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Vertical fluxes and micrometeorology during aerosol particle formation events

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ABSTRACT

Fluxes of aerosol particles with sizes larger than 10 nm together with fluxes of momentum, sensible and latent heat and CO_2 were measured 10 m above a Scots pine forest with the eddy covariance method. During days when nucleation events were observed particle size distribution measurements showed particle growth from 3 nm sizes to the Aitken mode. Analysis of the experimental data showed systematic differences in fluxes during the days when new particle production was observed compared to other days. During the nucleation events the particle flux measurements showed downward aerosol particle transport, i.e., indicating an elevated source, with respect to the measurement level, of particles larger than 10 nm. Furthermore the turbulence intensity and the heat fluxes were observed to be significantly higher. Evidences of mesoscale circulation were observed in wind speed records as well as in turbulent fluxes on nucleation days. The measurement results show that micrometeorology, the synoptic scale conditions and the particle formation are closely related.

1. Introduction

Recently, particle production events have been observed at different locations: remote continental (Mäkelä et. al., 1997), urban background (Birmilli and Wiedensohler, 2000) and marine coastal environment (O'Dowd et al., 1998). A field campaign BIOFOR (Biogenic aerosol formation in the boreal forest) was conducted to study biogenic aerosol formation observed in boreal Scots pine forest (Hyytiälä, SMEAR II station, southern Finland). Little is known about the nature of the new particle formation observed in the lower atmosphere. Weber et al. (1999) argued that ambient conditions determine the mechanism for atmospheric nucleation. Kulmala et al. (2000) suggested a hypothesis that thermodynamically stable cluster with sizes about 1 nm grow by condensation in the Atmospheric Boundary Layer (ABL) to detectable sizes, which is at least 3 nm. According to experimental evidence the particle production is dependent on solar radiation and pre-existing aerosol surface area as a condensational sink for precursor gases (Weber, 1997). Therefore sunny days are more favourable for new particle formation due to dependence of the precursor species on solar radiation (Weber, 1997; Clement et al., 2001). However, all sunny days do not lead to nucleation according to observations, indicating other limiting factors for nucleation to occur in these days. Unfortunately, neither the particle formation and the growth mechanism, nor the chemical composition of newly formed particles is known.

Recently, more detailed studies on meteorology give new interesting ideas. Nilsson et al. (2001a) showed that the particle formation events at the

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SMEAR II station occurred during the advection of cold Arctic and Polar air masses and that the formation could occur in the same air mass from northern to southern Finland and further to south on the same day. This particle formation is likely to occur within the ABL. The ABL processes during the nucleation days were extensively discussed by Nilsson et al. (2001b). New particle entrainment from the free troposphere was found to be very unlikely because the new particles were observed in the morning prior to the growth of the mixed layer through the residual layer into the free troposphere. There are also micro-meteorologically different conditions at smaller spatial scales inside the atmospheric surface layer (ASL) accompanying the cold air advection during the nucleation days, and these differences are demonstrated and discussed in detail in this study. The vertical fluxes of momentum, sensible heat, H_2O, CO_2, and aerosol particles in the ASL during the days categorised as nucleation event days, non-nucleation event days but with cold air advection, and other days are compared.

2. Measurement setup

The SMEAR II (Station for Measuring Forest Ecosystem-Atmosphere Relations) field measurement station is located in Hyytiälä, Southern Finland (61°51′N, 24°17′E, 181 m asl), within extended areas of pine dominated forests. Detailed description of the SMEAR II station can be found in Vesala et al. (1998) and the description of measurement arrangements during the BIOFOR campaigns in Kulmala et al. (2001). The tower of the atmospheric measurements at SMEAR II is located on a moderately sloping terrain and surrounded by a homogeneous Scots pine stand. The site is described in micrometeorological context by Rannik (1998). The conditions at the site are typical for background location. The local pollution from the station buildings (0.5 km) and city of Tampere (60 km), both located west-south-west from the station, affect occasionally the measurements at SMEAR II.

Two eddy covariance systems were used to measure vertical fluxes. The flux measurements were performed at 23 and 46 m height, approximately 10 and 33 m above the forest canopy. The systems consisted of a sonic anemometer (Solent Research R3, Gill, UK), and an infrared gas analyser for CO_2 and H_2O (LI 6262, Li-Cor, USA). A condensational particle counter (CPC), TSI model 3010 (for instrument description, see TSI (1996)), was mounted only at the lower level to measure the particle number fluxes. The trace gas flux instruments are typical of those used in eddy covariance flux systems of CO_2 and H_2O and their operation has been described extensively in the literature (Moncrieff et al., 1997; Aubinet et al., 2000). The signals were digitised and recorded together with three wind speed components and sonic temperature at 21 Hz.

Original details of the CPC eddy covariance system can be found in Buzorius et al. (1998) and various aspects of its application and operation are described by Buzorius et al. (2000). Additional technical details concerning its operation were studied during BIOFOR and are reported below. Contemporaneous aerosol size distribution measurements were performed using Differential Mobility Particle Sizer (DMPS). The size distributions were obtained for 10 min periods. The instrument and the measurement results are described in greater detail by Aalto et al. (2001). For the purpose of this paper the DMPS data are used to highlight periods when the nucleation mode dominated the aerosol size distribution in order to classify the particle number flux data.

During BIOFOR 1 (time periods are presented below) two CPCs (referred to as CPC1 and CPC2 hereafter) of the same type (model TSI 3010) but with different cut-off sizes were employed using a common sample inlet. The cut-off size of CPC1 was set to 14 nm by adjusting the temperature difference between the condenser and the saturator of the instrument to 14°C, and the cut-off size of CPC2 to 7 nm, with a corresponding temperature difference of 21°C. The cut-off sizes for the CPCs were subsequently confirmed by laboratory calibration using furnace-generated silver particles as described by Buzorius (2001). The purpose of using two CPCs was to observe the vertical transport of particles with sizes between 7 and 14 nm. During the summer measurement campaign in 1998 (BIOFOR 2, spring 1998) only one CPC with the cut-off size of 10 nm was employed in the flux measurements.

During BIOFOR 3 (spring 1999) the CPC TSI 3010 was modified to operate at 3 lpm sampling flow rate (10 nm cut-off size). This modification improved the response time of the counter (Buzorius, 2001), and thus, allowed better detection of the fast fluctuations in the aerosol number concentration time series. In parallel to this CPC the second CPC of the same type was operated under standard conditions, i.e., 1 lpm sampling flow rate corresponding to a response time of 0.8 s and a 10 nm cut-off size, to validate concentration measurements of the modified CPC. Measured concentrations were the same, indicating that the modified CPC operated reliably. Differences in fluxes were negligible, showing no substantial difference in the flux values measured at a height of 10 m above the canopy by two CPCs with time constants of 0.8 and 0.4 s. More detailed discussion on the frequency attenuation by CPC can be found in Buzorius et al. (2000).

3. Results

Three field campaigns were conducted from 14.04.1998 to 22.05.1998 (BIOFOR 1), from 27.07.1998 to 21.08.1998 (BIOFOR 2) and from 15.03.1999 to 30.04.1999 (BIOFOR 3). The flux observations during BIOFOR campaigns consist of 4587 half-hour periods. The micrometeorological fluxes of momentum (and friction velocity), heat, CO_2, H_2O and particles were calculated as 30 min average co-variances between the scalars (or horizontal wind speed) and vertical wind speed according to commonly accepted procedures (Aubinet et al., 2000). Except for momentum, upward fluxes were defined to be positive. The aerosol size distributions obtained by DMPS system were also averaged for half-hour periods.

In addition, wind speed and direction, temperature, water concentration of the air and global and net radiation, continuously measured at SMEAR II station with intervals varying from 1 min to 5 min depending on quantity, were averaged over half-hour periods for further analysis. Also the original records and 5 min average fluxes were used in the analysis to reveal short-time behaviour of some quantities.

3.1. *Particle flux distributions and statistics*

The average particle number flux over all BIOFOR campaigns was -1.05×10^6 particles $m^{-2} s^{-1}$ with a standard deviation of 13.88×10^6 particles $m^{-2} s^{-1}$. The negative sign indicates small net particle deposition. Over all observation periods the upward fluxes were almost as frequent as downward.

A sample of the half-hour average flux values, plotted according to wind direction, is shown in Fig. 1. Significant emission fluxes were observed during the south-west winds, suggesting a local source within the measurement source area. The Hyytiälä field station buildings were located in this direction some 700 m from the measurement tower, which were the likely source of these emissions. North-west winds brought relatively clean (small pre-existing aerosol concentration) air masses and the particle production was registered with the DMPS system. At the same time large deposition fluxes were measured, typical for the events. During other periods the fluxes were relatively small.

After removing the periods with wind from the station (a sector 215 to 265° was identified as being affected by the station), the dominant particle flux direction is downwards (Fig. 2). However, relatively small upward fluxes are still frequent. The upward flux values are not representative of the particle emissions from the forest and occur due to temporary pollution episodes from other directions than the direction of station buildings, advective non-stationarity in concentration time series (Gallagher et al., 1997a) and random uncertainty in flux estimates.

Fig. 2a shows the cumulative frequency of occurrence of particle number flux data obtained

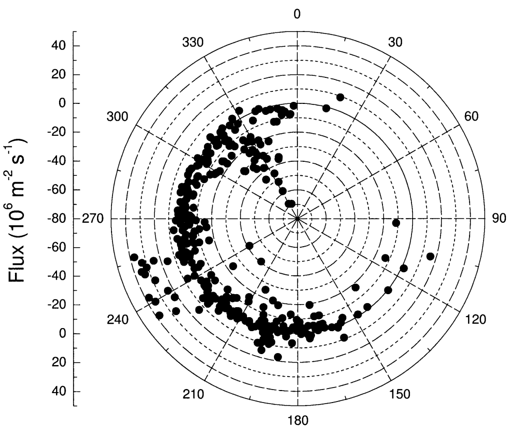

Fig. 1. Half-hour average aerosol particle number fluxes versus wind direction on 1–7 April 1999.

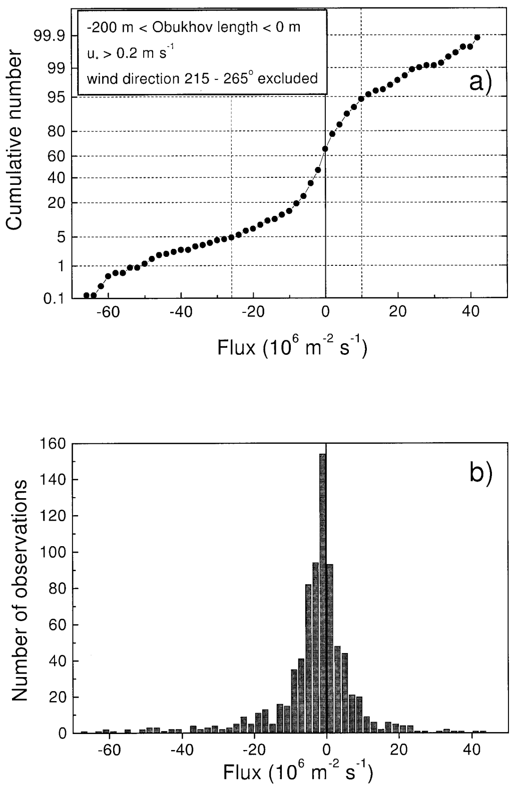

Fig. 2. Cumulative frequency distribution (a) and frequency distribution (b) of half-hour average particle number fluxes measured during BIOFOR campaigns. Data for near-neutral to unstable conditions were selected according to stability criterion indicated in the figure. Low turbulence conditions (friction velocity less than $0.2\ \mathrm{m\,s^{-1}}$) and wind direction from the station were excluded.

from all three measurement campaigns during unstable stratification conditions (for the definition of the Obukhov stability length see, e.g., Stull (1988)). In addition the data set was filtered to limit the observations to friction velocities greater than $0.2\ \mathrm{m\,s^{-1}}$. With the additional removal of data from the unsuitable fetch direction this eventually yielded a quality controlled data set consisting of 801 data points. Fig. 2b shows that the smallest fluxes occur most frequently, as is expected. The frequency distribution is negatively skewed with 67% of fluxes directed downwards. The 5% and 95% percentile levels are also shown. In 5% of cases the particle fluxes were larger than 10×10^6 particles $\mathrm{m^{-2}\,s^{-1}}$ and in 5% of cases the magnitude was larger than 26×10^6 particles $\mathrm{m^{-2}\,s^{-1}}$ in deposition regime.

If periods where the nucleation mode dominated the ambient aerosol size distribution are selected, the particle flux histogram looks quite different, Fig. 3a, b. The data in this figure includes the observations when more than half the particle concentration was given by particles with sizes below 20 nm as determined by the DMPS size distribution. 20 nm is roughly a size separating nucleation and Aitken modes in size spectrum. During particle formation events, when the number concentration of nucleation mode particles was relatively high, large downward fluxes dominated. In contrast to Fig. 2, the majority of fluxes during the events were downward (87% of cases). The result may be interpreted as being due to a source above the measurement level. This is

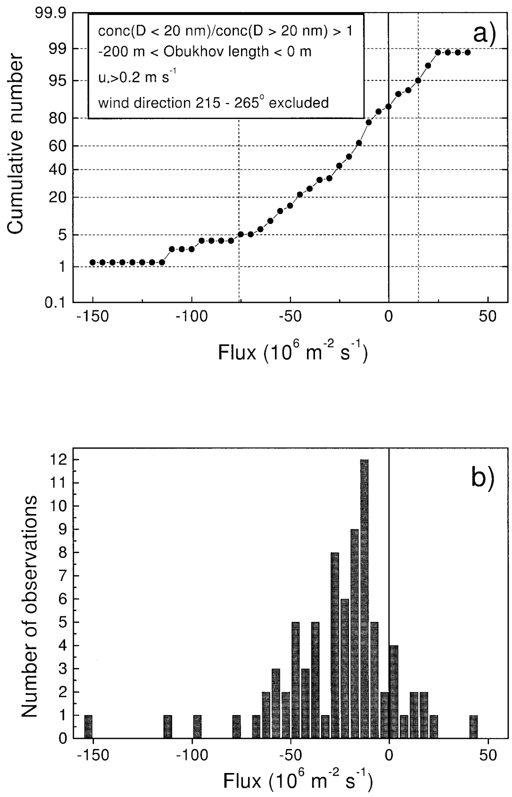

Fig. 3. The same as Fig. 2, but only for time periods when concentration of particles with diameters less than 20 nm was higher than concentration of larger particles according to DMPS measurements at 2 m level.

in agreement with Nilsson et al. (2001b) who found that nucleation probably occurred in the mixed layer or the entrainment zone, perhaps in the residual layer, but not in the free troposphere.

3.2. *Diurnal variation during different periods*

The data was classified according to 5 different categories listed below. All nucleation events occurred during cold air advection conditions (Nilsson et al., 2001a). Therefore the classification based on cold air advection and nucleation occurrence, and additionally on the season of the campaign.

The 1st category contained days when particle formation was observed. These days originate from the spring BIOFOR 1 (1998) and BIOFOR 3 (1999) measurement campaigns. The 2nd category corresponds to days from BIOFOR 2 campaign during the summer period (1998) when cold air advection occurred but new particle formation was not observed. The 3rd category corresponds to days when cold air advection occurred but no new particle production was observed, during the spring campaigns BIOFOR 1 and BIOFOR 3. The forth category was used to classify the remaining days from the summer campaign (not included in the category 2), and the category 5 the days not included in the categories 1 and 3. Days from BIOFOR 2 were analysed separately because of different conditions (temperature etc.) in the summer.

An average of particle fluxes over the event days is shown in Fig. 4a (open circles). Although the individual nucleation days were somewhat different in temporal evolution, the average diurnal course illustrates the general behaviour well. During the particle formation events large particle deposition (downward flux) was observed. Before the increase in downward particle flux, frequently upward particle transport occurred (around 9.00–10.00 a.m. in the figure). Examination of the DMPS data revealed that simultaneously particle concentrations dropped in the Aitken and accu-

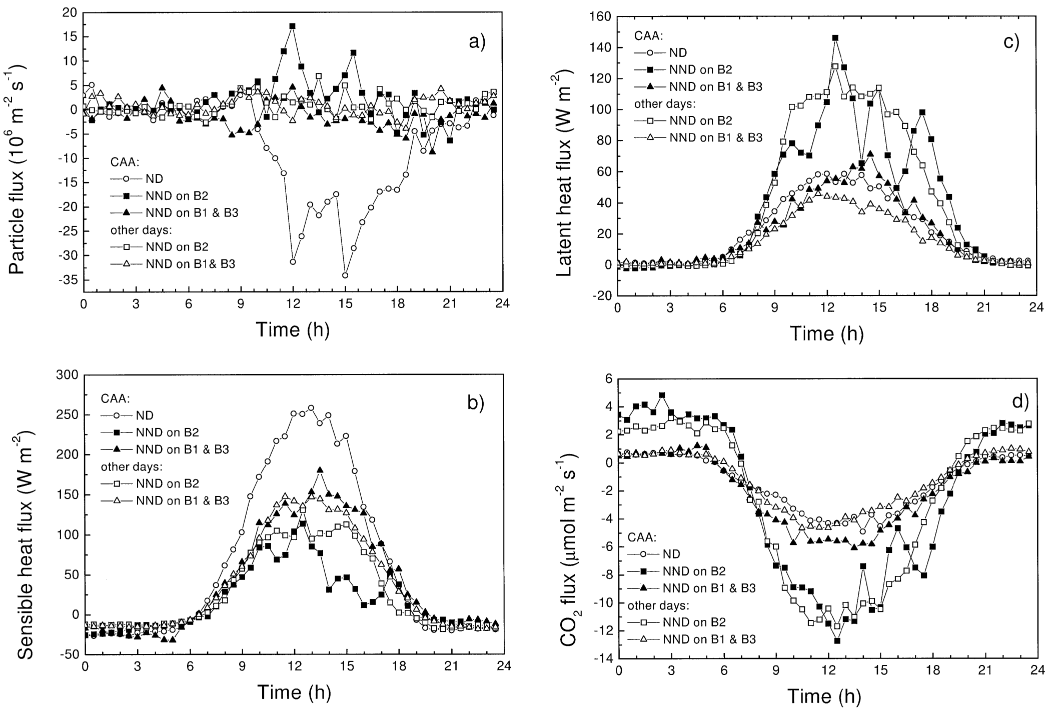

Fig. 4. Average daily courses of aerosol particle (a), sensible heat (b), latent heat (c), and CO_2 (d) fluxes for different categories of days. CAA — cold air advection; ND — nucleation day; NND — non-nucleation day; B1, B2, B3 — BIOFOR campaigns.

mulation modes before the beginning of the new particle production. Evidently, the concentration decrease and upward fluxes are connected. During the mixed layer growth the particle concentrations are reduced by the entrainment of air from the residual layer, which presumably has lower particle concentration (Nilsson et al., 2001b; Stull, 1988). This corresponds to upward particle transport from the air layers close to the surface, where initially the particle concentration was higher. It was typical that decreasing concentrations in Aitken and accumulation modes during the rapid growth of mixed layer coincided with upward fluxes.

Sensible heat fluxes (Fig. 4b) were clearly higher during the event days compared to other categories (Nilsson et al., 2001b). Latent heat fluxes (Fig. 4c) were highest during the summer season, when transpiration was much higher compared to springtime. Maximum CO_2 uptake occurred in summer during the highest photosynthetic activity of the canopy (Fig. 4d). Also the night-time respiration was significantly higher in summer. During the event days in spring CO_2 fluxes were slightly smaller than during the other days, on average, although on nucleation days incoming radiation was higher. This was the result of smaller CO_2 uptake during the event days in 2 to 6 April 1999. On those days the ambient temperature was lower compared to days before and after the period. Subsequently, primarily radiation controlled CO_2 uptake was inhibited due to low temperatures. Based on the CO_2 flux data, no apparent connection between the photosynthetic activity of the forest (a possible indicator of biogenic emissions of precursor gases for nucleation and condensation) and the particle formation occurrence was observed.

Fig. 5 shows additional meteorological information: temperature, water vapour concentration, net radiation, and standard deviation of vertical wind speed. During summertime temperatures (Fig. 5a) and water vapour concentrations (Fig. 5b) were on average almost twice as high as those in springtime. On particle formation days in spring the air was slightly cooler and especially dry. The average daily course of water content was different on nucleation days compared to non-nucleation day, decreasing before noon from 8 a.m. until 12 a.m. However, there were several non-event days, which were as dry as the event days. Among the event days the highest water content was 7 mmol mol^{-1} (volume mixing ratio). An examination showed that the highest concentration of nucleation mode particles coincided with the lowest water vapour concentration (below 5 mmol mol^{-1}). However, one has to remember that conditions close to surface might be very different from the conditions aloft where presumably nucleation takes place.

Nucleation event days were characterised by higher levels of net radiation (Fig. 5c), which was responsible for elevated turbulent heat fluxes (Fig 4b). This was the most noticeable difference between event and non-event days, primarily as a result of lower cloudiness during nucleation days (Nilsson et al., 2001a). In summer the net radiation fluxes were smaller, probably due to cloudiness, which lead to reduction in incoming solar radiation, and increased infrared energy loss from the surface. Fig. 5d shows averaged standard deviation of vertical wind speed. The event days are characterised by higher turbulence intensities and more intensive mixing compared to non-event days, allowing mixing of surface-based precursor gases to higher altitudes and possible initiation of nucleation, followed by subsequent growth of newly formed particles to detectable sizes.

3.3. *Nucleation mode particle fluxes*

During BIOFOR 1 (spring 98) two CPCs with cut-off sizes of 7 and 14 nm were employed in the eddy covariance system to measure vertical particle fluxes. Throughout most of the period the concentrations and fluxes measured by both instruments were very close. Before noon on May 20, 1998, the concentration difference, indicative of the 7–14 nm concentration, reached 1000 particles per cm^3 (Fig. 6). Measured fluxes were different too, but the exchange velocities, obtained through normalisation by average concentration as $V_e = -$flux/concentration, were the same. The positive exchange velocities (identical to deposition velocities) of particles for sizes between 7 and 14 nm were about 6–7 mm s^{-1}, being very close to the exchange velocities obtained by both CPCs. The deposition velocity is expected to be higher for smaller particles (between 7 and 14 nm) due to higher diffusivity compared to Aitken mode particles. The exchange velocity at the observation level might not represent well the surface depos-

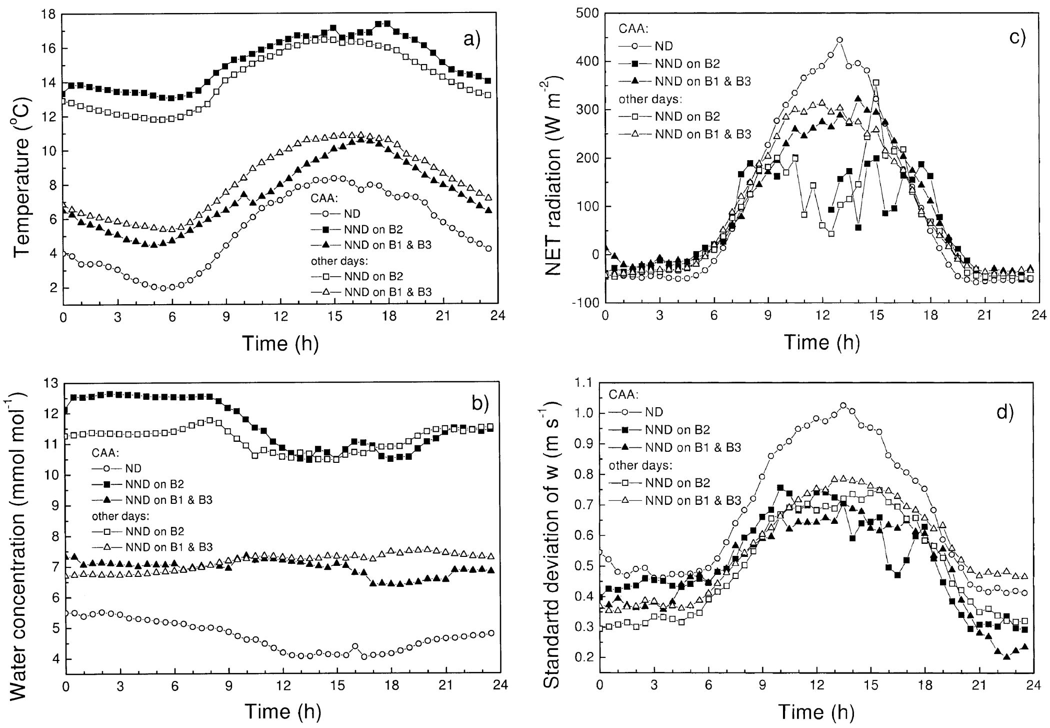

Fig. 5. Average daily courses of temperature (a), water vapour concentration (b), net radiation (c), and standard deviation of vertical wind speed (d) for different categories of days. CAA — cold air advection; ND — nucleation day; NND — non-nucleation day; B1, B2, B3 — BIOFOR campaigns.

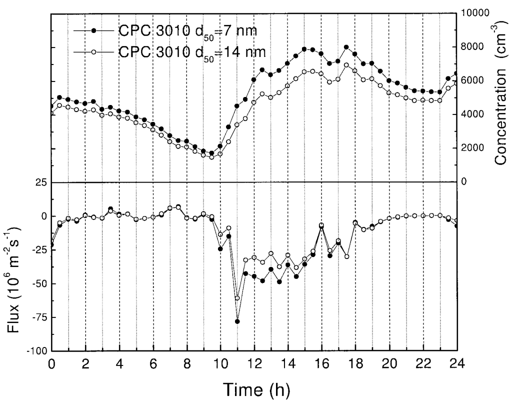

Fig. 6. Particle concentrations and fluxes as determined by two CPCs with cut-off sizes of 7 and 14 nm on 20 May 1998, during the event day when nucleation mode particle concentration was high.

ition velocity during the periods of concentration changes, when storage or release of concentration in the air layer occurs. Unfortunately, no more episodes were registered by the two CPCs with high concentration differences between 7 and 14 nm.

Modelled values of deposition velocities into the forest (Gallagher et al., 1997a,b) compare reasonably to our measurement results. Experimental results on deposition velocities of nucleation mode particles are scarce. Schery et al. (1998) have reported deposition velocities of nanometer-size particles over semiarid desert to be as high as 5–35 cm s^{-1}. Buzorius et al. (2000) have estimated experimentally deposition velocities for nucleation mode particles (most of particles between sizes from 10 to 20 nm as determined by DMPS size spectra) over forest at current experimental site from 1 to 40 mm s^{-1}, with the tendency for deposition to increase with turbulence intensity (friction velocity). The present experimental setup with two CPCs enabled more distinct separation of particle sizes.

3.4. *Evidences of mesoscale variation*

The particle flux time series in Fig. 6 reveals variation in consequent half-hour values. This was a common observation in particle flux time series on nucleation days and deserves additional attention. Variation on such a time scale is characteristic to mesoscale roll circulation in the boundary layer (Atkinson, 1996).

Roll circulation manifests itself close to the surface as periodic variation in wind speed (Smedman, 1991). Variation with periodicity of a few tens of minutes was observed in wind speed records. Fig. 7 shows spectra of vertical and crosswind speeds close to the surface, at 46.0 m height. The mean wind direction and correspondingly three wind components were determined over an hourly period, over which also the spectra were calculated. The variation is best seen as low frequency spectral peak in cross-wind component, with energy maximum at periodicity about 20 min. The variation in vertical wind speed is largest in the middle of the boundary layer, but is hardly seen in the vertical wind spectrum at 46.0 m as the vertical motion is limited by the presence of surface.

Turbulent fluxes are usually calculated over half-hour to hourly period to catch all the frequencies contributing to turbulent flux. However, in half-hour values the variation characteristic to roll circulation is averaged out. Momentum transport and heat fluxes reveal periodicity when averaged over 5 min (Fig. 8a, b), although part of the variability can certainly be ascribed to random uncertainty of flux estimates when averaged over a short period. The events of intensive momentum transport were accompanied with increased sensible and latent heat (not shown) fluxes. During the momentum transport events also increase in downward turbulent particle fluxes occurred (Fig. 8c). The periodicity in momentum and heat fluxes was observed already in the morning before the nucleation mode particles were detected by the DMPS system around 10 am. This was the case also in other nucleation days as far as the existence of roll circulation can be confidently detected by spectral analyses. Adiabatic cooling in rising air may be what initiates the nucleation, as suggested by Easter and Peters (1994) and Nilsson et al. (2001b). Thus mesoscale circulation might play an important role in initiation of

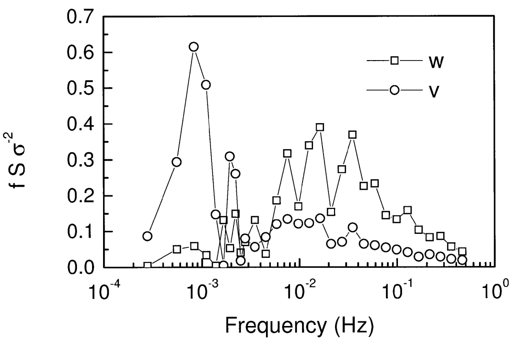

Fig. 7. Power spectra of vertical and cross-wind speeds observed close to the surface at 46.0 m height on 12 April 1999, 12:00–13:00.

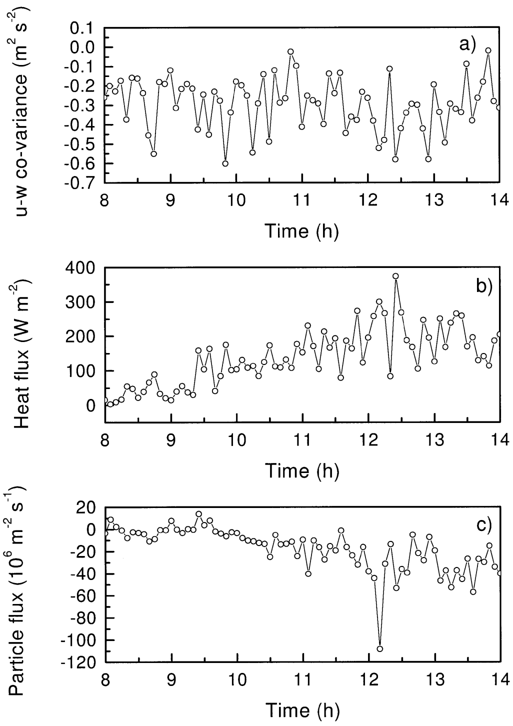

Fig. 8. 5-min average co-variances of vertical and along-wind speeds (a), heat fluxes (b), and aerosol number fluxes (c) on 12 April 1999, measured at 23.3 m height.

nucleation and/or particle growth to detectable sizes.

Although correlation between roll circulation events and particle transport was observed, no conclusions on correlation with nucleation can be made based on particle fluxes larger than 10 nm in diameter. If roll circulation drives the nucleation, this cannot be seen in current data. Only if condensational growth was extremely fast, the increase in nucleated clusters in an air parcel could be accompanied by an increase in aerosol number detected by our measurement system. However, unrealistically large vapour pressure of condensable gases is needed for nucleation to show up in several nm aerosol particle number virtually in the same air parcel (Nilsson et al., 2000).

Mesoscale rolls commonly occur in cold air outbreaks (Atkinson and Zhang, 1996), which was always the case on nucleation days (Nilsson et al., 2001a). Roll circulation frequently leads to formation of cloud streets, but rolls can also occur in clear air (Puhakka and Saarikivi, 1996). Roll circulation in combination with clouds can result in favourable conditions for nucleation to occur: the updrafts associated with rolls possibly transport precursor gases from close to surface into higher levels, where clouds have cleansed the air of the larger aerosol, otherwise being limiting factor as surface for condensational sink. During nucleation days fast reduction in Aitken and accumulation mode aerosol concentrations was sometimes (about one third of the nucleation days) observed during the ML growth in the morning (Nilsson et al., 2001b). Inspection of satellite images and global radiation records revealed that cloud streets and/or convective cells were frequently present during nucleation days, but not always. Note that even though roll vortices may exist earlier, they usually form cumulus clouds and cloud streets in the afternoon. On 20 May 1998, cloud streets and/or convective cells, being seen as a decrease in incoming solar radiation (Fig. 9a), were present already when the particle burst was observed in size spectrum at about 8:30 a.m. On 4 April 1999, however first disturbances in radiation due to clouds appeared more than an hour after nucleation had started before 10 a.m. (Fig. 9b). At midday visible cloud streets were observed over the whole southern Finland (Fig. 10). Nucleation occurred also in totally cloud-free days. More detailed analysis of the role

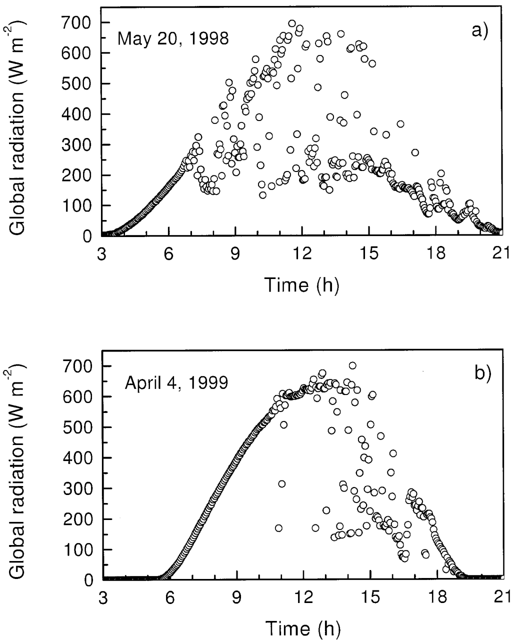

Fig. 9. Global radiation above the forest on 20 May 1998 (a), and on 4 April 1999 (b).

of mesoscale convection and cloudiness in nucleation events is out of the scope of this paper.

4. Concluding discussion

The fluxes of aerosol particles, sensible and latent heat and CO_2, and some meteorological parameters, measured 10 m above a Scots pine forest during the BIOFOR campaigns in Hyytiälä, southern Finland, have been analysed and presented. Days from the campaign periods were classified according to seasonality (BIOFOR 1 and BIOFOR 3 versus BIOFOR 2), occurrence of synoptic scale cold air advection and particle formation events. Table 1 summarises the differences in turbulent fluxes and some meteorological parameters during different categories of days.

Nucleation events occurred only in cold air advection conditions during the spring campaigns. The cold air advection days without nucleation and the days with no cold air advection did not differ significantly in spring as well as in summer in terms of studied paramaters. No clear nucleation events were observed during the summer

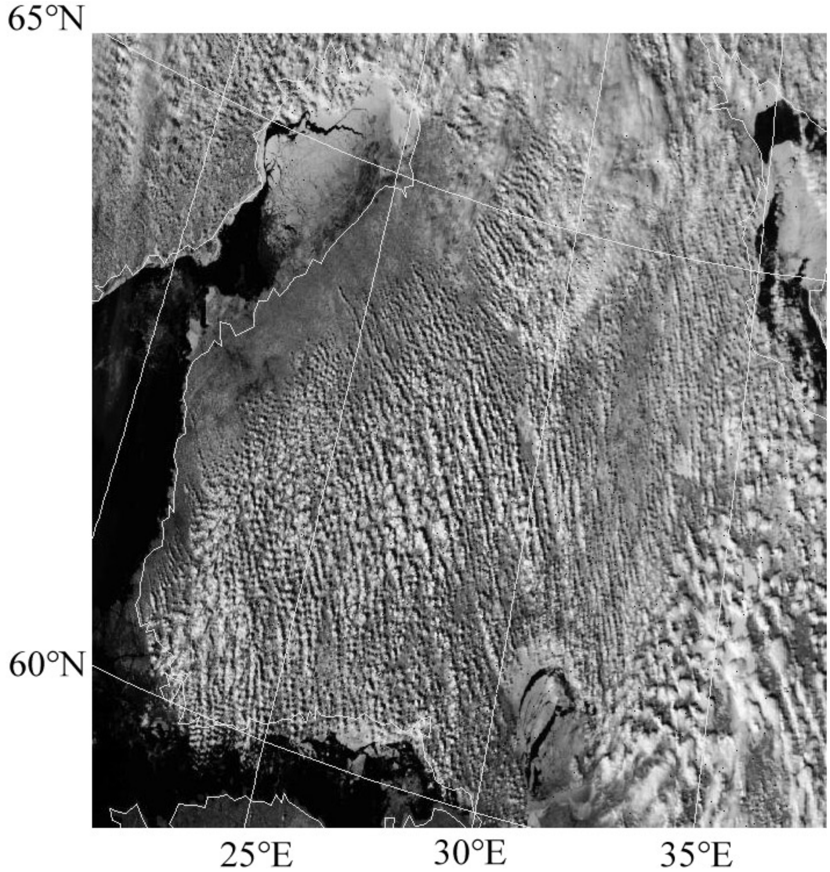

Fig. 10. NOAA-14 satellite image (from the satellite receiving station of the University of Dundee), visible light, channel 2, from 4 April 1999, 12:40 UTC, illustrating existance of cloud streets over the whole southern Finland.

Table 1. *Summary of differences between different data categories*[a]

	CAA N	CAA B2	CAA B1 + B3	Rest of B2	Rest of B1 + B3
particle number flux	large and down				
sensible heat flux	enlarged				
latent heat flux		larger		larger	
CO_2 flux		larger		larger	
temperature	lower	high		high	
H_2O concentration	low	high	intermediate	high	intermediate
NET radiation	enlarged	lower	intermediate	lower	intermediate
turbulence intensity	higher				

[a] CAA — cold air advection; N — event day; B1, B2 and B3 — BIOFOR campaigns.

BIOFOR 2 campaign. The summer period was different from spring with higher temperatures, higher water content of the air and water fluxes, and bigger CO_2 uptake by forest. The net radiation in summer was relatively low compared to springtime.

The biggest difference was observed between the nucleation days and days of other categories in spring. Large downward particle number fluxes, higher turbulence intensities and intensive vertical mixing, and high net radiation and sensible heat fluxes were observed during the nucleation days. On average the air temperature and water content were low during the nucleation days. Due to low temperatures on nucleation days also the CO_2 uptake by forest was low, on average. The biogenic emissions of precursor gases for nucleation and/or condensation can be linked to photosynthetic activity of forest, but no connection between the photosynthetic activity of the forest and the particle formation occurrence was observed.

Evidences of mesoscale BL circulation were observed during the nucleation days. The surface layer wind spectra revealed secondary maxima in energy, with the periodicity of few tens of minutes. The roll circulation was accompanied with periodic variation in surface layer momentum, heat, and aerosol particle fluxes. Rolls circulation frequently leads to formation of cloud streets, but they can also occur in clear air. The nucleation days were generally sunnier, with higher shortwave radiation input at the surface, which in turn led to the higher heat fluxes and turbulence intensity. However, clouds (streets and convective cells) were frequently present during nucleation days, but not always. Nucleation occurred also in totally cloud-free days.

The rôle of mesoscale circulation and cloudiness in particle production and transport deserves more attention in future studies. The results show that micro-meteorology, meso- and synoptic scale conditions and particle formation are closely related.

5. Acknowledgements

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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.

Atkinson, B. W. and Zhang, J. W. 1996. Mesoscale shallow convection in the atmosphere. *Reviews of Geophysics* **34**, 403–431.

Aubinet, M., Grelle, A., Ibrom, A., Rannik, Ü., Moncrieff, J., Foken T., Kowalski, A. S., Martin, P. H., Berbigier, P., Bernhofer, Ch., Clement, R., Elbers, J., Granier, A., Grünwald, T., Morgenstern, K., Pilegaard, K., Rebmann, C., Snijders, W., Valentini, R. and Vesala, T. 2000. Estimates of the annual net carbon and water exchange of European forests: the EUROFLUX methodology. *Adv. Ecol. Res.* **30**, 113–173.

Birmili, W. and Wiedensohler, A. 2000. New particle formation in the continental boundary layer: Meteorological and gas phase parameter influence. *Geophys. Res. Letters* **27**, 3325–3328.

Buzorius, G., Rannik, Ü., Mäkelä, J. M., Vesala, T. and Kulmala, M. 1998. Vertical Aerosol particle fluxes measured by eddy covariance technique using condensational particle counter. *J. Aerosol Sci.* **29**, 157–171.

Buzorius, G. 2001. Cut-off sizes and time constants of CPC TSI 3010 operating at flow rates from 1 to 3 lpm. *J. Aerosol Sci.and Techn.*, in press.

Buzorius, G., Rannik, Ü., Mäkelä., J. M., Keronen, P., Vesala, T. and Kulmala, M. 2000. Vertical aerosol fluxes measured by the eddy covariance method and deposition of nucleation mode particles above a Scots pine forest in southern Finland, *J. Geophys. Res.* **105**, 19,905–19,916.

Clement, C. F., Pirjola, L., dal Maso, M., Mäkelä, J. M. and Kulmala, M. 2001. Analysis of particle formation bursts observed in Finland. *J. Aerosol Sci.*, in press.

Easter, R. C. and Peters, L. K. 1994. Binary homogeneous nucleation: Temperature and relative humidity fluctuations, non-linearity and aspects of new particle production in the atmosphere. *J. Appl. Meteorol.* **33**, 775–784.

Gallagher, M. W., Beswick, K. M., Duyzer, J., Westrate, H., Choularton, T. W. and Hummelshøj, P. 1997a. Measurements of aerosol fluxes to Speulder forest using a micrometeorological technique. *Atmos. Environ.* **31**, 359–373.

Gallagher, M., Fontan, J., Wyers, P., Ruijgrok, W., Duyzer, J., Hummelshøj, P., Pilegaard, K. and Fowler, D. 1997b. Atmospheric particles and their interaction with natural surfaces. In: *Biosphere — atmosphere exchange of pollutants and trace substances. Transport of pollutants in the troposphere* (ed. S. Slanina). Springer-Verlag, Heidelberg, pp. 45–92.

Kulmala, M., Hämeri, K., Aalto, P. P., Mäkelä, J. M., Pirjola, L., Nilsson, E. D., Buzorius, G., Rannik, Ü., Maso, M. D., Seidl, W., Hoffmann, T., Janson, R., Hansson, H.-C., Viisanen, Y., Laaksonen, A. and O'Dowd, C. D. 2001. Overview of the international project on Biogenic aerosol formation in the boreal forest (BIOFOR). *Tellus* **53B**, 327–343.

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

Mäkelä, J. M., Aalto, P., Jokinen, V., Pohja, T., Nissinen, A., Palmroth, S., Markkanen, T., Seitsonen, K., Lihavainen, H. and Kulmala, M. 1997. Observations of ultrafine aerosol particle formation and growth in boreal forest. *Geophys. Res. Letters* **24**, 1219–1222.

Moncrieff, J. B., Massheder, J. M., de Bruin, H., Elbers, J., Friborg, T., Heusinkveld, B., Kabat, P., Scott, S., Sogaard, H. and Verhoef, A. 1997. A system to measure surface fluxes of momentum, sensible heat, water vapour and carbon dioxide. *J. Hydrology* **188–189**, 589–611.

Nilsson, D., Pirjola, L. and Kulmala, M. 2000. The effect of atmospheric waves on aerosol nucleation and size distribution. *J. Geophys. Res.* **105**, 19,917–19,926.

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

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

O'Dowd, C., Geever, M., Hill, M. K., Smith, M. H. and Jennings, S. G. 1998. New particle formation: nucleation rates and spatial scales in the clean marine coastal environment, *Geophys. Res. Letters* **25**, 1661–1664.

Puhakka, T. and Saarikivi, P. 1986. Doppler radar observations of horizontal roll vortices in Finland. *Geophysica* **22**, 101–118.

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

Schery, D. S., Wasiolek, P. T., Nemetz, B.M, and Yarger, F. D. 1998. Relaxed eddy accumulator for flux measurement of nanometer-size particles. *Aerosol Science and Technology* **28**, 159–172.

Smedman, A.-S. 1991. Occurrence of roll circulations in a shallow boundary layer. *Boundary-Layer Meteorol.* **57**, 343–358.

Stull, R. B. 1988. *An introduction to boundary layer meteorology*. Kluwer Academic Publishers, Boston, Mass., USA, 666 pp.

TSI, 1996 Model 3010 Condensational Particle Counter Instruction Manual, TSI Inc., St. Paul, MN.

Vesala, T., Haataja, J, Aalto, P., Altimir, N., Buzorius, G., Garam, E., Hämeri, K., Ilvesniemi, H., Jokinen, V., Keronen, P., Lahti, T., Markkanen, T., Mäkelä, J. M., Nikinmaa, E., Palmroth, S., Palva, L., Pohja, T., Pumpanen, J., Rannik, Ü., Siivola, E., Ylitalo, H., Hari, P. and Kulmala, M. 1998. Long-term field measurements of atmosphere-surface interactions in boreal forest combining forest ecology, micrometeorology, aerosol physics and atmospheric chemistry. *Trends in Heat, Mass and Momentum Transfer* **4**, 17–35.

Weber, R. J., Marti, J. J. and McMurry, P. H., Eisele, F. L., Tanner, D. J. and Jefferson, A. 1997. Measurements of new particle formation and ultrafine particle growth rates at a clean continental site. *J. Geophys. Res.* **102**, 4375–4385.

Weber, R. J., McMurry, P. H., Mauldin, R. L., Tanner, D. J., Eisele, F. L., Clarke, A. D. and Kapustin, V. N. 1999. New particle formation in the remote troposphere: A comparison of observations at various sites. *Geophys. Res. Letters* **26**, 307–310.