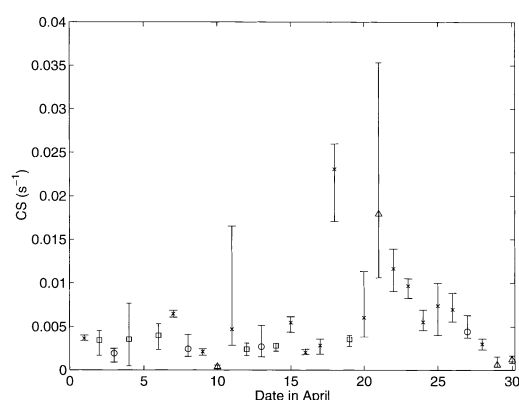
Fig. 11. The variation of the condensation sink during April 1999. Average value as well as maximum and minimum values observed are plotted. Squares (class 1), circles (class 2) and triangles (class 3) indicate the nucleation event days and crosses indicate the non-event days.

Using the near surface measurements of monoterpenes, SO_2, NH_3, NO_x, O_3, and radiation, Janson et al. (2001) calculated the steady state $\cdot OH$ and NO_3^- concentrations for the summer-98 and spring-99 campaigns and then the source terms for the production of condensable gases from the monoterpenes (Q_{SOC}) and SO_2 (Q_{H2SO4}). They found that the spring-99 values for Q_{SOC} varied from 5×10^4 to $7 \times 10^7 \, cm^{-3} \, s^{-1}$ with an average of $2 \times 10^6 \, cm^{-3} \, s^{-1}$. Values on event days were generally higher than non-event days, although the difference was only weakly significant. Daytime values were higher during the summer, averaging $5 \times 10^6 \, cm^{-3} \, s^{-1}$ by the $\cdot OH$ oxidation alone. These values are all well above the source rate needed for growth as calculated by Kulmala et al. (2001) suggesting that about 10–15% of oxidation products will transfer to condensable vapours. Janson et al. (2001) also observed that the nighttime production of condensable gases was generally larger than the daytime production. Since the NO_3 radical is the most important oxidant at night, followed by O_3, the nighttime products ought to be more prone to condense, yet no nucleation was observed at nighttime. Most likely, at least some of the products condensed onto existing particles as the source term $Q_{SOC}(NO_3^-)$ was found to correlate to accumulation mode organic carbon.

Pirjola and Kulmala (2001) used aerosol dynamics model (AEROFOR2) to obtain the

source rate and very nice agreement with the analytical method was found: they found that the fraction of oxidation products to form condensable vapours is about 15%. As a conclusion, the source rate can be estimated within one order of magnitude.

In order to be able to explain the observed particle growth the concentration of the condensable vapour should be over $2 \cdot 10^7 \, cm^{-3}$ as discussed before. However, according to recent simulations (Pirjola and Kulmala, 2001) the sulphuric acid concentrations were typically below $1 \cdot 10^7 \, cm^{-3}$. Therefore some other condensable vapour was needed to explain the growth to nucleation and Aitken modes (see more details Kulmala et al., 2000a).

4.2. Possible particle formation mechanisms

In principle the atmospheric particle formation can occur via different pathways. Recently e.g., Korhonen et al. (1999) investigated ternary nucleation, Yu and Turco (2000) ion-induced nucleation and Leck and Bigg (1999) amino acids as possible pathways. All the three hypothesis were investigated during BIOFOR. Kulmala et al. (2000b) have compared ternary (water–sulphuric acid–ammonia), binary (water–sulphuric acid) and ion induced (water–sulphuric acid–ions) nucleation. According to our simulations both ammonia and ions can considerably enhance the nucleation rates compared to the binary (water–sulphuric acid) systems. These simulations also showed, that ternary nucleation seems to be the most effective process producing the new particles under atmospheric conditions when ammonia concentration exceeds 1–2 pptv. At measured temperatures (typically below 5°C), at measured ammonia concentrations (always over 10 ppt, often over 50 ppt) and estimated daytime sulphuric acid concentrations (10^6–$10^7 \, cm^{-3}$) the nucleation rates calculated using the recently developed ternary nucleation code were high enough (more than $100 \, cm^{-3} \, s^{-1}$). Although we did not study all possible nucleation pathways (such as nucleation of organic compounds) in the present study, we can conclude that ternary nucleation seems to be very effective under ambient conditions.

The other possible mechanism is related to the biological activity. Bigg (2001) investigated the possibility that amino acid, L-methionine, is

responsible for new particle formation. Although this mechanism is plausible there are still several unsolved problems connected with this mechanism. The ion induced nucleation mechanism is not strong enough to explain the observed aerosol formation.

Nevertheless, Janson et al., (2001) found that the calculated source rate of sulphuric acid times the ammonia concentration was correlated to the number concentration of nucleation mode particles during the events, thus corroborating the suggestion that the initial nuclei are produced by ternary nucleation processes involving sulphuric acid and ammonia and that the number of particle actually produced may be determined by the sulphuric acid source rate. The calculated source rates of sulphuric acid were not, however, high enough to sustain observed particle growth as estimated by Kulmala et al. (2001), although the sulphuric acid may also enhance the condensation growth.

We have also found obvious connections between the aerosol formation and the special meteorological conditions found necessary for the nucleation. There are several possible explanations for the correlation between the onset of turbulence and nucleation: (1) new aerosols or clusters may have been entrained from the residual layer into the mixed layer where they then (in the case of clusters) underwent growth to detectable sizes; (2) two or more precursor gases may have been mixed with each other over the entrainment zone; (3) the adiabatic cooling in the rising convective plumes and the turbulent fluctuation in temperature and vapours by the entrainment flux may have enhanced aerosol formation; (4) a sudden decrease in pre-existing aerosol due to dilution of the mixed layer aerosol by entrained air may have reduced the vapour sink enough to initiate nucleation.(Nilsson et al., 2001b). However theoretically the ternary nucleation mechanism seems to be strong enough to produce aerosol particles without any enhancement.

4.3. Estimation of emissions of secondary organic aerosols during BIOFOR

During the BIOFOR period, a continuous DMPS data set has also been measured in the SMEAR I station (Värriö, Lappland) and in downtown Helsinki (at the Department of Physics

University of Helsinki). The data sets show that new particle formation can occur simultaneously over a large region under normal atmospheric conditions. Around 15–20% of these particle formation events can be seen at least in two out of three of these sites.

Using the experimental data from three measurement sites, Mäkelä et al. (1998) estimated aerosol production over the larger area. Using the BIOFOR data we repeat the estimation here. Events are observed on about 50 days/year in Finland producing 1000–5000 particles cm^{-3} per event, amounting to 50,000–100,000 particles cm^{-3} $year^{-1}$. Rawisonde soundings indicate that the planetary boundary layer height is typically 1000 m in Southern Finland during the events. If detectable particles are formed only within the mixed layer, we get the particle production rate ca. $5–10 \times 10^{13}$ particles m^{-2} $year^{-1}$ over Finland. For global average over vegetated land we get $3–6 \times 10^{27}$ particles $year^{-1}$. This order of magnitude is comparable with other global estimates (Radke and Hobbs, 1976). However, the result is very uncertain.

During the BIOFOR campaign we were able to show that a significant (mass) fraction of the new aerosols is organic compounds, which very probably is of biogenic origin. In order to be able to observe new particle production, the particle diameter should be at least 3 nm of size. Although the probable nucleation mechanism is ternary nucleation, other vapours are required (e.g., organic acids) to grow the particles to 3 nm size. Since the nucleation itself seems to happen easily the key question is how, and under what conditions, these small clusters grow to observable sizes (see also Kulmala et al., 2000a). For this reason, we conclude that the observed aerosols have a natural origin, and we can estimate that the production of natural (biogenic) aerosols from forested areas probably constitute a significant fraction of the global atmospheric aerosol.

5. Conclusions

The major scientific impact of the project is to provide quantitative estimation of aerosol formation and growth at a boreal forest site. The results will also be applied in aerosol packages in regional and global scale numerical models.

As scientific conclusions we are able to give preliminary answers to our objectives

(1) The most probable formation mechanism is ternary nucleation (water–sulphuric acid–ammonia) and the growth to observable sizes takes place mainly owing to condensation of organic vapours. Nevertheless, there is no direct proof of this phenomenon because the composition of 1–5 nm size particles is very difficult to determine using present state-of-art instrumentation. However nucleation was found only in correlation with increased sulphuric acid source term and ammonia concentrations.

(2) Nucleation takes place always in very specific weather conditions: cold air advection in Polar and Arctic air masses, at low cloudiness, and no precipitation. Furthermore, the nucleation was closely connected to the onset of strong turbulence in the morning-noon transition from stable to unstable stratification, which should also correspond to the onset of convection and entrainment from aloft.

(3) The emissions rates for several gaseous compounds have been determined. Using 4 independent ways the amount of the condensable vapour needed for observed growth of aerosol particles was estimated to 2–10×10^7 vapour molecules cm^{-3}. The estimations for source rate gives 7.5–11×10^4 cm^{-3} s^{-1}.

6. Future directions

Resulting from the BIOFOR project, close connection between physics and chemistry of particle formation with meteorology and partly also with biological activity has been shown. Although, nucleation bursts have been observed simultaneously around Finland (Mäkelä et al., 1998; Väkevä et al., 2000), see also Fig. 6 in Nilsson et al. (2001a)

more experiments are needed to verify the actual geographical scale of this phenomena. In addition, more accurate measurements of condensable organic compounds are necessary in order to identify the condensable vapours needed for aerosol growth.

The IPCC (1996) report highlights the fact that the current estimates of the global mean radiative forcing due to anthropogenic aerosols is highly variable (0.3 to -3.5 Wm^{-2}) and of comparable magnitude but opposite sign to the radiative forcing owing to anthropogenic greenhouse gases. To estimate the radiative forcing due to anthropogenic aerosols, the formation and growth mechanisms of natural aerosols and their importance in climate processes needs to be known. As a result, predictions of climate change due to anthropogenic influences are inaccurate and it appears difficult for the policy makers to make decisions about strategic policy changes related to climate change. The understanding of natural, biogenic aerosols that results from BIOFOR measurements and process model studies will enable more reliable climate model predictions. This in turn will provide improved understanding of natural process to allow more informed decision making.

7. Acknowledgements

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