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Seasonal variation of CCN concentrations and aerosol activation properties in boreal forest

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ACPD

10, 28231–28272, 2010

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

As a part of EUCAARI activities, the annual cycle of cloud condensation nuclei (CCN) concentrations and critical diameter for cloud droplet activation as a function of supersaturation were measured using a CCN counter and a HTDMA (hygroscopic tandem DMA) at SMEAR II station, Hyytiälä, Finland. The critical diameters for cloud droplet activation were estimated from (i) the CCN concentration and particle size distribution data, and (ii) the hygroscopic growth factors by applying κ -Köhler theory. The critical diameters derived by these two methods were in good agreement with each other. The effect of new particle formation on the diurnal variation of CCN concentration and critical diameters was studied. New particle formation was observed to increase the CCN concentrations by 70–110%, depending on the supersaturation level. The average value for the κ -parameter determined from hygroscopicity measurements was $\kappa = 0.18$ and it predicted well the CCN activation in Hyytiälä boreal forest conditions. The derived critical diameters and κ -parameter indicate that aerosol particles at CCN sizes in Hyytiälä are mostly organic, but contain also more soluble, probably inorganic salts like ammonium sulphate, making the particles more CCN active than pure secondary organic aerosol.

1 Introduction

Atmospheric aerosol particles influence clouds, and thereby climate, by altering the albedo, lifetime and precipitation patterns of clouds (Twomey, 1974; Lohmann and Feichter, 2005; Rosenfeld et al., 2008; Stevens and Feingold, 2009). While aerosol-cloud interactions depend on a number of environmental factors, the primary aerosol-related quantity in this respect is the cloud condensation nuclei (CCN) concentration or, more specifically, the CCN supersaturation spectrum (Andreae and Rosenfeld, 2008).

During the last decade or so, CCN number concentrations and spectra have been measured in a variety of different environments (Snider and Brenguier, 2000; Bigg

ACPD

10, 28231–28272, 2010

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



and Leck, 2001; Hudson and Yum, 2002; Roberts et al., 2006; Hudson and Noble, 2009). Another class of investigations, that has gained a large interest, is so-called CCN closure studies (Covert et al., 1998; Rissler et al., 2004; Ervens et al., 2007; Medina et al., 2007; Duplissy et al., 2008; Good et al., 2010; Poulain et al., 2010). In closure studies, the measured CCN concentrations are compared with those obtained from equilibrium model calculations based on measured aerosol properties, such as the aerosol size distribution and chemical composition or hygroscopicity.

Determining the relative role of humans and nature in aerosol-cloud interaction requires identifying and quantifying different CCN sources. In addition to primary natural and anthropogenic sources, increasing empirical evidence has been obtained on secondary CCN production in the atmosphere via nucleation and growth of new aerosol particles (Lihavainen et al., 2003; Laaksonen et al., 2005; Kuwata et al., 2008; Kuang et al., 2009; Wiedensohler et al., 2009). Recent model studies indicate that secondary production may be an important CCN source in the global atmosphere (Spracklen et al., 2008a; Makkonen et al., 2009; Wang and Penner, 2009), even though uncertainties in the overall magnitude of this source are still quite large (Pierce and Adams, 2009).

Boreal forests constitute one of the largest and most active areas of atmospheric new particle formation (Kulmala et al., 2001; Tunved et al., 2006; Dal Maso et al., 2007, 2008). While the potential of particles nucleated in boreal forests to act as CCN has been identified already some time ago (Lihavainen et al., 2003; Kerminen et al., 2005), direct CCN measurements in this environment have been extremely rare (Aalto and Kulmala, 2000). Such measurements would be of great help when studying aerosol-cloud interactions over boreal forests and, especially, when attempting to narrow down the uncertainties in indirect radiative effects due to boreal forest aerosols (Kurtén et al., 2003; Spracklen et al., 2008b; Lihavainen et al., 2009).

In this paper, we present an analysis of simultaneous measurements of CCN concentrations, aerosol number size distributions and hygroscopic properties at a boreal forest site. We aim to shed light on the following questions: (i) what is the seasonal variation of CCN concentrations and related aerosol activation quantities, (ii) how CCN

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



concentrations are affected by atmospheric new particle formation events, and (iii) can we predict the CCN activity of boreal forest aerosols based on measured aerosol size distributions and hygroscopic properties. Our analysis relies on one year of continuous measurements conducted at the SMEAR II station in Hyytiälä, Finland.

2 Experimental

The SMEAR II (Station for Measuring Forest Ecosystem-Atmosphere Relations) station is located in a rather homogenous Scots pine (*Pinus sylvestris* L.) stand on a flat terrain at Hyytiälä Forestry Field Station (61°51 N, 24°17 E, 181 m.a.s.l.) of the University of Helsinki, 220 km north-west from Helsinki. 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 south-west from the measurement site with about 200 000 inhabitants. Kulmala et al. (2001) and Hari and Kulmala (2005) have described the station and its operation in detail.

2.1 Cloud condensation nuclei (CCN) concentrations

Cloud condensation nuclei concentrations (N_{CCN}) were measured with a commercial diffusion-type CCN counter (CCNC, Droplet Measurements Technologies Inc.; Roberts and Nenes, 2005). The CCNC consists of a saturator unit which mimics the supersaturated conditions inside a cloud. The sample flow, surrounded by a sheath flow (1/10 flow ratio), is led through the saturator unit, which is a vertical column with walls wetted by water. Inner walls of the column are covered with filter paper in order to maintain constant wetting. The sheath flow is humidified by using Nafion-humidifier. The column temperature increases along the flow, generating a stable supersaturation along the centre-line. Particles that have a critical saturation ratio smaller than the peak saturation ratio will activate and grow to droplets by condensation. After the saturator, the droplets are counted by an optical particle counter (OPC). The OPC is heated to a temperature 2°C higher than the bottom column temperature in order to avoid

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



condensation of water vapour to the optics. The concentration of cloud condensation nuclei is thus obtained as a function of the water supersaturation ($SS = S - 1$, where S is saturation ratio). Supersaturations of 0.1, 0.2, 0.4, 0.6 and 1.0% were used by selecting the correct temperature difference between the upper and bottom part of the saturator column.

The CCN counter was calibrated with ammonium sulphate particles, which were produced using an atomizer (TOPAS ATM 220). The size of the particle was selected with a differential mobility analyzer (DMA). A condensation particle counter (CPC, model TSI 3772) was used to count all the particles (N_{CN}) entering the CCNC. Calibrations were performed by keeping the temperature difference constant and varying the size of the ammonium sulphate particles entering the CCNC. The CCNC concentration was compared to the CPC concentration in order to define the critical diameter ($N_{CCN}/N_{CN} = 50\%$) for each temperature difference, i.e. for each supersaturation. From this data the peak saturation ratio inside the column can be calculated by using Köhler theory.

2.2 Hygroscopicity measurements

A Hygroscopic Tandem Differential Mobility Analyzer (HTDMA, Liu et al., 1978; Ehn et al., 2007) was used to measure the hygroscopicity of aerosol particles. In this instrument, the sample aerosol is first brought through a drier and a radioactive charger in order to reach a charge equilibrium and a relative humidity below 20%. After this, a certain dry particle size, d_{dry} , is selected using a differential mobility analyzer (DMA). Next the particles are passed through a humidifier so that a certain controlled relative humidity is reached. Finally the humidified aerosol passes through another DMA which is used as a DMPS. Thus a humidified size distribution for a certain d_{dry} is measured. The second DMA was located in a temperature controlled box, maintained at a temperature of 19 °C and relative humidity of 90%. Four dry sizes (110 nm, 75 nm, 50 nm and 35 nm), which belong to the standard sizes agreed inside the EUCAARI project (Kulmala et al., 2009), were used.

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2.3 Aerosol number size distribution

The total aerosol particle size distribution was measured using a differential mobility particle sizer (DMPS) in size range from 3 nm to 1 µm (Aalto et al., 2001). The aerosol sample is first dried to a RH < 30%, after which the particles are charged using a radioactive bipolar charger. Then the particles are classified according to their (mobility) size using two separate DMAs for different size ranges (Twin-DMPS). The particle concentration after the DMA, corresponding to a certain particle size range, is measured with condensation particle counters (CPC, TSI 3025 for small particles and TSI 3010 for bigger particles).

3 Theoretical framework

3.1 CCN activation

The activation of an aerosol particle into a cloud droplet is described by the Köhler equation (Köhler, 1936):

$$S = \gamma_w x_w \exp\left(\frac{4M_w \sigma}{RT \rho d_p}\right), \quad (1)$$

where S is the water saturation ratio, γ_w is the activity coefficient, x_w is the mole fraction of water, M_w is the molar mass of water, σ is the surface tension of the solution, R is the universal gas constant, T is temperature, ρ is the density of the solution and d_p is the wet particle diameter. This equation connects the particle properties (size, surface tension, density and water activity) with the thermodynamic state of the surroundings (temperature, saturation ratio). Usually Köhler equation is presented in the following form (Seinfeld and Pandis, 1998, e.g.):

$$\ln(S) = \frac{A}{d_p} - \frac{B}{d_p^3 - d_{\text{insol}}^3}, \quad A = \frac{4M_w \sigma_w}{RT \rho_w}, \quad B = \frac{6n_s M_w}{\pi \rho_w}, \quad (2)$$

28236

ACPD

10, 28231–28272, 2010

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



where σ_w is the surface tension and ρ_w the density of water, and n_s is the number of moles of the soluble material (= solute). This equation takes also into account the (possible) insoluble core of diameter d_{insol} inside the aerosol particle.

For a particle of a certain composition and surrounding conditions, the equilibrium saturation ratio, calculated according to Köhler equation (Eq. 1), has its maximum at the critical particle size d_c . Particles having larger diameter than this are referred as being activated and can become cloud droplets by condensational growth by water vapour. The critical saturation ratio (S_c) and the corresponding critical (wet) diameter (d_c , the maximum point of the Köhler curve) are given by:

$$\ln(S_c) = \sqrt{\frac{4A^3}{27B}}, \quad d_c = \sqrt{\frac{3B}{A}}. \quad (3)$$

To find out the dependence of S_c on the properties of the dry particle, we expand B (Eq. 2; A does not depend on the properties of aerosol particle) and calculate the number of moles of solute in the droplet:

$$n_s = v_s \frac{m_s}{M_s} = v_s \frac{\rho_s \varepsilon_s V_{\text{dry}}}{M_s} = v_s \frac{\rho_s \varepsilon_s \frac{\pi}{6} d_{\text{dry}}^3}{M_s} = v_s \frac{\rho_s \varepsilon_s \pi d_{\text{dry}}^3}{6M_s}, \quad (4)$$

where m_s and ρ_s are the (dry) mass and density of the solute material, M_s is its molar mass, v_s is the number of ions per one solute molecule in water solution ($v_s = 3$ for $(\text{NH}_4)_2\text{SO}_4$), and V_{dry} and d_{dry} are the volume and diameter of the dry particle containing the volume fraction ε_s of soluble material. Combining Eqs. (2, 3, and 4), we get for the critical saturation ratio (Seinfeld and Pandis, 1998, e.g.):

$$\ln(S_c) = \sqrt{\frac{4A^3}{27}} \sqrt{\frac{\rho_w M_s}{v_s \rho_s M_w \varepsilon_s d_{\text{dry}}^3}} \propto d_{\text{dry}}^{-3/2}. \quad (5)$$

This equation describes the connection between the critical saturation ratio and the dry particle size. Thus, at a given saturation ratio S_c , d_{dry} is the smallest dry particle

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



size that gets activated. In this paper, we call this diameter the critical diameter and denote it by d_{crit} . Note, that this diameter is different than the point corresponding to the maximum of the Köhler curve, which is given by Eq. 3 (wet diameter d_c).

The total CCN concentrations (N_{CCN}) were compared to the aerosol size distribution data obtained from the DMPS measurements in order to estimate the activated fraction and the critical diameter from the ambient data. The activated fraction (F_{act}) for a certain supersaturation is defined here as:

$$F_{\text{act}} = \frac{N_{\text{CCN}}}{N_{\text{CN}}}, \quad (6)$$

where N_{CN} is the total concentration of the condensation nuclei obtained from the DMPS data.

The DMPS data can also be used to estimate the critical particle diameter for all the supersaturations used. In order to obtain this, the activation probability as a function of the particle size is assumed to behave like a step function, with all particles above the critical size (d_{crit}) activating. The particle size distribution is summed from the largest particle size towards the smaller sizes until the measured value of the CCN concentration is obtained. At that size the diameter corresponds to the critical diameter:

$$\sum_{d_p=d_{\text{crit}}}^{d_{\text{max}}} N_i = N_{\text{CCN}}. \quad (7)$$

This method includes the assumption that the aerosol is completely internally mixed. The aerosol size distribution was measured by using 38 size bins from 3 to 1000 nm (Aalto et al., 2001). In order to increase the resolution of the critical diameter estimate, a linear interpolation was used between each size bin of the DMPS.

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3.2 HTDMA methods

In the simplest form the hygroscopicity of a certain sized dry particle can be described as a growth factor:

$$g_a = \frac{d_{\text{wet}}}{d_{\text{dry}}}, \quad (8)$$

- 5 where d_{wet} is a mean diameter of the humidified size distribution for a certain dry particle size d_{dry} .

The critical particle diameter for CCN activation at certain supersaturation can be calculated also from the HTDMA data using the so called κ -Köhler equation (Petters and Kreidenweis, 2007) (see Eq. 1 for the notation):

$$10 \quad S(d_{\text{wet}}) = \frac{d_{\text{wet}}^3 - d_{\text{dry}}^3}{d_{\text{wet}}^3 - d_{\text{dry}}^3(1 - \kappa)} \exp\left(\frac{4M_w\sigma_w}{RT\rho_w d_{\text{wet}}}\right). \quad (9)$$

- In this theory the Köhler equation is modified so that all the physico-chemical properties of the aerosol are captured into a single κ -parameter (κ) while all other parameters are kept constant. Equation 9 can be derived from Eq. 1 by using Zdanovskii, Stokes and Robinson (ZSR) assumption for multicomponent-water solution at equilibrium, volume equivalent diameters (spherical particles) and the following parametrization for the water activity of the solution:

$$15 \quad \frac{1}{a_w} = 1 + \kappa \frac{V_{\text{dry}}}{V_w}. \quad (10)$$

- In the previous equation $a_w = \gamma_w x_w$ is the water activity, V_{dry} is the volume of the dry particulate matter and V_w is the volume of water in the solution ($V_{\text{tot}} = V_{\text{dry}} + V_w$). For HTDMA data Eq. 9 can be rearranged to a more convenient form by expressing the wet diameter with the growth factor using Eq. 8:

$$20 \quad S = \frac{g_a^3 - 1}{g_a^3 - (1 - \kappa)} \exp\left(\frac{A}{d_{\text{dry}} g_a}\right), \quad A = \frac{4M_w\sigma_w}{RT\rho_w}. \quad (11)$$

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The κ -values were obtained by solving the Eq. 11 for κ and applying the saturation ratio (now $S < 1$), dry size, growth factor and temperature of the HTDMA measurement. The critical particle diameter (d_{crit}) with a certain discrete kappa can be determined by searching the dry size (d_{dry}) which produces the maximum value of κ -Köhler curve (Eq. 11) at the point of activation equal to a certain supersaturation. The critical dry diameters were determined for the supersaturations used in the CCN measurements, i.e. 0.1, 0.2, 0.4, 0.6 and 1.0%. For consistency the calculations were done using the surface tension of water, $\sigma_w = 0.072 \text{ J m}^{-2}$, as suggested by Petters and Kreidenweis (2007).

4 Results

4.1 The measured CCN concentrations

The time series of CCN concentrations for the analyzed one-year period (July 2008–June 2009) is presented in Fig. 1. The CCN concentrations show high variability from day to day, reflecting the variations in aerosol number concentration, size distribution and chemical composition. From this figure no clear seasonal pattern in the CCN concentration can be observed. Mostly the CCN concentrations seem to be lower on new particle formation event days (marked with a grey background) as compared to other days (this issue will be returned to later in Sect. 4.3.2). When comparing the time series for different supersaturations with each other, there appears to be larger difference between the different supersaturations in spring-summertime (May–August) than in wintertime. This might be related to differences in the aerosol chemical composition: in summertime the aerosol in Hyytiälä is mainly secondary organic aerosol (SOA), whereas in winter we expect to have lower contribution of SOA from the forest and more anthropogenic influence.

The seasonal variation in CCN concentration becomes clearer when looking at monthly averages (Table 1 and Fig. 2). CCN concentrations are highest in the

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



summertime and lowest in wintertime, with minimum values occurring in November–January. Comparison with the total particle concentration (CN, condensation nuclei) shows that the seasonal variation of CCN concentration is not only due to variation in the CN concentration (see Fig. 2a), but there are also differences in the size distribution and chemical composition, both of which contribute to the variation of CCN concentration. This is reflected in the activated fraction (CCN/CN, Fig. 2b), which has somewhat different annual profile than the CCN concentrations. For instance, the minimum is shifted to October–November. The activated fraction at the highest supersaturations (0.4, 0.6 and 1.0%) shows quite a large variation over the year, with a maximum in July. At those supersaturations, the activated CCN contain more (small) particles produced by nucleation events than with lower supersaturations.

We also examined the mean CCN concentration as a function of supersaturation, and determined parameters for the so called “CCN spectrum”, i.e. the parameterisation

$$[\text{CCN}] = c\text{SS}^k, \quad (12)$$

where SS is supersaturation in percents. The values for the parameters c and k from this data set were $c = (1100 \pm 220) \text{ cm}^{-3}$ and $k = (0.6 \pm 0.2)$ (for mean [CCN]; for median [CCN] $c = 990 \text{ cm}^{-3}$ and $k = 0.7$). These are within the range of values reported in literature for continental aerosol ($c = 600\text{--}3500 \text{ cm}^{-3}$ and $k = 0.4\text{--}0.9$; e.g. Seinfeld and Pandis, 1998).

In January–February (21 January–10 February 2009) there was a break in the measurements due to some changes made for the measurement setup (a DMA was added in front of the CCNC to enable size-resolved measurements in the future), and therefore data for this period is missing (see Fig. 1). The concentrations stayed at similar level after the break, so we can assume that the observations before and after the break are comparable to each other.

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


4.2 The critical diameter for CCN activation

4.2.1 The critical diameter estimated from CCN and size distribution measurements

The critical diameter for cloud droplet activation was estimated from the CCN concentrations and particle size distributions measured with the DMPS (see Sect. 3.1). In principle, the variations in critical diameter reflect the variations in chemical composition of the aerosol: the more hygroscopic the aerosol, the smaller the critical diameter for activation.

Figure 3 shows the time series of the estimated critical diameters for the five supersaturations inside the CCN counter. The critical diameter varies from ~ 50 nm at a supersaturation of 1.0% to ~ 180 nm at a supersaturation of 0.1%. Day-to-day variability in the critical diameter is quite high; note that the linear scale in the y-axis hides the variations for small critical diameters (i.e. high supersaturations).

Similarly as the CCN concentration, the critical diameter has larger variation between different supersaturation levels in summertime than in wintertime. Especially d_{crit} for the lowest supersaturation ($SS = 0.1\%$) decreases in winter (from December to March), and starts to increase again towards summer. Similarly as with the CCN concentration, Fig. 3 does not reveal any clear differences in the critical diameter between new particle formation days and other days.

Monthly mean values show that the critical diameter is smallest in April and largest in November for supersaturations $SS > 0.1\%$ (Table 1 and Fig. 4). The maximum of d_{crit} in November corresponds to the minimum of CCN concentration and activated fraction (see also Fig. 2). For $SS = 0.1\%$ the seasonal behaviour of d_{crit} is clearly different than for other supersaturations.

When calculating the critical diameters from CCNC and DMPS data, a uniform chemical composition was assumed. It must be noted that this assumption may not be justified, if the aerosol population is highly externally mixed, having for example a big fraction of primary soot particles. However, with this method we can get an estimate

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



for the activation diameter with a high time resolution, and investigate the general characteristics of its variation.

4.2.2 Comparison of d_{crit} from the CCN concentration and hygroscopicity measurements

5 For comparison, we determined the critical diameter values also from hygroscopicity measurements by applying κ -Köhler theory. HTDMA-data was available only for shorter periods between July 2008 and June 2009, and therefore we do not present the time series for it, but examine only the average values over a longer period. The monthly averages of the critical diameter at supersaturation of 0.4% are listed
10 in Table 1. The values from HTDMA data differed to some extent from the values estimated from the CCN concentrations, but no systematic difference was observed. The seasonal behaviour was fairly similar for both critical diameter estimates, with smallest values occurring in March–April. One reason for the differences between the two critical diameter estimates are the gaps in HTDMA data, making the monthly averages not
15 truly comparable with each other. Therefore, in the following we consider the one-year averages of the critical diameters.

Figure 5 compares the average critical diameters over the one-year data set estimated from the CCN concentrations and hygroscopic measurements. The critical diameter decreases linearly with supersaturation (with logarithmic scale in both axes),
20 with a slope that is close to the theoretical value $-2/3$ given by the Köhler equation (see Eq. 5; approximately $d_{\text{crit}} \sim \text{SS}^{-2/3}$). The values determined by these two methods are very close to each other. This gives confidence that our simple method to determine the critical diameter by integrating the size distribution seems to work reasonably well; and vice versa that the κ -Köhler model works for the Hyytiälä aerosol population. The
25 variation in critical diameters, as shown by the error bars, is slightly bigger for the CCNC data than for the HTDMA data.

Figure 5 also shows the data points for pure secondary organic aerosol (SOA) from the oxidation of α -pinene and trimethylbenzene (TMB) in laboratory measurements

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



by Duplissy et al. (2008). The critical diameters in Hyytiälä are somewhat smaller than the laboratory measurements for the SOA, but clearly bigger than those for pure ammonium sulphate. Aerosol particles at Hyytiälä measurement station, located in a pine forest in boreal forest region, are expected to have a high organic fraction, with α -pinene and other monoterpenes being the most important VOCs emitted by coniferous trees (Hakola et al., 2003; Tarvainen et al., 2005). Our measurements and the derived critical diameters indicate that the aerosol particles in Hyytiälä are not purely organic, but consist also of more soluble compounds, such as ammonium sulphate.

Besides the measured data points, Fig. 5 shows a line from the κ -Köhler equation (Eq. 11) using the average κ -value determined from the HTDMA-data, $\kappa = 0.18$. Also the lines with κ -values reported earlier from laboratory studies of monoterpene SOA ($\kappa = 0.04 \dots 0.24$, e.g. Engelhart et al., 2008) are plotted. Figure 5 shows that the κ -Köhler theory with $\kappa = 0.18$ is on average a good model for cloud droplet activation in Hyytiälä.

Recently, Gunthe et al. (2009) reported a value $\kappa = 0.1$ for pure secondary organic aerosol in Amazonian tropical rainforest, and Dusek et al. (2010) found the same value for SOA at a rural Central-European site. The value $\kappa_{\text{org}} = 0.1$ is supported also by laboratory studies of SOA from α - and β -pinene (Petters and Kreidenweis, 2007; Duplissy et al., 2008; Jurányi et al., 2009). The average κ -value $\kappa = 0.18$ obtained in this study is somewhat bigger, and it takes into account the average deviation of aerosol particles in Hyytiälä from pure organic aerosol. The effective κ -parameter can be calculated as a linear superposition from the aerosol constituents, for which κ -values are known:

$$\kappa = f_{\text{org}}\kappa_{\text{org}} + f_{\text{inorg}}\kappa_{\text{inorg}} \quad (13)$$

where κ_{org} and κ_{inorg} are kappa-values of the organic and inorganic part, and f_{org} and f_{inorg} their volume fractions (Gunthe et al., 2009).

If we assume $\kappa_{\text{org}} = 0.1$ for the organic part and $\kappa_{\text{inorg}} = 0.6$ for the inorganic part ($\kappa = 0.6$ for ammonium sulphate; Petters and Kreidenweis, 2007), we get the following fractions for organic and inorganic part in Hyytiälä aerosol: $f_{\text{org}} = 84\%$ and $f_{\text{inorg}} = 16\%$.

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



These fractions, although having some uncertainty, tell that the aerosol in Hyytiälä is largely organic, but includes also more hygroscopic (non-organic) compounds. The organic fraction predicted here from the kappa parameter (84%) corresponds well to the estimates from growth rate analyses, which predict the organic contribution to particle growth rate to be on average about 90% in Hyytiälä (Boy et al., 2005). On the other hand, our estimate is somewhat higher than the organic mass fraction of ~70% observed in aerosol mass spectrometer (AMS) studies in Hyytiälä (Jimenez et al., 2009). One reason for the discrepancy between the organic fraction estimates could be that κ -parameter deals with volume fractions whereas AMS measurement gives the mass fraction. Another reason might be that there are also organic compounds with $\kappa_{\text{org}} < 0.1$ present in Hyytiälä aerosol population.

In this data set we did not observe a systematic difference between d_{crit} estimated from CCNC and HTDMA-measurements (although some non-systematic differences were seen), which would result from a non-linear hygroscopicity behaviour at RH close to 100%, as reported by some studies (Petters and Kreidenweis, 2007; Wex et al., 2009; Good et al., 2010). In those studies, HTDMA data yielded bigger d_{crit} values (or smaller κ -values) than CCN activity measurements. This discrepancy has been explained by the fact that when applying the kappa-Köhler theory using hygroscopicity measurements, the CCN activation is “extrapolated” from the measurement in subsaturated conditions, at about RH = 90–98%. The hygroscopicity may increase at relative humidities close to 100%, which would cause a systematic error to d_{crit} (and κ) values estimated from the hygroscopicity data. However, in this study this kind of effect was not seen; instead, at least on average, both methods gave similar results.

CCN concentrations have been measured previously in Hyytiälä by Aalto and Kulmala (2000), but with a different instrument and measurement setup. In addition, the data analysis was performed in a different way concentrating mainly on estimating the soluble fraction from the CCNC data. Due to differences in the instrument and data analysis, the results of this and previous study are not directly comparable with each other. Nevertheless, the critical diameter values obtained here are broadly consistent

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



with the measurements by Aalto and Kulmala (2000), who reported an approximate activation diameter of 70 nm at supersaturation of 0.5% for spring–summer 1998 in Hyytiälä.

4.3 Effect of new particle formation to CCN population

5 New particle formation (NPF) is an important source of particles in atmosphere, and some fraction of the formed particles reaches a size where they can act as CCN. In this section we study the effect of new particle formation on CCN properties in Hyytiälä by comparing the CCN concentrations, activated fractions and critical diameters on new particle formation days (event days) and on days with no particle formation (non-event
10 days) classified according to Dal Maso et al. (2005). During the analysed one-year period (July 20008–June 2009) 75 new particle formation events were observed.

4.3.1 A case study of CCN activation on new particle formation days

An example of a new particle formation period with four consecutive particle formation events is shown in Fig. 6. New particle formation occurs typically around midday
15 following the sunlight, and the growth of the nucleation mode particles continues until midnight and even during the next day. During the period presented in Fig. 6, both new particle formation events and changes in background aerosol created interesting variations in the CCN concentration, activated fraction and critical diameter.

Each new particle formation event shows as a rapid increase in total particle concentration (CN, black curve in Fig. 6). After the particle formation burst, the total concentration decreases due to coagulation. Meanwhile, the CCN concentration shows less rapid changes and increases steadily during the first three event days. Because the background aerosol concentration was rather stable during this period, the increase in CCN concentration can be attributed to the particles produced by the NPF events. The
20 continuous increase in CCN concentration was observed for all the supersaturations, but the increase was greatest for $SS = 1.0\%$ (almost one order of magnitude from 100
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Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



to 1000 cm^{-3}), because at higher supersaturation a bigger fraction of nucleation mode particles gets activated. Before the fourth event (on 23 April 2009) the background concentration dropped dramatically, which led to a rapid decrease in CCN concentration at $SS = 0.1\text{--}0.6\%$, although the total number concentration increased because of high number of newly formed particles. The particles from the NPF event contribute to the increase of CCN concentration at the supersaturation of 1.0% (which includes particles above $\sim 40\text{ nm}$).

Because of quite large variations in the total particle concentration, the activated fraction varies a lot even inside one day. Typically during a NPF event, the activated fraction is very low due to a high number of small particles produced. During the growth of the nucleation mode (especially for the two strong events on 21.4. and 22.4.), the activated fraction increased greatly to 0.5 and above (for $SS = 1.0\%$).

The effect of the background aerosol is seen also on the first measurement day (20 April), when there was a clear change of airmass. The new air mass was very clean and had very low particle concentration. However, despite the decrease of particle concentration in all particle sizes, the CCN concentrations increased and the activated fraction rose to close to 0.5 during this period. This indicates that aerosol was much more water soluble than typically in Hyytiälä, as can be seen also from the critical diameter, which decreased to as low values as $20\text{--}40\text{ nm}$. Based on the observed wind direction (about 330°), this was a northern marine airmass, which are typically very clean and contain sea salt, making the particles more hygroscopic than in continental airmasses.

The critical diameters stayed quite constant and close to their mean values during the whole new particle formation period (21 April–24 April). A more careful inspection reveals that for the two highest supersaturations (0.6 and 1.0%) the critical diameter decreased slowly and steadily from 21 April to 23 April, meaning that the particles around $30\text{--}60\text{ nm}$ became more hygroscopic in the course of time. This particle range contains a big fraction of recently nucleated particles. The increase in hygroscopicity is probably due to aging of the secondary organic aerosol, as reported by Jimenez et al. (2009).

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



For the two smallest supersaturations (0.1 and 0.2%) the critical diameter slowly increased from 21 April to 24 April. This particle size range, 90–180 nm, consists mainly of long-range transported particles or primary biogenic particles.

4.3.2 The diurnal profiles of CCN-properties on event and non-event days

5 To further investigate the effect of new particle formation on CCN-properties, we calculated mean diurnal profiles of CCN quantities on event and non-event days over the whole one-year data set. The diurnal variation of CCN concentration for two consecutive days, at five different supersaturation levels is presented in Fig. 7. Figures 8 and 9 show the profiles for activated fraction and critical diameter, respectively. A 48-h period was chosen, because particles produced by NPF events continue growing on the following day and some fraction of them reach CCN-sizes only on the next day after the event (typical growth rates in Hyytiälä are 2–8 nm/h, e.g. Manninen et al., 2009). The classification of the 48 h period as an event period or a non-event period was done based on whether the first day (first 24 h in the figure) was an event or a non-event day, and paying no attention to the classification of the following day (last 24 h in the figure).

CCN concentration

Before noon the CCN concentrations were observed to be lower on NPF event days than on non-event days for all supersaturations except for $SS = 1.0\%$ (see Fig. ??A1). This is explained by the fact that NPF events typically occur on days with low background aerosol concentration (leading to small coagulation and condensation sink) before the event (Hyvönen et al., 2005; Dal Maso et al., 2007), which also leads to a low number of particles activated as CCN.

On days with no particle formation, the diurnal profile of CCN concentration is rather stable. On the other hand, on new particle formation days big variations are observed: the CCN concentration has a small minimum around noon, and after the NPF event

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



(which occurs around noon) the concentrations increase towards the evening and stay approximately at the same level on the following day. The increase in CCN is most pronounced and occurs first for the highest supersaturation ($SS = 1.0\%$), whereafter the increase can be seen consecutively at 0.6% , 0.4% and 0.2% supersaturation levels. The increase in CCN concentration on event day evenings can be attributed to particles produced by NPF events. The increase is observed first at the highest supersaturation, corresponding to the activation of particles above ~ 50 nm, the size range which nucleated particles reach in the afternoon. While the particles grow bigger, they are seen at smaller and smaller supersaturations, corresponding to larger activation size. For the two lowest supersaturations (0.1% and 0.2%), the CCN concentration continues to increase on the day after the event, as it takes more time for particles to grow to the corresponding sizes (110 – 180 nm).

In percentages, the CCN concentration increases on event days by about 70% from event day midday to next day's midnight at supersaturation of 1.0% , and by 82% , 70% , 110% and 106% at supersaturation levels 0.6% , 0.4% , 0.2% and 0.1% , respectively (for absolute changes see Fig. 7). At the two smallest supersaturations (0.2% and 0.1%) the increase in CCN concentration is smaller in absolute numbers, but percentually the increase is greater (upto 110% by the end of the two day period). Thus, new particle formation considerably increases the CCN number on the following day, the increase being even a factor of two from the concentration before the event.

Activated fraction

The activated fraction (CCN/CN) was observed to be lower on event days than on non-event days (see Fig. 8). Naturally, the activated fraction follows a similar form as the CCN concentration (Fig. 7): the profiles are flat on non-event days, whereas on event days there is a minimum in the afternoon after which the activated fraction increases towards the next day. However, during the 48 h period, the activated fraction on event days always stays lower than on non-event days. This is mainly because

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the total particle number concentration (CN, condensation nuclei) is higher on event days than on non-event days (see Fig. A1 in Appendix). A high CN further makes the activated fraction low on event days, despite the substantial increase in CCN due to particle formation (as shown in Fig. 7). At first sight it is surprising that also before noon the total particle concentration is higher on event days than on non-event days, as events should occur on days with low background aerosol concentration. However, for event occurrence it is the background aerosol surface area (i.e. the condensation and coagulation sinks) that is crucial, and for that the number of bigger particles has more influence (as seen also in Fig. 7, where the difference between event day and non-event day mornings disappears for the highest supersaturation). The high CN concentration in the morning of an event day is explained by the fact that during the “event-rich period” from spring to autumn the NPF events often occur on consecutive days. This makes the total particle concentration high also in the morning of an event day, when the tail of the nucleated particle population is often observed.

In summary, the difference in the activated fraction between event and non-event days results mainly from the differences in the shape of the particle size distribution, event days having a bigger number of small particles and, before the event, less big particles. Also the differences in the particle chemical composition between event and non-event days may have some effect on the CCN concentrations, but the effect of size distribution is expected to dominate, as reported by Dusek et al. (2006).

Critical diameter

The diurnal profile of the critical diameter on event and non-event days is presented in Fig. 9. On average, the critical diameter is somewhat lower on event days than on non-event days, but the difference is quite small, only a few nanometers. On both event and non-event days, there is a diurnal variation in critical diameter with a minimum around 03:00 p.m. in the afternoon. The variation seems to be more pronounced on event days, especially at lower supersaturations. In principle, the variations in critical

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



diameter should be related to the chemical composition (hygroscopicity or water activity) of the particles. The diurnal variation of d_{crit} is opposite to the variation of solar radiation, and it could be related to photo-chemistry (mainly through oxidation by OH radical). During daytime the oxidation products of VOCs condense on the particles and there might be chemical reactions on particle surfaces, which could change the hygroscopicity of the particles in the course of the day.

A rather small difference in d_{crit} between event and non-event days indicates that there is no big difference in the particle chemical composition on event and non-event days. However, it should be noted that in Fig. 9 the points are averaged over one year, during which the critical diameter shows some seasonal variation (see Fig. 4). This averaging may hide some differences in aerosol composition between event and non-event days.

5 Conclusions

As a part of EUCAARI (Kulmala et al., 2009) activities, the CCN properties at Hyytiälä SMEAR II station were investigated by analyzing one year data of CCN concentrations and hygroscopicity measurements. The critical diameters for cloud droplet activation were estimated using two methods: from the CCN concentrations by integrating the particle size distribution, and from the hygroscopic growth factors by applying the κ -Köhler theory. Both methods gave results very close to each other. No systematic difference between the values from hygroscopicity and CCN measurements was observed, as reported in some earlier studies (Petters and Kreidenweis, 2007). This means that our method to calculate the critical diameter by simply integrating the size distribution, without any knowledge of the chemical composition, seems to work reasonably well in Hyytiälä conditions. The critical diameter values obtained here were broadly consistent with the earlier measurements by Aalto and Kulmala (2000).

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The CCN number concentrations showed a large seasonal variation, with the concentrations being more than a factor of two higher in summer than in winter. The critical diameter had a maximum in November and a minimum in April. The seasonal variation was similar for supersaturations 0.2–1.0%, but clearly different for the supersaturation of 0.1%. This is related to the fact that the CCN measurements at supersaturations 0.2–1.0% (corresponding to critical diameters from 120 to 50 nm) are affected by new particle formation, whereas the measurements at the supersaturation of 0.1% (critical diameter ~180 nm) reflect mainly the long-range transported background aerosol.

New particle formation was observed to increase the CCN concentrations in the evening of a new particle formation day. The CCN concentrations increased on average by 70–110% from event day midday to next day midnight, depending on the supersaturation level. The activated fraction was smaller on event days than on non-event days, and showed only small increase towards the evening on event days, due to high number of small particles produced by nucleation. For the critical diameter, no significant difference between event and non-event days was observed. On both event and non-event days the critical diameter had a small sinusoidal diurnal variation with a minimum in the afternoon, probably related to OH-driven chemistry.

Comparison with the laboratory measurements by Duplissy et al. (2008) showed that the critical diameter in Hyytiälä is somewhat smaller than that for pure secondary organic aerosol from α -pinene or trimethylbenzene. The average value for κ -parameter, determined from the hygroscopicity measurements, was $\kappa = 0.18$. Both the critical diameters and the κ -parameter value indicate that the aerosol in Hyytiälä is slightly more hygroscopic than pure secondary organic aerosol.

Our value for the κ -parameter corresponds well to the values reported earlier: $\kappa = 0.15$ has been reported by Engelhart et al. (2008) for monoterpene SOA produced in a smog chamber and by Gunthe et al. (2009) for a measurement campaign in Amazonian rainforest. By extrapolating the data, a value $\kappa_{\text{org}} = 0.1$ for pure organic aerosol has been proposed (Gunthe et al., 2009; Dusek et al., 2010). The value of κ -parameter in Hyytiälä, $\kappa = 0.18$, indicates that aerosol is mostly organic, but includes also more

soluble salts, e.g. ammonium sulphate. By assuming $\kappa_{\text{org}} = 0.1$ for organic part and $\kappa_{\text{inorg}} = 0.6$ for inorganic part, we estimated the average organic fraction to be 84% and inorganic fraction 16% for the aerosol in Hyytiälä. This organic fraction is close to the organic contribution of $\sim 90\%$ estimated from the particle growth rates (Boy et al., 2005), but somewhat higher than the value of $\sim 70\%$ obtained from aerosol mass spectrometer studies in Hyytiälä (Jimenez et al., 2009).

Our measurements give several implications for modelling CCN activation in boreal forest areas in global models. The seasonal variation in critical diameter is significant and should be taken into account in the models. New particle formation has a significant effect on the CCN concentration and should be taken into account for accurate prediction of CCN concentrations. The κ -Köhler theory with κ -parameter $\kappa = 0.18$ and a typical growth factor for secondary organic aerosol was attested to be a good model for CCN activation in Hyytiälä boreal forest conditions.

In future, a more detailed analysis applying data of the particle chemical composition would be needed to better understand the role of chemical composition on CCN activation in boreal forest conditions. Also, analyses of continuous multi-year datasets are needed to more reliably distinguish the seasonal patterns and interannual variation of CCN properties.

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Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



References

- Aalto, P. and Kulmala, M.: Using a cloud condensation nuclei counter to study CCN properties and concentrations, *Boreal Env. Res.*, 5, 349–359, 2000. 28233, 28245, 28246, 28251
- 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.: Physical characterization of aerosol particles during nucleation events, *Tellus B*, 53, 344–358, 2001. 28236, 28238
- Andreae, M. O. and Rosenfeld, D.: Aerosol-cloud-precipitation interactions. Part 1. The nature and sources of cloud-active particles, *Earth-Science Rev.*, 89, 13–41, 2008. 28232
- Bigg, E. K. and Leck, C.: Cloud-active particles over the central Arctic Ocean, *J. Geophys. Res.*, 106, 32155–32166, 2001. 28232
- Bonan, G. B.: Forests and climate change: forcings, feedbacks, and the climate benefits of forests, *Science*, 320, 1444–1449, doi:10.1126/science.1155121, 2008.
- Boy, M., Kulmala, M., Ruuskanen, T. M., Pihlatie, M., Reissell, A., Aalto, P. P., Keronen, P., Dal Maso, M., Hellen, H., Hakola, H., Jansson, R., Hanke, M., and Arnold, F.: Sulphuric acid closure and contribution to nucleation mode particle growth, *Atmos. Chem. Phys.*, 5, 863–878, doi:10.5194/acp-5-863-2005, 2005. 28245, 28253
- Broekhuizen, K., Chang, R.Y.-W., Leaith, W. R., Li, S.-M., and Abbatt, J. P. D.: Closure between measured and modeled cloud condensation nuclei (CCN) using size-resolved aerosol compositions in downtown Toronto, *Atmos. Chem. Phys.*, 6, 2513–2524, doi:10.5194/acp-6-2513-2006, 2006.
- Covert, D. S., Gras, J. L., Wiedensohler, A., and Stratmann, F.: Comparison of directly measured CCN with CCN modeled from the number-size distribution in the marine boundary layer during ACE 1 at Cape Grim, Tasmania, *J. Geophys. Res.*, 103, 16597–16608, 1998. 28233
- Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen, K. E. J.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, *Boreal Env. Res.*, 10, 323–336, 2005. 28246
- Dal Maso, M., Sogacheva, L., Aalto, P. P., Riipinen, I., Komppula, M., Tunved, P., Korhonen, L., Suur-Uski, V., Hirsikko, A., Kurtén, T., Kerminen, V.-M., Lihavainen, H., Viisanen, Y., Hansson, H.-C., and Kulmala, M.: Aerosol size distribution measurements at four Nordic field stations:

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



identification, analysis and trajectory analysis of new particle formation bursts, *Tellus B*, 59, 350–361, 2007. 28233, 28248

Dal Maso, M., Sogacheva, L., Anisimov, M. P., Arshinov, M., Baklanov, A., Belan, B., Khodzher, T. V., Obolkin, V. A., Staroverova, A., Vlasov, A., Zagaynov, V. A., Lushnikov, A., Lyubovtseva, Y. S., Riipinen, I., Kerminen, V.-M., and Kulmala, M.: Aerosol particle formation events at two Siberian stations inside the boreal forest, *Boreal Env. Res.*, 13, 81–92, 2008. 28233

Duplissy, J., Gysel, M., Alfara, M. R., Dommen, J., Metzger, A., Prévôt, A. S. H., Wein-gartner, E., Laaksonen, A., Raatikainen, T., Good, N., Turner, S. F., McFiggans, G., and Baltensperger, U.: Cloud forming potential of secondary organic aerosol under near atmo-spheric conditions, *Geophys. Res. Lett.*, 35, L03818, doi:10.1029/2007GL031075, 2008. 28233, 28244, 28252, 28267

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Bormann, S., and Andreae, M. O.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, *Science*, 312, 1375–1378, 2006. 28250

Dusek, U., Frank, G. P., Curtius, J., Drewnick, F., Schneider, J., Kürten, A., Rose, D., Andreae, M. O., Bormann, S., and Pöschl, U.: Enhanced organic mass fraction and decreased hygroscopicity of cloud condensation nuclei (CCN) during new particle formation events, *Geophys. Res. Lett.*, 37, L03804, doi:10.1029/2009GL040930, 2010. 28244, 28252

Ehn, M., Petäjä, T., Aufmhoff, H., Aalto, P., Hämeri, K., Arnold, F., Laaksonen, A., and Kul-mala, M.: Hygroscopic properties of ultrafine aerosol particles in the boreal forest: diurnal variation, solubility and the influence of sulfuric acid, *Atmos. Chem. Phys.*, 7, 211–222, doi:10.5194/acp-7-211-2007, 2007. 28235

Engelhart, G. J., Asa-Awuku, A., Nenes, A., and Pandis, S. N.: CCN activity and droplet growth kinetics of fresh and aged monoterpene secondary organic aerosol, *Atmos. Chem. Phys.*, 8, 3937–3949, doi:10.5194/acp-8-3937-2008, 2008. 28244, 28252, 28267

Ervens, B., Cubison, M., Andrews, E., Feingold, G., Ogren, J. A., Jimenez, J. L., DeCarlo, P., and Nenes, A.: Prediction of cloud condensation nucleus number concentration using mea-surements of aerosol size distributions and composition and light scattering enhancement due to humidity, *J. Geophys. Res.*, 112, D10S32, doi:10.1029/2006JD007426, 2007. 28233

Furutani, H., Dallosto M., Roberts, G. C., and Prather, K. A.: Assessment of the relative impor-tance of atmospheric aging on CCN activity derived from measurements, *Atmos. Environ.*, 42, 3130–3142, 2008.

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Gasparini, R., Collins, D. R., Andrews, E., Sheridan, P. J., Ogren, J. A., and Hudson, J. G.: Coupling aerosol size distributions and size-resolved hygroscopicity to predict humidity-dependent optical properties and cloud condensation nuclei spectra, *J. Geophys. Res.*, 111, D05S13, doi:10.1029/2005JD006092, 2006.

5 Good, N., Topping, D. O., Duplissy, J., Gysel, M., Meyer, N. K., Metzger, A., Turner, S. F., Baltensperger, U., Ristovski, Z., Weingartner, E., Coe, H., and McFiggans, G.: Widening the gap between measurement and modelling of secondary organic aerosol properties?, *Atmos. Chem. Phys.*, 10, 2577–2593, doi:10.5194/acp-10-2577-2010, 2010. 28233, 28245

10 Gunthe, S. S., King, S. M., Rose, D., Chen, Q., Roldin, P., Farmer, D. K., Jimenez, J. L., Artaxo, P., Andreae, M. O., Martin, S. T., and Pöschl, U.: Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity, *Atmos. Chem. Phys.*, 9, 7551–7575, doi:10.5194/acp-9-7551-2009, 2009. 28244, 28252

15 Hakola, H., Tarvainen, V., Laurila, T., Hiltunen, V., Hellen, H., and Keronen, P.: Seasonal variation of VOC concentrations above a boreal coniferous forest, *Atmos. Environ.*, 37, 1623–1634, 2003. 28244

Hämeri, K., Väkevää, M., Aalto, P. P., Kulmala, M., Swietlicki, E., Zhou, J., Seidl, W., Becker, E., and O'Dowd, C. D.: Hygroscopic and CCN properties of aerosol particles in boreal forests, *Tellus*, 58B, 359–379, 2001.

20 Hari, P. and Kulmala, M.: Station for Measuring Ecosystem-Atmosphere relations, *Boreal Env. Res.*, 10, 315–322, 2005. 28234

Hudson, J. G.: Variability of the relationship between particle size and cloud-nucleating ability, *Geophys. Res. Lett.*, 34, L08801, doi:10.1029/2006GL028850, 2007.

25 Hudson, J. G. and Noble, S.: CCN and cloud droplet concentrations at a remote ocean site, *Geophys. Res. Lett.*, 36, L13812, doi:10.1029/2009GL038465, 2009. 28233

Hudson, J. G. and Yum, S. S.: Cloud condensation nuclei spectra and polluted and clean clouds over the Indian Ocean, *J. Geophys. Res.*, 107(D19), 8022, doi:10.1029/2001JD000829, 2002. 28233

30 Hyvönen, S., Junninen, H., Laakso, L., Dal Maso, M., Grönholm, T., Bonn, B., Keronen, P., Aalto, P., Hiltunen, V., Pohja, T., Launiainen, S., Hari, P., Mannila, H., and Kulmala, M.: A look at aerosol formation using data mining techniques, *Atmos. Chem. Phys.*, 5, 3345–3356, doi:10.5194/acp-5-3345-2005, 2005. 28248

Jimenez, J. L., Canagaratna, M., Donahue, N., Prevot, A., Zhang, Q., Kroll, J., DeCarlo, P.,

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- Allan, J., Coe, H., Ng, N., Aiken, A., Docherty, K., Ulbrich, I., Grieshop, A., Robinson, A., Duplissy, J., Smith, J., Wilson, K., Lanz, V., Hueglin, C., Sun, Y., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J., Collins, D., Cubison, M., Dunlea, E., Huffman, J., Onasch, T., Alfarra, M., Williams, P., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Grin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J., Zhang, Y., Dzepina, K., Kimmel, J., Sueper, D., Jayne, J., Herndon, S., Trimborn, A., Williams, L., Wood, E., Middlebrook, A., Kolb, C., Baltensperger, U., and Worsnop, D.: Evolution of Organic Aerosols in the Atmosphere, *Science*, 326, 1525–1529, doi:10.1126/science.1180353, 2009. 28245, 28247, 28253
- Jurányi, Z., Gysel, M., Duplissy, J., Weingartner, E., Tritscher, T., Dommen, J., Henning, S., Ziese, M., Kiselev, A., Stratmann, F., Georgec, I., and Baltensperger, U.: Influence of gas-to-particle partitioning on the hygroscopic and droplet activation behaviour of α -pinene secondary organic aerosol, *Phys. Chem. Chem. Phys.*, 11, 8091–8097, doi:10.1039/b904162a, 2009. 28244
- Kerminen, V.-M., Lihavainen, H., Komppula, M., Viisanen, Y., and Kulmala, M.: Direct observational evidence linking atmospheric aerosol formation and cloud droplet activation, *Geophys. Res. Lett.*, 32, L14803, doi:10.1029/2005GL023130, 2005. 28233
- Köhler, H.: The nucleus in and the growth of hygroscopic droplets, *T. Faraday Soc.*, 32, 1152–1161, 1936. 28236
- Kuang, C., McMurry, P. H., and McCormick, A. V.: Determination of cloud condensation nuclei production from measured new particle formation events, *Geophys. Res. Lett.*, 36, L09822, doi:10.1029/2009GL037584, 2009. 28233
- Kulmala, M., Hämeri, K., Aalto, P. P., Mäkelä, J. M., Pirjola, L., Nilsson, E. D., Buzorius, G., Rannik, Ü., Dal Maso, M., Seidl, W., Hoffman, T., Janson, R., Hansson, H.-C., Viisanen, Y., Laaksonen, A., and O'Dowd, C. D.: Overview of the international project on biogenic aerosol formation in the boreal forest (BIOFOR), *Tellus*, 53B, 324–343, 2001. 28233, 28234
- Kulmala, M., Asmi, A., Lappalainen, H. K., Carslaw, K. S., Pöschl, U., Baltensperger, U., Hov, Ø., Brenquier, J.-L., Pandis, S. N., Facchini, M. C., Hansson, H.-C., Wiedensohler, A., and O'Dowd, C. D.: Introduction: European Integrated Project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales, *Atmos. Chem. Phys.*, 9, 2825–2841, doi:10.5194/acp-9-2825-2009, 2009. 28235, 28251
- Kurtén, T., Kulmala, M., Dal Maso, M., Suni, T., Reissell, A., Vehkamäki, H., Hari, P., Laakso-

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



nen, A., Viisanen, Y., and Vesala, T.: Estimation of different forest-related contributions to the radiative balance using observations in southern Finland, *Boreal Env. Res.*, 8, 275–285, 2003. 28233

Kuwata, M., Kondo, Y., Miyazaki, Y., Komazaki, Y., Kim, J. H., Yum, S. S., Tanimoto, H., and Matsueda, H.: Cloud condensation nuclei activity at Jeju Island, Korea in spring 2005, *Atmos. Chem. Phys.*, 8, 2933–2948, doi:10.5194/acp-8-2933-2008, 2008. 28233

Laaksonen, A., Korhonen, P., Kulmala, M., and Charlson, R. J.: Modification of the Köhler Equation to Include Soluble Trace Gases and Slightly Soluble Substances, *American Meteorological Society*, 55, 853–862, 1998.

Laaksonen, A., Hamed, A., Joutsensaari, J., Hiltunen, L., Cavalli, F., Junkermann, W., Asmi, A., Fuzzi, S., and Facchini, M. C.: Cloud condensation nucleus production from nucleation events at a highly polluted region, *Geophys. Res. Lett.*, 32, L06812, doi:10.1029/2004GL022092, 2005. 28233

Lihavainen, H., Kerminen, V.-M., Komppula, M., Hatakka, J., Aaltonen, V., Kulmala, M., and Viisanen, Y.: Production of “potential” cloud condensation nuclei associated with atmospheric new-particle formation in northern Finland, *J. Geophys. Res.*, 108(D24), 4782, doi:10.1029/2003JD003887, 2003. 28233

Lihavainen, H., Kerminen, V.-M., Tunved, P., Aaltonen, V., Arola, A., Hatakka, J., Hyvärinen, A., and Viisanen, Y.: Observational signature of the direct radiative effect by natural boreal forest aerosols and its relation to the corresponding first indirect effect, *J. Geophys. Res.*, 114, D20206, doi:10.1029/2009JD012078, 2009. 28233

Liu, B. Y. H., Pui, D. Y. H., Whitby, K. T., Kittelson, D. B., Kousaka, Y., and McKenzie, R. L.: The aerosol mobility chromatograph: a new detector for sulfuric acid aerosols, *Atmos. Environ.*, 12, 99–104, 1978. 28235

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, *Atmos. Chem. Phys.*, 5, 715–737, doi:10.5194/acp-5-715-2005, 2005. 28232

Makkonen, R., Asmi, A., Korhonen, H., Kokkola, H., Järvenoja, S., Räisänen, P., Lehtinen, K. E. J., Laaksonen, A., Kerminen, V.-M., Järvinen, H., Lohmann, U., Bennartz, R., Feichter, J., and Kulmala, M.: Sensitivity of aerosol concentrations and cloud properties to nucleation and secondary organic distribution in ECHAM5-HAM global circulation model, *Atmos. Chem. Phys.*, 9, 1747–1766, doi:10.5194/acp-9-1747-2009, 2009. 28233

Manninen, H. E., Nieminen, T., Riipinen, I., Yli-Juuti, T., Gagné, S., Asmi, E., Aalto, P. P., Petäjä, T., Kerminen, V.-M., and Kulmala, M.: Charged and total particle formation and

ACPD

10, 28231–28272, 2010

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



growth rates during EUCAARI 2007 campaign in Hyytiälä, Atmos. Chem. Phys., 9, 4077–4089, doi:10.5194/acp-9-4077-2009, 2009. 28248

Medina, J., Nenes, A., Sotiropoulou, R.-E. P., Cottrell, L. D., Ziemba, L. D., Beckman, P. J., and Griffin, R. J.: Cloud condensation nuclei closure during the international Consortium for Atmospheric Research on Transport and Transformation 2004 campaign: Effects of size-resolved composition, J. Geophys. Res., 112, D10S31, doi:10.1029/2006JD007588, 2007. 28233

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971, doi:10.5194/acp-7-1961-2007, 2007. 28239, 28240, 28244, 28245, 28251

Pierce, J. R. and Adams, P. J.: Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates, Atmos. Chem. Phys., 9, 1339–1356, doi:10.5194/acp-9-1339-2009, 2009. 28233

Poulain, L., Wu, Z., Petters, M. D., Wex, H., Hallbauer, E., Wehner, B., Massling, A., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and CCN activation for secondary organic aerosols – Part 3: Influence of the chemical composition on the hygroscopic properties and volatile fractions of aerosols, Atmos. Chem. Phys., 10, 3775–3785, doi:10.5194/acp-10-3775-2010, 2010. 28233

Quinn, P. K., Bates, T. S., Coffman, D. J., and Covert, D. S.: Influence of particle size and chemistry on the cloud nucleating properties of aerosols, Atmos. Chem. Phys., 8, 1029–1042, doi:10.5194/acp-8-1029-2008, 2008.

Ramanathan, V., Crutzen, P. J., Kiehl, J. T., and Rosenfeld, D.: Atmosphere – Aerosols, Climate and the Hydrological Cycle, Science, 294, 2119–2124, doi:10.1126/science.1064034, 2001.

Raymond, T. M. and Pandis, S. N.: Formation of cloud droplets by multicomponent organic particles, J. Geophys. Res., 108(D15), 4469, doi:10.1029/2003JD003503, 2003.

Rissler, J., Swietlicki, E., Zhou, J., Roberts, G., Andreae, M. O., Gatti, L. V., and Artaxo, P.: Physical properties of the sub-micrometer aerosol over the Amazon rain forest during the wet-to-dry season transition – comparison of modeled and measured CCN concentrations, Atmos. Chem. Phys., 4, 2119–2143, doi:10.5194/acp-4-2119-2004, 2004. 28233

Roberts, G. and Nenes, A.: A Continuous-Flow Streamwise Thermal-Gradient CCN Chamber for Atmospheric Measurements, Aerosol Sci. Tech., 39, 206–221, 2005. 28234

Roberts, G., Mauger, G., Hadley O., and Ramanathan, V.: North American and Asian aerosols over the eastern Pacific Ocean and their role in regulating cloud condensation nuclei, J.

- Geophys. Res., 111, D13205, doi:10.1029/2005JD006661, 2006. 28233
- Rosenfeld, D., Lohmann U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell A., and Andreae, M. O.: Flood or drought: How do aerosols affect precipitation?, Science, 321, 1309–1313, 2008. 28232
- 5 Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, A Wiley-Interscience Publication, USA, 1998. 28236, 28237, 28241
- Snider, J. R. and Brenguier, J.-L.: Cloud condensation nuclei and cloud droplet measurements during ACE-2, Tellus, 52B, 828–842, 2000. 28232
- Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V.-M., Sihto, S.-L., Riipinen, I.,
 10 Merikanto, J., Mann, G. W., Chipperfield, M. P., Wiedensohler, A., Birmili, W., and Lihavainen, H.: Contribution of particle formation to global cloud condensation nuclei concentrations, Geophys. Res. Lett., 35, L06808, doi:10.1029/2007GL033038, 2008a. 28233
- Spracklen, D. V., Bonn, B., and Carslaw, K. S.: Boreal forests, aerosols and the impacts of clouds and climate, Phil. Trans. Royal Soc. A., 366, 4613–4626, 2008b. 28233
- 15 Stevens, B. and Feingold, G.: Untangling aerosols effects on clouds and precipitation in a buffered system, Nature, 461, 607–613, doi:10.1038/nature08281, 2009. 28232
- Swietlicki, E., Zhou, J., Berg, O. H., Martinsson, B. G., Frank, G., Cederfelt, S.-I., Dusek, U., Berner, A., Birmili, W., Wiedensohler, A., Yuskiewich, B., and Bower, K. N.: A closure study of sub-micrometer aerosol particle hygroscopic behaviour, Atmos. Res., 50, 205–240, 1999.
- 20 Swietlicki, E., Hanson, H.-C., Hämeri, K., Svenningsson, B., Massling, A., McFiggans, G., Murry, P., H., Petäjä, T., Tunved, P., Gysel, M., Topping, D., Weingartner, E., Baltensperger, U., Rissler, J., Wiedensohler, A., and Kulmala, M.: Hygroscopic properties of submicrometer atmospheric aerosol particles measured with H-TDMA instruments in various environments – a review, Tellus, 60B, 432–469, 2008.
- 25 Tarvainen, V., Hakola, H., Hellén, H., Bäck, J., Hari, P., and Kulmala, M.: Temperature and light dependence of the VOC emissions of Scots pine, Atmos. Chem. Phys., 5, 989–998, doi:10.5194/acp-5-989-2005, 2005. 28244
- Tunved, P., Hansson, H.-C., Kerminen, V.-M., Ström, J., Dal Maso, M., Lihavainen, H., Viisanen, Y., Aalto, P. P., Komppula, M., and Kulmala, M.: High Natural Aerosol Loading over Boreal Forests, Science, 312, 261–263, doi:10.1126/science.1123052, 2006. 28233
- 30 Twohy, C. T. and Anderson, J. R.: Droplet nuclei in non-precipitating clouds: composition and size matters, Environ. Res. Lett. 3, 045002, doi:10.1088/1748-9326/3/4/045002, 2008.
- Twomey, S.: Pollution and the planetary albedo, Atmos. Environ., 8, 1251–1256,

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



doi:10.1016/0004-6981(74)90004-3, 1974. 28232

Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, Atmos. Chem. Phys., 9, 239–260, doi:10.5194/acp-9-239-2009, 2009. 28233

Wex, H., Petters, M. D., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol: Part 1 – Evidence from measurements, Atmos. Chem. Phys., 9, 3987–3997, doi:10.5194/acp-9-3987-2009, 2009. 28245

Wiedensohler, A., Chen, Y. F., Nowak, A., Wehner, B., Achtert, P., Berghof, M., Birmili, W., Wu, Z. J., Hu, M., Zhu, T., Takegawa, N., Kita, K., Kondo, Y., Lou, S. R., Hofzumahaus, A., Holland, F., Wahner, A., Gunthe, S. S., Rose, D., Su, H., and Pöschl, U.: Rapid aerosol particle growth and increase of cloud condensation nucleus activity by secondary aerosol formation and condensation: A case study for regional air pollution in northeastern China, J. Geophys. Res., 114, D00G08, doi:10.1029/2008JD010884, 2009. 28233

ACPD

10, 28231–28272, 2010

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Table 1. Monthly averages of CCN concentration, activated fraction (CCN/CN) and critical diameter, measured at SMEAR II station in Hyytiälä July 2008–June 2009. The data are for supersaturation of 0.4% representing an average case in the range of measured supersaturations.

Month	CCN concentration [cm ⁻³]	Activated fraction	d_{crit} (CCNC) [nm]	d_{crit} (HTDMA) [nm]
Jul 08	804	0.42	81	96
Aug 08	563	0.41	79	89
Sep 08	514	0.36	83	85
Oct 08	328	0.22	91	89
Nov 08	261	0.21	99	88
Dec 08	327	0.36	94	78
Jan 09	336	0.31	82	74
Feb 09	648	0.39	66	78
Mar 09	567	0.35	62	73
Apr 09	675	0.36	58	77
May 09	744	0.33	68	79
Jun 09	791	0.40	72	85

All values are at supersaturation SS = 0.4%.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



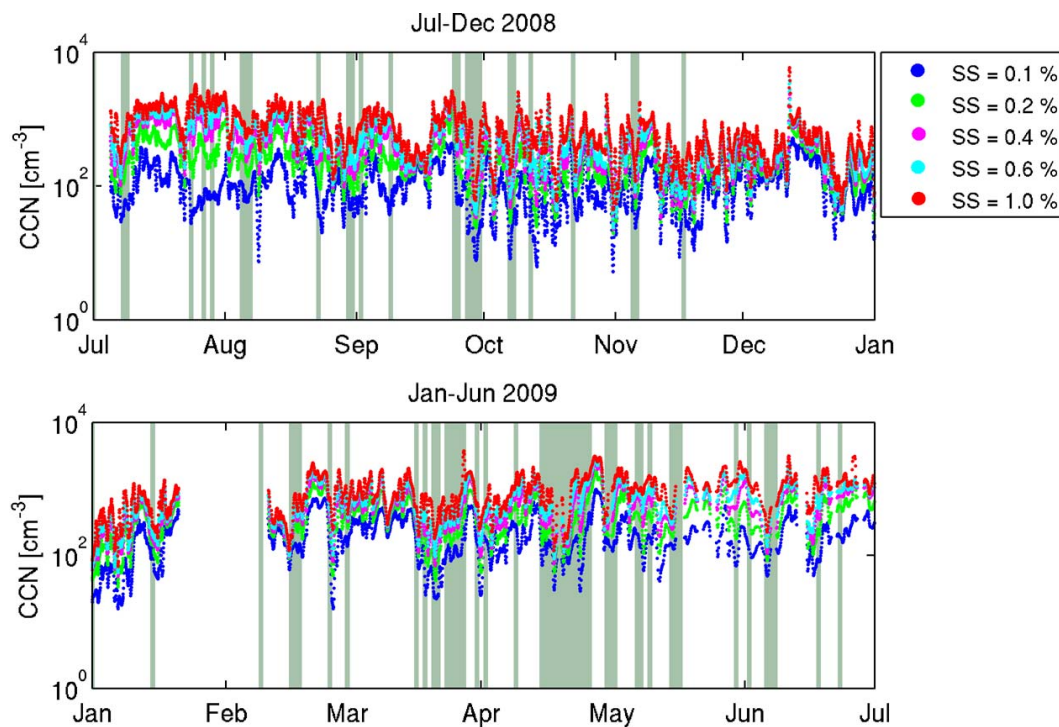


Fig. 1. Time series of CCN concentration at Hyytiälä SMEAR II station measured with five water supersaturations (SS): 0.1% (blue), 0.2% (green), 0.4% (purple), 0.6% (light blue), 1.0% (red). The data points are 1 h averages, which have been smoothed by taking a moving average with a 10 points window. Days with new particle formation (event days) are marked with a grey background.

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

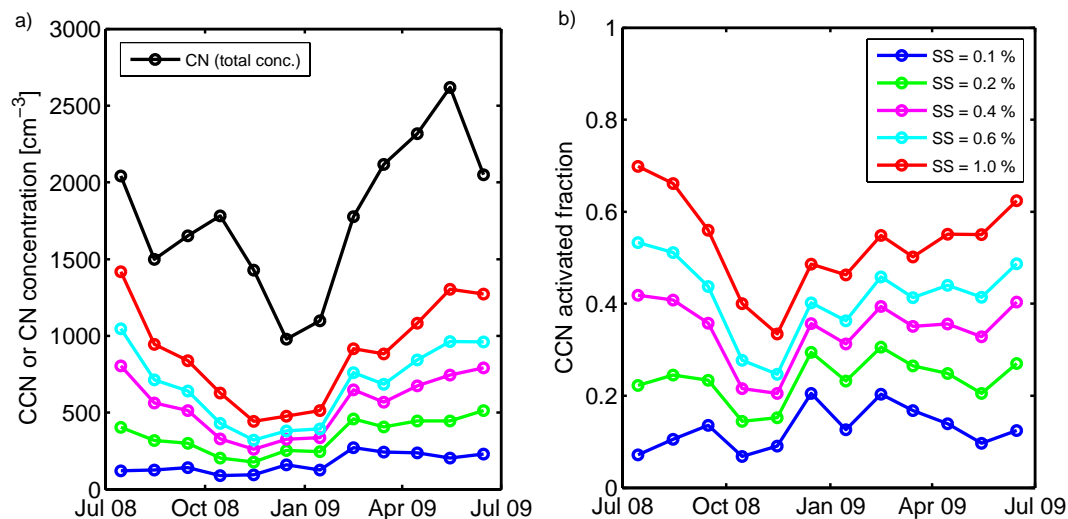


Fig. 2. (a) Monthly mean values of the CCN concentrations at different supersaturations and the total number concentration (CN), and (b) monthly means of the activated fraction (CCN/CN).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

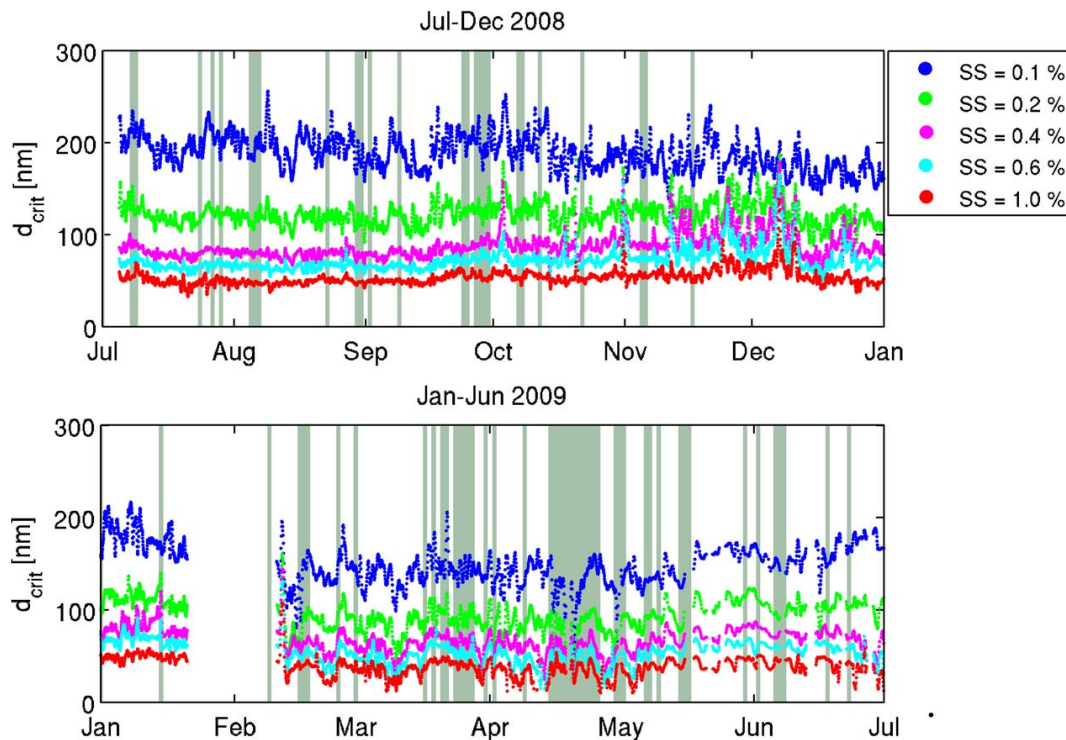


Fig. 3. Time evolution of critical diameter, estimated from CCN data and particle size distribution data. Different colours correspond to five supersaturation (SS) levels: 0.1% (blue), 0.2% (green), 0.4% (purple), 0.6% (light blue), 1.0% (red). The data points are 1 h averages which have been smoothed by taking a moving average with a 10 points window. Days with new particle formation (event days) are marked with a grey background.

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

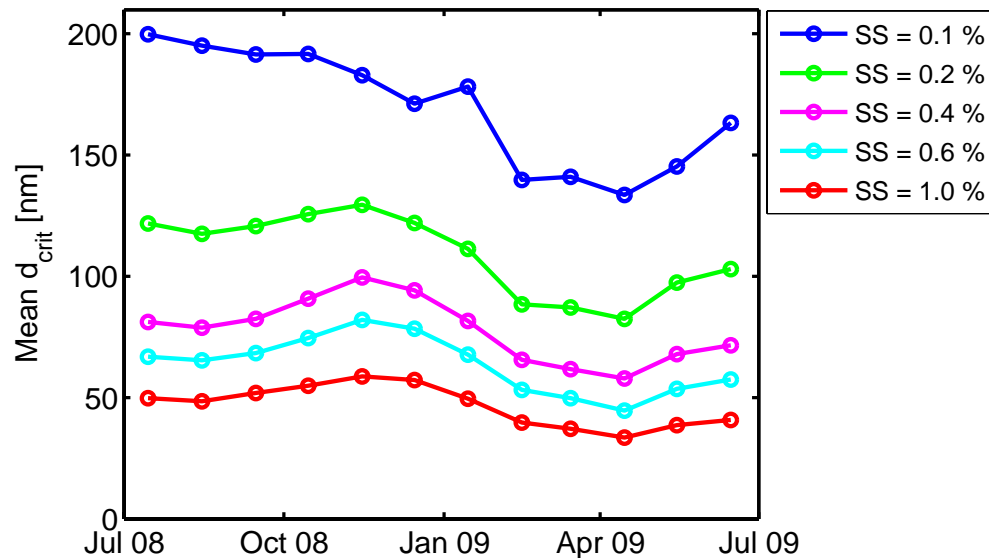


Fig. 4. Monthly mean values of critical diameter estimated from the CCN concentrations and size distribution measurements.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

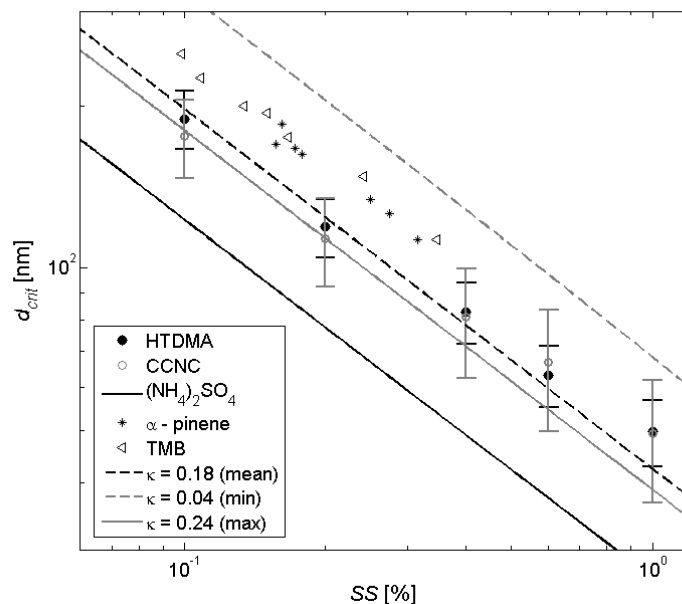


Fig. 5. Average critical diameter estimated from CCNC and HTDMA data as a function of water supersaturation. Error bars show the standard deviations over the one-year data set. Data from laboratory measurements by Duplissy et al. (2008) with α -pinene and trimethylbenzene (TMB) aerosol are plotted with markers (stars and triangles). Lines show theoretical values from Köhler equation for pure ammonium sulphate particles (black solid line) and from κ -Köhler theory with mean κ -value determined in this study ($\kappa = 0.18$), and range of κ -values from literature for organic (monoterpene) aerosol (Engelhart et al., 2008).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



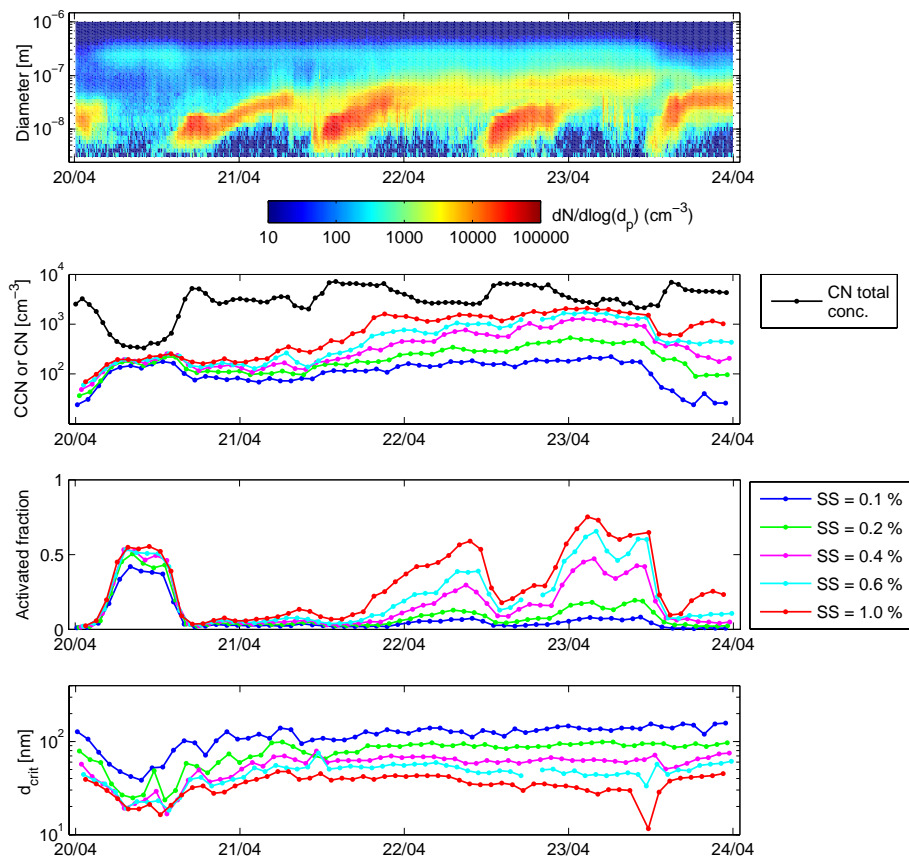


Fig. 6. Comparison of the time evolution of particle size distributions, cloud condensation nuclei (CCN) and total particle concentrations (CN), activated fraction (CCN/CN), and critical diameter (estimated from CCN and size distribution measurements) on four consecutive new particle formation days in Hyytiälä (20 April–23 April 2009).

Seasonal variation of CCN concentrations in boreal forest

S.-L. Sihto et al.

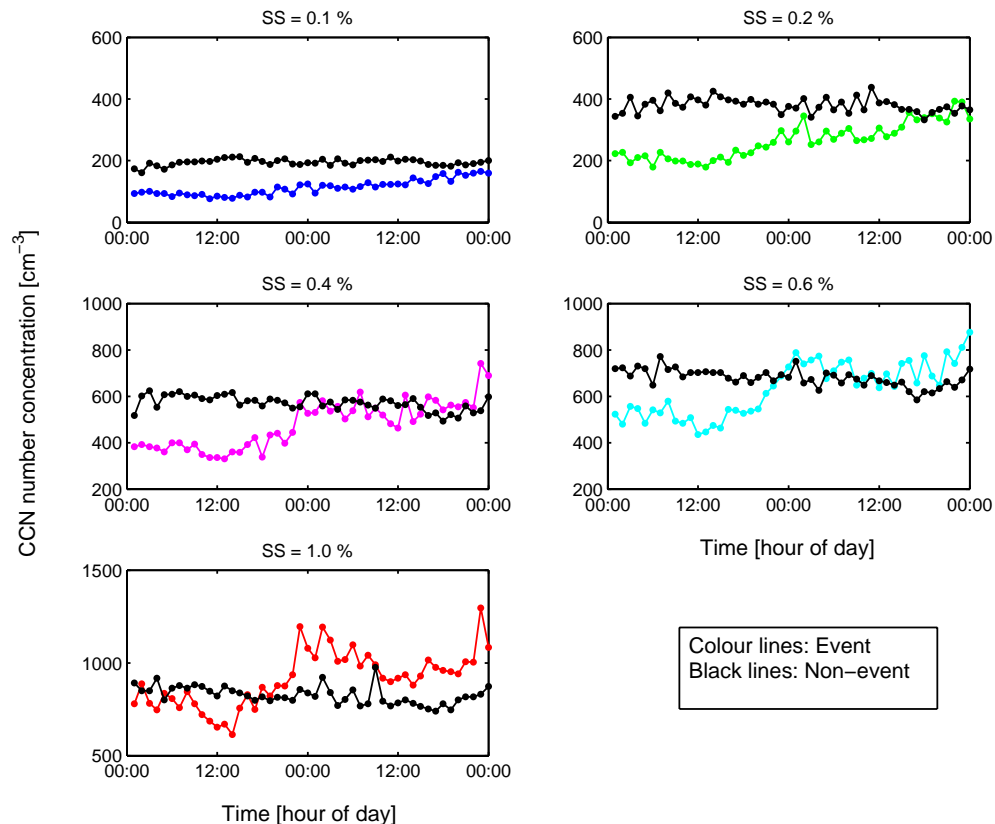
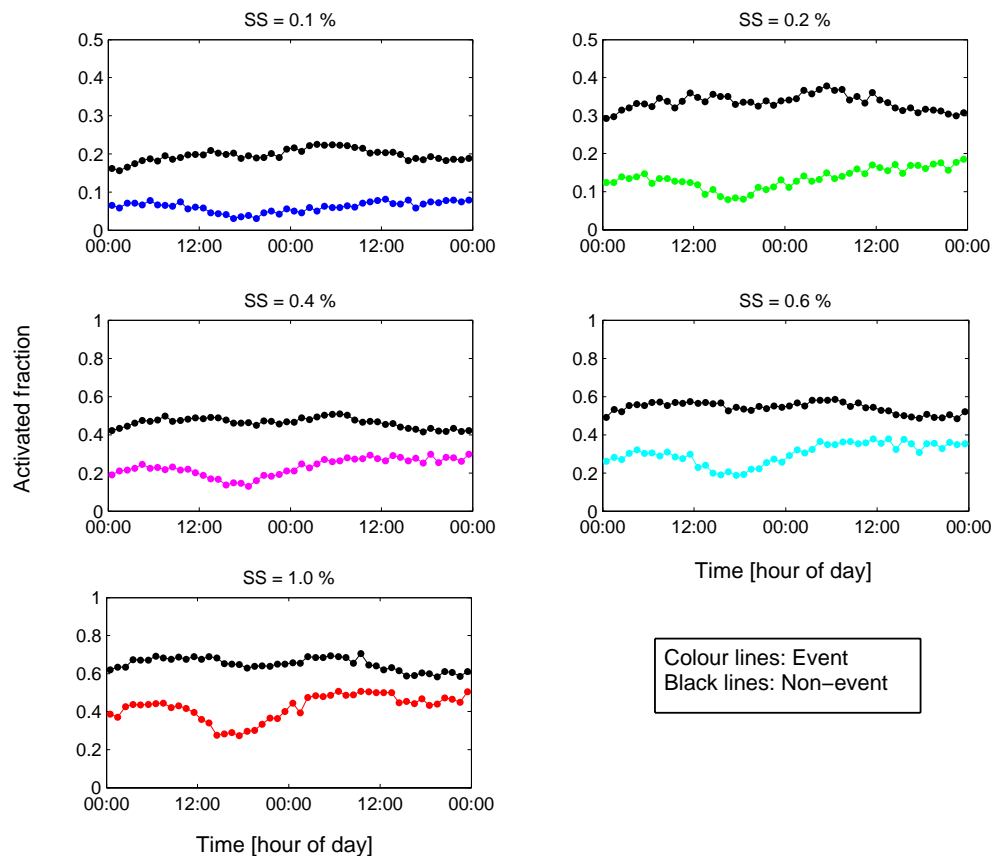


Fig. 7. Mean diurnal variation of CCN concentrations at different supersaturations on two consecutive days, separated according to whether the first day was an event day (coloured markers) or a non-event day (black markers). The points are hourly averages over the one year data set.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


**Seasonal variation of
CCN concentrations
in boreal forest**

S.-L. Sihto et al.

**Fig. 8.** Same as Fig. 7, but for the activated fraction (CCN/CN).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Seasonal variation of
CCN concentrations
in boreal forest**

S.-L. Sihto et al.

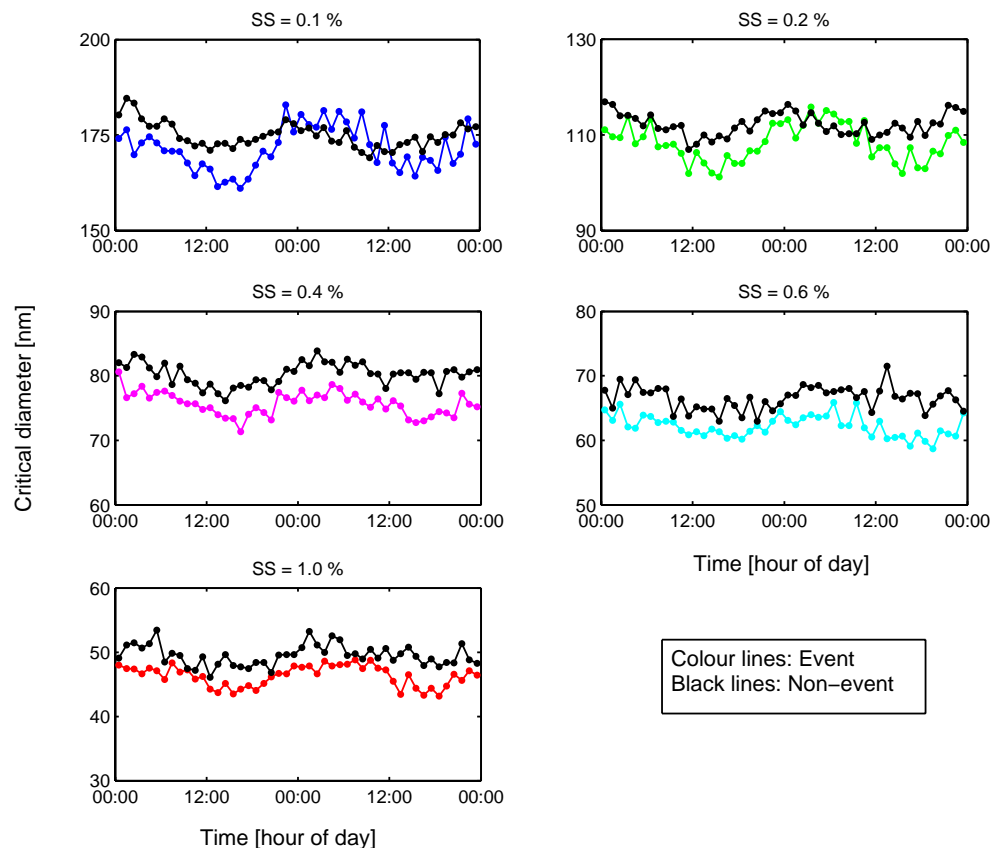


Fig. 9. Same as Fig. 7, but for the critical diameter determined from the CCN and size distribution measurements.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Seasonal variation of
CCN concentrations
in boreal forest**

S.-L. Sihto et al.

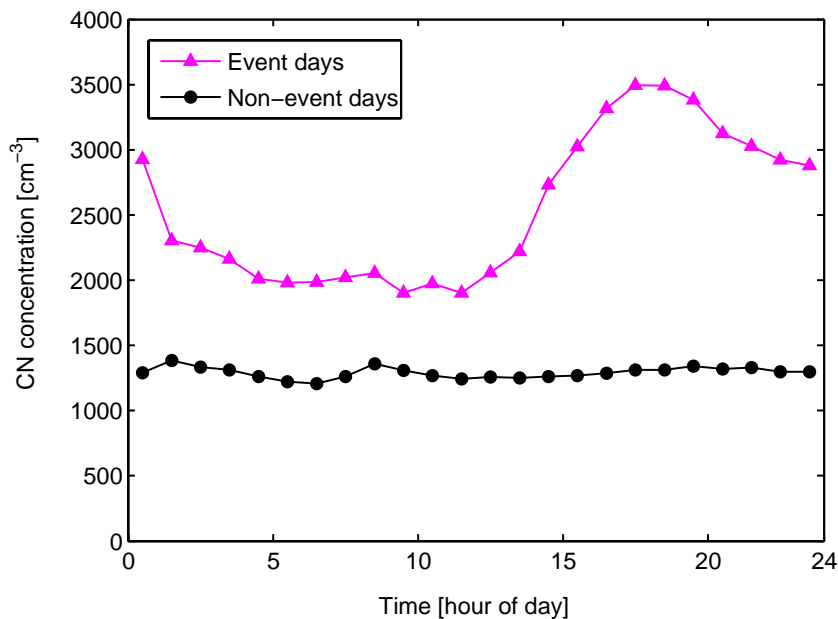


Fig. A1. Mean total particle number concentration (CN, diameter range 3–1000 nm) on event and non-event days at Hyytiälä SMEAR II station.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)