Tellus SERIES B CHEMICAL AND PHYSICAL METEOROLOGY
PUBLISHED BY THE INTERNATIONAL METEOROLOGICAL INSTITUTE IN STOCKHOLM

Tellus (2008), 60B, 495–508
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TELLUS

Annual and interannual variation in boreal forest aerosol particle number and volume concentration and their connection to particle formation

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(Manuscript received 31 October 2007; in final form 6 May 2008)

ABSTRACT

We investigated size-resolved submicrometre aerosol particle number and volume concentration time series as well as aerosol dynamic parameters derived from Differential Mobility Particle Sizer (DMPS) measurements at five background stations in the Nordic boreal forest area. The stations in question were Aspvreten, Hyytiälä and Utö in southern Finland and Sweden, and Värriö and Pallas in the Finnish Lapland. The objective of our investigation was to identify and quantify annual and interannual variation observable in the time series. We found that the total number and mass concentrations were much lower at the Lapland stations than at the southern stations and that the total particle number was strongly correlated to particle formation event frequency. The annual total number concentration followed the annual distribution of particle formation events at the southern stations but much less clearly at the Lapland stations. The volume concentration was highest during summer, in line with higher condensation growth rates; this is in line with the assumption that a large part of the particle volume is produced by oxidized plant emissions. The decrease of sulphate emissions in Europe was not visible in our data set. Aerosol dynamic parameters such as condensation sink, condensation sink diameter and the power law exponent linking coagulation losses and condensation sink are presented to characterize the submicron Nordic background aerosol.

1. Introduction

Aerosol particles play a major role in the atmospheric matter and energy transfer processes. They directly affect the radiation balance by scattering incoming short-wave solar radiation and by absorbing long-wave radiation. They also play a critical role in cloud formation by acting as nuclei for cloud droplet condensation; changes in the aerosol population alter the cloud microphysical properties. These changes in turn can lead to changes in the albedo or lifetime of clouds, affecting the radiation balance and water cycle (see e.g. Albrecht, 1989; Twomey, 1991; Garrett et al., 2002). In addition to these climatic effects, high aerosol loads have been found to have adverse effects on human health, causing increased mortality as well as pulmonary and cardiovascular problems (Goldberg et al., 2001; Slaughter et al., 2003).

The climatic effects, both direct by scattering and indirect by changing cloud properties, constitute a significant uncertainty in understanding the behaviour of our climate system (IPCC, 2007). It has been pointed out that in the past century, aerosol particles emitted by human activities might have been protecting the Earth from a stronger increase in the greenhouse effect (Andreae, 2007a). This is an important issue, because as sulphate emissions will decrease (see e.g. Berglen et al., 2007), this protection might also become less effective.

Instrument technology has allowed continuous measurements of submicron aerosol particle concentrations for only a few decades. Long-term, size-resolved concentration measurements with a good particle size resolution in the planetary boundary layer are quite scarce. Some of the longest data series of such measurements have been recorded at a network of measurement

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DOI: 10.1111/j.1600-0889.2008.00366.x

stations situated in Finland and Sweden, inside the boreal forest zone. Particle size distributions have been measured at several stations continuously, the earliest measurements starting in 1996 in Hyytiälä, southern Finland (Mäkelä et al., 1997). The importance of size-resolved, continuous number concentration measurements has been demonstrated by the findings that new-particle formation by nucleation was occurring very often in the boreal forest boundary layer. Several studies have looked at the measured time series with the focus on this particle formation phenomenon (e.g. Kulmala et al., 2001; Komppula et a., 2003; Dal Maso et al., 2005, 2007), and found annual trends and interannual differences in the particle formation activity. These studies have often focused on the concept of particle formation events, their frequency and their dynamic behaviour in connection with different atmospheric variables. Continuous time series of size-resolved aerosol number data, however, can be used for other purposes as well. In this study, our aim is to have a look at the long-term time series of particle number concentrations measured in several size ranges in five field stations, and to analyse the time series of aerosol dynamic parameters that can be derived from the size distribution measurements.

This kind of investigation is of interest for several reasons. First, it gives quantitative insight into the submicron particle concentrations prevailing over the Nordic boreal forest environment. The data measured at the stations investigated here can be considered representative of a background aerosol because all the stations are away from areas of significant industrial or residential activities, so that measured aerosols are usually either long-range transported or produced by natural sources. Second, the data set allows us to investigate possible trends in particle number and volume concentrations to find out, for example, whether it is possible to identify a decrease in the submicron particle number concentration that would follow from the decrease in sulphate emissions. Third, the analysis provides insight into the effect of particle formation to the background aerosol concentration. Particle number concentrations in different size ranges, as well as particle volume concentrations, will be compared with trends, or interannual changes, in the nucleation event frequency, nucleation rates and particle growth rates presented by Dal Maso et al. (2007).

As already mentioned, we will also investigate various size distribution parameters that were calculated for all five measurement time series. The calculated parameters include the condensation sink, average particle diameter, condensation sink diameter (see Lehtinen et al. (2003) for definition) and the fresh-particle coagulation loss exponent (Lehtinen et al., 2007). These characteristics are of interest to both modellers and experimentalists, as they can be used to get an estimate of the average character of the submicrometre aerosol. Physically, they provide information on the processes influencing the aerosol system. The condensation sink, for example, is just the inverse of the atmospheric lifetime of vapour lost by condensation onto existing aerosol particles, whereas the coagulation loss exponent can be used to relate this lifetime to the corresponding lifetime of freshly formed particles.

2. Materials and methods

2.1. Measurement sites

We present an analysis of aerosol size distribution data measured at five field stations, all situated inside or at the border of the Nordic the boreal forest area: the SMEAR (Station for Measuring Ecosystem–Atmosphere Relations) I and II stations at Värriö and Hyytiälä, respectively; the Sammaltunturi monitoring station at Pallas and the background measurement station at Aspvreten, Sweden; and the Utö EMEP station. At all these sites the aerosol size distribution measurements are performed using a Differential Mobility Particle Sizer (DMPS), an instrument consisting of a bipolar charger, one or two Differential Mobility Analyzers (DMA) and one or two Condensation Particle Counters (CPC). Depending on the setup, the DMPS can measure particle size distributions with a high size resolution from sizes as low as 3 nm. The sampling time per distribution is usually ca. 10 min.

The SMEAR II station at Hyytiälä (61°51N 24°17′E, 180 m a.s.l.) has extensive facilities for measuring forest-atmosphere relations. The station is surrounded by Scots Pine forest. The nearest urban pollution sites are Tampere (ca. 50 km to the southwest) and Jyväskylä (ca. 100 km to the northeast).

The Aspvreten measurement station (58°46′N, 17°24′E, 25 m a.s.l.) is located near the Swedish coastline in Sörmland, ca. 70 km south of Stockholm and 2 km from the seaside.

The Utö Measurements were conducted at the EMEP-station (European Monitoring and valuation Programme) Utö (59°47′N, 21°23′E), located in Baltic Sea. The station is situated 8 m above sea level some 60 km from the Finnish southwest coast. The wind direction is dominantly southwesterly in fall and winter and north or southwesterly during spring and summer. The wind speed is highest in winter.

The SMEAR I station at Värriö (67°46′N 29°35′E, 400 m a.s.l.) is situated in Lapland, in a rural area far removed from any settlements. It is surrounded by a Scots Pine (*Pinus sylvestris* l.) forest, which is over 40 yr old in the station's immediate vicinity. The measurements are performed on a hill top (Hari et al., 1994).

The Sammaltunturi Global Atmospheric Watch station (67°58′N 24°07′E, 565 m a.s.l.), operated by the Finnish Meteorological Institute, is situated on the top of a field in western Lapland. The Pallas area is in the sub-Arctic region near the northern limit of the boreal forest zone. The area has no significant local or regional pollution sources and the distance to the nearest town (Muonio, with 2500 inhabitants) is about 20 km.

Table 1 presents the locations of the stations and data acquisition periods for all the five stations. More detailed station

Table 1. An overview of the stations and measurements used in this study

Site	Latitude	Longitude	Size range	Meas. start
Smear I, Värriö	67°46′N	29°35′E	8–460 nm	01. Jan 1998
Smear II, Hyytiälä	61°51′N	24°17′E	3–500 nm	30. Jan 1996
Utö, Baltic Sea	59°47′N	21°23′E	7–500 nm	11. Mar 2003
Sammaltunturi, Pallas	67°58′N	24°07′E	10–490 nm	12. Apr 2000
Aspvreten, Sweden	58°46′N	17°24′E	10–450 nm	01. Jun 2000

Fig. 1. A map showing the locations of the five measurement stations Hyytiälä, Värriö, Pallas, Utö and Aspvreten.

descriptions, as well as descriptions of the setup of the aerosol measurement instrumentation can be found in Tunved et al. (2003), Engler et al. (2007), Hyvärinen et al. (2008) Aalto et al. (2001), Hari and Kulmala (2005) and Hatakka et al. (2003) and also references therein. A map showing the locations of all the stations is showed in Fig. 1.

2.2. *Data quality control*

The particle size distribution measurements at the five stations presented here are performed with the same type of instrument, the DMPS. This is of advantage when comparing the measurements to each other. The instrument works by causing a known charge distribution in the sample aerosol, then using a Differential Mobility Analyzer (DMA) to select an electronical mobility fraction of the sample aerosol and then using a Condensation Particle Counter (CPC) to count the particles selected. To retrieve the real ambient particle size distribution, one needs to invert the voltage-particle count data.

The intercomparability of our data set is improved by the fact that the data inversion at all the stations is performed with essentially the same inversion algorithm, which takes into account the same corrections: losses in the sample line, losses in the DMA and CPC counting efficiency. The inversion routine is a pseudo inversion routine with DMA kernels by Stolzenburg (1988) and the charging probabilities of Wiedensohler (1988).

While the instruments have not been all intercompared, some comparisons have been performed. The Aspvreten DMPS system took part in a EUSAAR workshop where DMPS systems from various institutes were compared, and was found to perform well when compared to other DMPS systems with ambient air. The Utö DMAs (exchanged once due to salt corrosion) have been intercompared at the University of Helsinki prior to deployment. The Pallas station is part of the Global Atmospheric Watch (GAW) program; thus the instruments are audited by the GAW authorities and are sent to calibration/comparison workshops in continuous basis. The DMPS data quality assurance is done parallel with the CPC data, which reveals the possible errors in the data that are not seen earlier. At the Hyytiälä and Värriö stations, measurements using only CPCs have been ongoing in parallel with the size distribution measurements. Comparisons to these measurements show an agreement within approximately 10% of the concentration, except when the size distribution is dominated by very small particles, in which case the differing cut sizes of the instruments have an effect. All stations are visited routinely for maintenance checks. The instrument flows are calibrated typically every month.

We estimate the error in particle number concentrations to be typically 10–20% for the total concentration. The large effect of diffusion losses for the smallest particles causes the error to be larger for smaller particle concentrations.

The error in the measurement is a combination of random and systematic errors. In this study, we investigate the particle number concentrations using sliding averages over long time periods, of the order of months. The long averaging periods largely eliminate the random element, which is expected to vary in both the positive and negative direction. To our knowledge, the main source of systematic error or difference between the instruments comes from the cut size of each instrument. Therefore, the error is largest when there are a large number of the smallest particles present. This aspect is discussed below, together with the data treatment.

2.3. Data treatment

The particle size distributions were measured at intervals ranging from 5.4 to 10 min. For each size distribution, we calculated a number of parameters, described in following section.

2.3.1. Number concentrations. We calculated the total particle number concentration, N_{tot}, as well as the number concentrations in different size ranges. Since the lifetime and properties of aerosol particles are strongly dependent on the particle size, it is of interest to investigate the number concentrations in different size ranges. For the submicron aerosol, the typical size ranges used are the nucleation mode (<25 nm), Aitken mode (25–90 nm) and accumulation mode (90–1000 nm). These size ranges are based on the observations of local minima in the size distribution function around the corresponding boundaries of the size ranges (Laakso et al., 2003). 90 nm is also considered a conservative estimate for the size at which particles are capable of activating as CCN (Lihavainen et al., 2003; Kurten et al., 2003; Kerminen et al., 2005; Andreae et al., 2007b) The number concentrations of the nucleation mode (N_{nuc}), Aitken mode (N_{aitk}), accumulation mode (N_{acc}), as well as the total particle number concentration, were integrated numerically from the measured size distribution functions. The accumulation mode is in this study assumed to range from 90 nm to the upper range of the particle sizing instrument; most of the particles are in the range covered by the instruments used here. Therefore, no corrections for the differing size limits are performed. The error caused by the differing size limits was investigated, and we found that the difference between the widest range (Hyytiälä) and the narrowest range (Aspvreten) was an average systematic error of about 5%. This error comes mainly from the low diameter end of the distribution. During events of high small particle concentrations, the error is of course larger. In this study, however, we averaged the data over long time periods, which diminished the importance of these short-lasting events.

2.3.2. Particle volume concentrations. In addition of particle numbers, the measured size distributions can be used to estimate the volume concentration of the particulate matter in the submicron size range. For this to be possible, one has to assume some relation between the measured electrical mobility diameter and the particle volume. In this study, we assumed the particles to be of a spherical shape, based on the assumption that submicron particles are mostly liquid. The total particulate volume concentrations V_{tot} was then integrated numerically from particle number size distributions. The differing size ranges were also investigated in terms of particle volume, and we found an average underprediction of the total particle volume by 5–10% when comparing the widest range with the narrowest range. This depends, of course, on the shape of the particle size distribution. This error is small when compared to the overall variation of the particle volume concentration.

2.3.3. Other parameters. For modelling the source, transformation and loss processes of atmospheric aerosols, some parameters characterizing different aspects of the size distribution come in useful. In this study we have calculated some of these, as they might be of interest for modellers trying to simulate typical Nordic Boreal aerosols.

One of these parameters is the condensation sink *CS*, which is basically the loss rate of a non-volatile vapour undergoing condensation. It can be thought of as the inverse lifetime of a non-volatile condensing vapour. It is strongly dependent on the particle number and size, and is a key parameter in determining condensable vapour concentrations and lifetimes. *CS* was integrated from the measurements using the measured dry size of the particles, as described in Kulmala et al. (2001).

Closely linked to the condensation sink is the condensation sink diameter *CSD*, described by Lehtinen et al. (2003). It describes the diameter of a monodisperse mode that has the same number concentration and condensation sink as the measured size distribution. It is of particular interest to researchers using simple models with monodisperse aerosol representations, as the *CSD* has been shown to yield the best results especially for aesrosol undergoing condensational transformations. The calculation of the *CSD* requires finding the diameter D_p at which $N_{tot} \cdot CS|_{Single}(Dp) = CS|_{observed}$. Here $\cdot CS|_{Single}(Dp)$ is the condensation sink caused by a single particle, which depends on the particle diameter, and $CS|_{observed}$ is the CS caused by the whole measured distribution. *CSD* is found by finding the diameter at which $CS|_{Single}(Dp) - CS|_{observed}./N_{tot} = 0$.

Another parameter calculated was the power-law exponent *m* used for estimating the real nucleation rates from observed apparent nucleation rates or vice versa, as described in Lehtinen et al. (2007). This parameter is the exponent in the power law describing how the coagulation sink for small particles depends on the diameter of the particle being scavenged. The exponent is not a constant but depends on the shape of the number size distribution, and can be numerically calculated if the size distribution function is known. Because the condensation sink *CS* can be thought of as the coagulation sink of vapour molecule-sized particles, the exponent presents a simple way to connect *CS* to the coagulation loss of fresh particles. Typical values for *m* are of interest to researchers trying to couple nucleation models to atmospheric models, as the particle coagulation losses during the early stages of growth can be parametrized with *CS* and *m*. As described by Lehtinen et al. (2007), if the condensation sink is calculated using the properties of sulphuric acid, as in this study, this parametrization takes the form

$$CoagS_{Dp} = CS \cdot \left(\frac{0.71}{D_p}\right)^{-m}, \quad (1)$$

where $CoagS_{Dp}$ is the coagulation sink of particles of size D_p. The factor 0.71 comes from the sulphuric acid hydrate size. The physical significance of the coagulation sink is that of an inverse lifetime and its unit is s^{-1}, similar to the condensation sink.

2.4. Data processing

2.4.1. Averaging. The size distribution characteristics described above were calculated for each measured size distribution. The obtained values were then averaged to 2-h average values to save computer memory and processing time.

To filter out short-term variations from the observations and focus on the long-term features, running means of varying lengths were used. Running means can cause some distortions at the start and end of the time series as well as at the edges of gaps. No gap filling was used.

2.4.2. Removing of outliers. Atmospheric concentration data often contains some extreme values, caused by either instrument malfunctions or some abnormal occurrence in the vicinity of the measurement station. These extreme values, even if rare, can cause severe distortions if some averaging filter such as a running mean is applied to the data. As the aim of this study is to get a general view of the size distribution characteristics and running means were used, we removed measurements that were more than 3 *SD*s smaller or larger than the mean value of the parameter. The removal was performed to the original data, not the 2-h averages.

3. Results and discussion

3.1. Overview of the time series

An overview of the statistics of the particle size distributions calculated for this study is given in Table 2. In the following, we will discuss the general findings in more detail.

3.1.1. Number concentrations. An overview of the particle number and volume concentrations as well as the condensation sink, is presented in Figs. 2 and 3, where we show the time series of the 2-h average concentration as well as the 2-month sliding averages. At all the stations, the particle number concentrations vary between 100 and 10 000 cm^{-3}. The lowest panel of the figure shows all 2-month sliding average concentrations in the same figure. In the total number concentrations, two features are clearly observable: first, the clear annual pattern with minimum concentration occurring in the winter, second, the very clear difference between the more northern Lapland stations (Värriö and Pallas) and the stations situated in the south of Finland/Sweden (Hyytiälä, Aspvreten and Utö). The mean concentrations for the time series at the Lapland stations are 686 (median 461) cm^{-3} and 875 (median 593 cm^{-3}) cm^{-3} for Pallas and Värriö, respectively. The average concentrations in the south are 2220, 2570 and 3210 cm^{-3} for Hyytiälä, Aspvreten and Utö, respectively; these concentrations are three to four times higher than in Lapland.

In the top three panels of Fig. 3 the moving 2-month average particle number concentrations in the nucleation, Aitken and accumulation modes are shown for all the stations. The average concentration in the nucleation mode are from 330 (Aspvreten) to 610 (Hyytiälä) cm^{-3} at the southern stations and 120–140 in

Table 2. A summary of the statistics of the parameters calculated for this study

	Hyytiälä	Aspvreten	Utö	Värriö	Pallas
Total number (cm^{-3})					
Mean	2217.5	2567.4	3210.3	875.5	686.0
Median	1838.5	2158.6	2908.8	593.3	461.1
STD	1669.7	1671.1	1868.4	933.7	687.3
P25	1167.6	1422.7	1743.2	257.5	182.7
P75	2760.8	3256.8	4362.6	1201.7	980.3
Nucleation mode (cm^{-3})					
Mean	607.7	334.1	397.1	142.8	119.1
Median	275.4	149.1	184.3	34.1	33.0
STD	1109.8	564.1	638.7	378.7	283.9
P25	124.4	54.8	84.0	12.0	13.4
P75	619.1	375.6	426.5	118.9	94.8
Aitken mode (cm^{-3})					
Mean	1023.0	1328.5	1841.4	429.0	339.9
Median	780.8	1057.9	1577.8	227.8	192.3
STD	860.9	1024.4	1236.2	606.1	422.3
P25	452.9	617.3	890.2	84.1	72.1
P75	1318.9	1732.2	2522.2	541.9	440.7
Accumulation mode (cm^{-3})					
Mean	581.6	905.8	969.7	304.0	226.6
Median	454.3	703.4	714.1	197.4	115.5
STD	455.9	734.9	839.9	313.1	268.2
P25	243.3	379.8	339.8	87.2	44.6
P75	799.7	1210.3	1362.4	406.7	303.3
Total volume (μm^{-3} cm^{-3})					
Mean	2.9	4.0	3.9	1.5	1.0
Median	2.2	3.0	2.8	1.0	0.5
STD	2.3	3.3	3.5	1.4	1.2
P25	1.2	1.5	1.3	0.5	0.2
P75	3.9	5.5	5.2	2.0	1.4
Condensation sink (10^{-3} s^{-1})					
Mean	3.2	4.8	5.1	1.7	1.2
Median	2.6	3.9	4.0	1.2	0.7
STD	2.3	3.6	4.1	1.8	1.3
P25	1.6	2.3	2.1	0.5	0.3
P75	4.2	6.3	6.8	2.3	1.7
Geom. mean diameter (nm)					
Mean	60.0	74.9	67.0	78.6	69.1
Median	56.8	72.2	66.0	75.9	63.7
STD	27.3	30.2	24.3	35.3	31.9
P25	39.0	52.1	48.9	50.3	43.9
P75	77.8	94.0	83.0	104.2	90.4
CS diameter (nm)					
Mean	101.0	110.0	98.2	119.2	106.0
Median	100.0	109.2	96.5	120.1	102.9
STD	36.2	34.9	30.7	42.0	40.5
P25	73.7	84.1	75.8	87.6	74.2
P75	126.4	133.6	119.0	150.1	134.4
m					
Mean	1.68	1.69	1.67	1.70	1.68
Median	1.70	1.69	1.67	1.71	1.69
STD	0.07	0.06	0.06	0.07	0.07
P25	1.73	1.73	1.71	1.74	1.73
P75	1.65	1.66	1.63	1.67	1.65

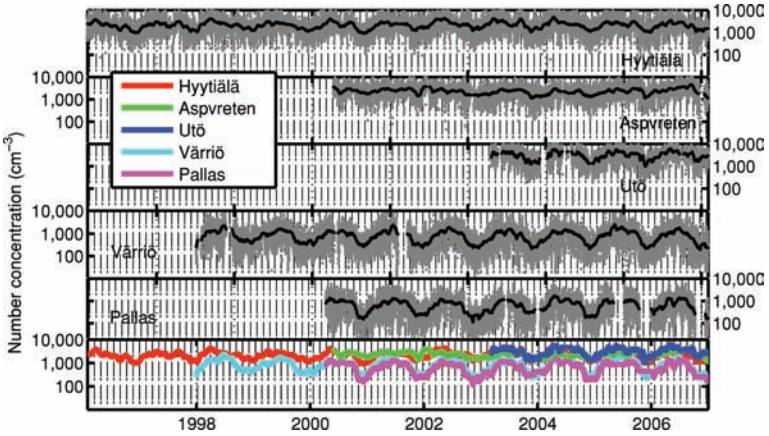

Fig. 2. Total number concentrations for each station, 2-h mean values and 2-month sliding averages (top five panels). Sixth panel: comparison of the sliding means for all five stations. The colour code for each station is given in the legend.

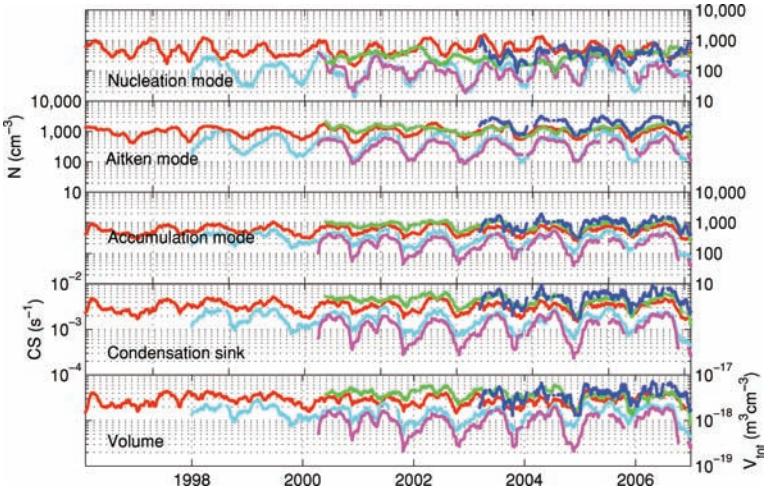

Fig. 3. Sliding 2-month means of the number concentrations in the nucleation mode (panel 1), Atiken mode (panel 2), accumulation mode (panel 3), condensation sink (panel 4) and total particle volume (panel 5). The colour code for each station is the same as in Fig. 1. (Red: Hyytiälä; Green: Aspvreten; Blue: Utö; Cyan: Pallas; Magenta: Värriö)

Lapland. The higher concentrations in Hyytiälä are most likely due to the fact that in Hyytiälä, smaller particles can be detected compared to the other stations. The nucleation mode concentration is highly variable, as can be seen from the difference between the median and mean concentrations, which are typically smaller by a factor of 2–3, and from the standard deviations, which are large compared to the mean concentrations.

The average Aitken mode concentrations in at the southern stations were between 1020 (Hyytiälä) and 1840 (Utö) cm^{-3}, while the Lapland stations measured average concentrations of 430 and 340 for Värriö and Pallas, respectively. The accumulation mode concentrations in the south were on average from the Hyytiälä 580 to 970 cm^{-3} at Utö, while the concentrations in Lapland were 300 and 230 for Värriö and Pallas, respectively.

By comparing the time series to each other, one can get an idea how well the measured concentrations represent a 'general' aerosol field. In Tunved et al. (2003) it was found that for short-time variations, it is difficult to predict aerosol characteristics from, for example, the source area of the air mass, as local sources and aerosol dynamic processes alter the size distribution very much on shorter timescales. However, when the data is averaged over timescales that are longer than the typical timescales of these processes, their effect is diminished and more general trends become visible.

From the data presented above one can see that the particle number concentrations at the three southern stations Hyytiälä, Aspvreten and Utö are very similar. The way the particles are distributed in the size ranges, however, is somewhat different. In Hyytiälä, the nucleation mode concentration is higher when compared to the other modes. Utö and Aspvreten have lower nucleation mode concentrations even if the effect of differing cut sizes is neglected. The Aitken and accumulation mode concentrations in Hyytiälä are lower than at the other southern stations. The number concentration at the Lapland stations is clearly lower than at the other three stations. The concentrations in all three modes are very similar between these two stations.

To get a measure of well the long-term number concentrations follow each other, we calculated the correlation coefficients (R) for the 2-month average concentrations. Table 3 we show the correlation coefficient table for the number concentrations at all the five stations. All correlation coefficients are very high,

Table 3. Correlation coefficients between the 2-month sliding mean number concentrations measured at the different stations

	Hyytiälä	Aspvreten	Utö	Värriö	Pallas
Hyytiälä	–	0.50	0.77	0.71	0.72
Aspvreten	0.50	–	0.66	0.59	0.60
Utö	0.77	0.66	–	0.85	0.83
Värriö	0.71	0.59	0.85	–	0.86
Pallas	0.76	0.60	0.83	0.86	–

representing a high probability that there is a relationship between the concentrations at each station. The highest correlation is found between the two Lappish stations, while they are less well correlated to the Hyytiälä and Aspvreten stations. The Aspvreten station shows the weakest correlation with all the other stations, but the time series correlates best with the Utö time series. This is expected, as both stations sample air that has spent some time over the Baltic Sea.

3.1.2. Particle volume concentration. The 2-month sliding average of the total particulate volume assuming spherical particles is shown in the lowest panel of Fig. 3. Here again, a clear difference between the Lapland stations and the other three stations can be observed. The mean volume concentrations at the Lapland stations are 1.5 and 1.0 $\mu m^3 cm^{-3}$ for Värriö and Pallas, respectively. In the south, the highest volume concentrations can be found at Aspvreten (4.0 μm^3 cm^{-3}) and the lowest at Hyytiälä (2.9 μm^3 cm^{-3}). The volume concentration fluctuates much more strongly than the number concentration. This was evident also when the correlation coefficients for the volume time series were calculated; using a shorter averaging window for the moving average increased the correlation coefficients. Despite the higher magnitude of the short-term variability, the volume concentration time series set showed similar characteristics as the number concentration, namely a quite clear separation between the Lapland stations and the more southern ones.

3.1.3. Other parameters. As mentioned before, also other characteristic parameters of the size distributions were calculated, such as the geometric mean diameter D_{pg}, the condensation sink and the condensation sink diameter (CSD), and the coagulation loss exponent m. Statistics for these parameters are given in Table 2.

At the Hyytiälä station, the geometric mean diameter was between 50 and 80 nm, being on average 60 nm. The value of D_{pg} at Aspvreten was on average higher, on average 75 nm, while Utö has an average D_{pg} of 67 nm. In Värriö and Pallas the mean values of D_{pg} were 79 and 69 nm, respectively. The values of D_{pg} in Lapland did not differ significantly from the other stations. Of the southern stations, Aspvreten usually has the larger particles, while in Lapland the particles are larger at Värriö than at Pallas. The average differences, however, are not significant. According to these results, the submicron size distribution over the Nordic background area is dominated by Aitken-mode particles; this is typical for a rural aerosol.

The 2-month sliding-average time series of the condensation sink is shown in panel 4 of Fig. 3. The distinction between the Lapland stations and the other stations is evident also in this data. The highest condensation sinks are observed in Utö, 5.1×10^{-3} s^{-1}, which corresponds to a condensing vapour characteristic lifetime of 3.3 min. Aspvreten has almost similar sinks, 4.8×10^{-3} s^{-1} (3.5 min). The Hyytiälä sinks are clearly lower, 3.2×10^{-3} s^{-1} (5.2 min), but not as low as in Lapland, where the sinks are 1.2×10^{-3} and 1.7×10^{-3} s^{-1} for Pallas and Värriö, respectively, yielding respective vapour lifetimes of 13.9 and 9.8 min. The lifetime estimates are made assuming a non-volatile vapour, meaning that the condensing vapour is assumed to have negligible vapour pressure and thus condense irreversibly on the particles.

The average condensation sink diameter was larger than the geometric mean diameter by 30–40 nm. Its magnitude was largest at the Värriö station (119 nm) and lowest at the Utö station (98 nm). Modellers using simple representations for submicrometre aerosol particles are advised to use this diameter instead of the geometric mean diameter, if coagulation or condensation of low saturation vapour pressure onto pre-existing particles or vapours is the main process being studied.

The power law exponent m in the expression to connect the nucleation rate to the rate of particle appearance in the size distribution was very similar at all stations. The sliding-average value ranged from -1.62 to -1.74, with station averages varying between -1.67 and -1.71. The standard deviations at all the stations are of the order of 0.05–0.07. There is some annual variation in the value of m, being higher during times with higher particle concentration; the magnitude of the variation, however, is less than 0.05. Approximating m with a constant value of -1.7 in models is thus proposed at least for the background aerosol types presented here. Applying eq. (1) for this value for an example case of calculating the coagulation sink of 1 nm particles from the calculated condensation sink gives $CoagS_{1nm} = 0.56 \times CS$.

3.2. *Annual trends*

3.2.1. Overview of annual trends. The averaged time series for the aerosol number and volume concentration showed clear structure despite the short-term variations having been lost to the averaging process. To investigate the annual trends more closely, we investigated the time series as a function of the day of the year. In order to remove the possible effects of a general trend towards higher or lower concentrations, we investigated the concentrations relative to the annual mean. For each year, we calculated the annual mean concentration and subtracted it from the 1-month sliding-average concentration. The resulting concentrations as a function of the day of year are shown in Fig. 4. The thin grey lines show the normalized concentrations,

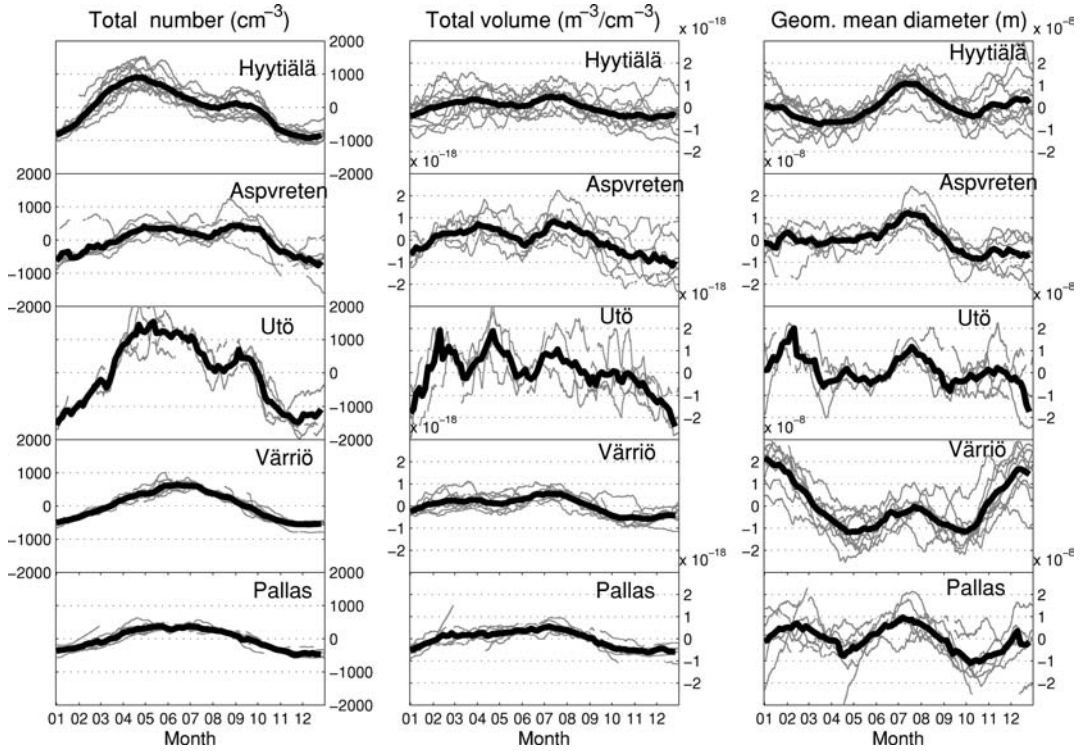

Fig. 4. The annual variation of total number concentration, total volume concentration and the geometric mean diameter. The grey lines are 1-month sliding averages with the yearly average subtracted; the black lines are the average of these values over all the years.

while the thick black lines show the normalized concentrations averaged over all the years that measurements were performed at the station.

The annual variation of the number concentration at the southern stations shows a bimodal structure, with maximum concentrations reached in springtime around April–May and another maximum in autumn, around September. The variation is less pronounced in Aspvreten than in Hyytiälä and Utö. At the Lapland stations the annual pattern is different. There, the number concentration has a maximum in summer and a minimum in winter. Breaking down the size distribution into the size ranges shows the picture a little more clearly; this is done in Fig. 5. The number concentrations in the nucleation mode show clear peaks in springtime in spring and autumn for the Hyytiälä and Utö stations, with minima in the summer and winter. The other three stations show less pronounced peaks, the autumn peaks visible only very weakly, with the Aspvreten station, surprisingly, showing no spring peak at all. In the Aitken mode, the southern stations all exhibit a two-peaked pattern, while the Lapland stations show highest concentrations in summer with only minor peaking in springtime. The accumulation mode numbers peak in summer at all stations, with a minor peak visible in springtime for the southern stations.

The difference in the annual concentration pattern between the accumulation mode and smaller particles is interesting when considering the CCN number. Particles larger than 90 nm in diameter are thought to be able to activate as cloud droplets. The fact that the number concentrations of these particles peaks in the summer at all stations, despite the total number peaks in spring and autumn for the southern stations, implies that the growth of the particles is the key factor in the CCN production. As shown by Tunved et al. (2006a,b), terpene emissions lead to particle mass formation over the boreal forest; these emissions have an annual cycle that peaks in summer. One cannot, of course, ignore the role of particle formation when considering CCN production: for one, the particles have in any case to be formed before they can grow to larger sizes. Also, the fact that the particles grow faster in summertime causes them to spend a shorter time in the nucleation and Aitken mode size ranges; this lowers their average number observed. By fully understanding this seasonality requires more knowledge of the plant emissions and their oxidation to form condensable vapours.

The annual variation of the total particle volume concentration is shown in Fig. 4, (centre panel), plotted similarly as that of the particle number concentration. The variation of the particle volume concentration is different from the number concentration variation. At all sites, a volume maximum occurs during summer, centred around July. All sites except Utö also exhibit a smaller maximum in springtime. None of the stations show any marked increase in particle volume during the autumn, when the secondary maximum in the particle number is occurring. Winter is the time of the least particle volume at all stations.

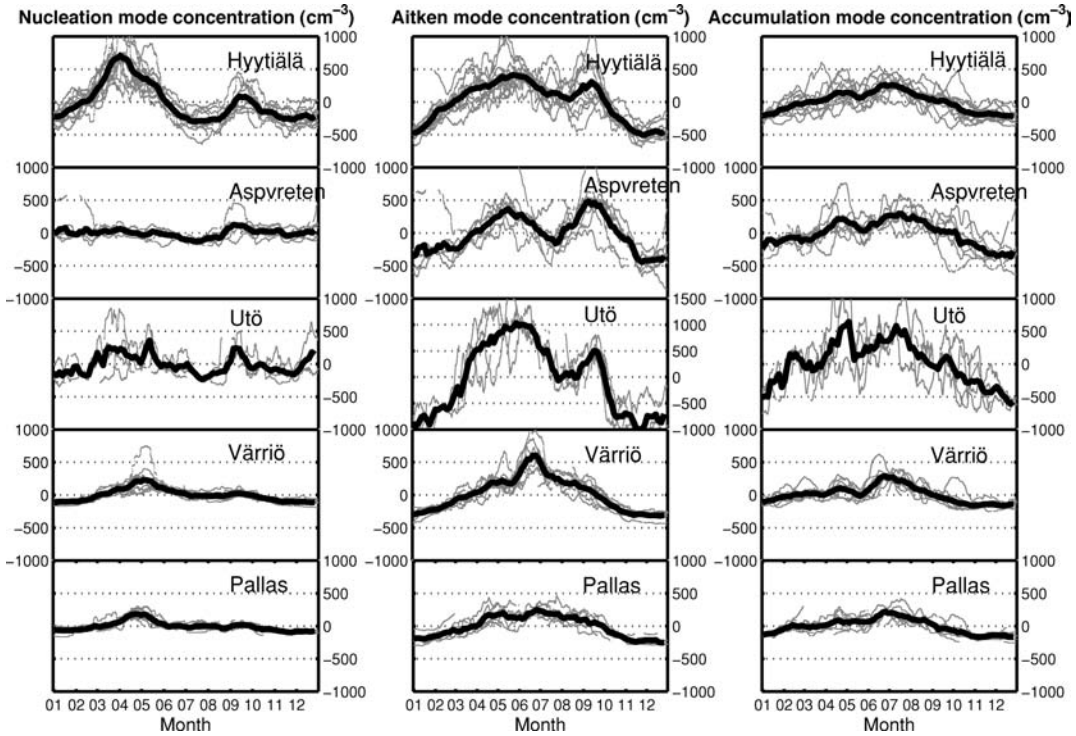

Fig. 5. The annual variation of the number concentration, in the nucleation, Aitken and accumulation size ranges. The grey lines are 1-month sliding averages with the yearly average subtracted; the black lines are the average of these values over all the years.

The annual variation of the geometric mean diameter shows a clear maximum in particle size during summer and two minima in spring and autumn. A maximum can also be found in wintertime; at most stations it is less pronounced than the summer maximum, however at Värriö the particles are clearly larger during wintertime. The annual variation of the condensation sink diameter is very similar to the number geometric mean diameter.

3.2.2. Comparison with nucleation event data. The north–south variation of the particle concentration has been reported previously also by Tunved et al. (2005) in a 1-yr study of the Nordic aerosol. Tunved et al. (2006b) also studied this increase in the special case of clean marine air passing over the Pallas and Värriö areas under clear sky conditions. In their study they found that boundary-layer nucleation events were the main factor controlling the particle number concentration and evolution during this southerly transport. Spracklen et al. (2006) used a global modelling approach to conclude that a significant number of the particles found in the troposphere are originated from regional new-particle formation events. Keeping this in mind, it is of interest to compare the observed number concentrations and their annual variation to the frequency of nucleation events observed at the respective stations. An analysis of the nucleation frequency was published in Dal Maso et al. (2007) for four of the stations discussed here. The defining feature of the annual distribution of the nucleation event occurrence was a maximum in spring, a local minimum in summer and a smaller maximum in September. Winter was a time of little formation activity.

We investigated the nucleation event fraction and compared it to the particle concentration. The nucleation event fraction we define as the number of nucleation event days in a 2-month moving window divided by the number of classified days in the same window. Thus, the nucleation event fraction is a number varying between 0 and 1. To eliminate differences between the stations, we normalized the number concentration to the maximum 2-month sliding-average value found, thus mapping the concentrations also on an axis between 0 and 1. The result of this analysis is shown in Fig. 6 for the four stations analysed in Dal Maso et al. (2007). In this figure, it is clearly seen that the total submicron aerosol number concentration is strongly correlated with the frequency of new-particle formation events. The correlation for the two data sets is very high ($R^2 = 0.65$). A linear fit to the data set gives a relationship of the fraction of the maximum number to the frequency of particle formation. It would be tempting to interpret the intercept at $x = 0$ (0.33) of this line as the fraction of aerosol number that would be present without particle formation; this would, however not be fully correct as atmospheric transport complicates the picture. It would, for example be fully possible for a station to observe no new-particle formation events at all and still measure particles formed in such events occurring elsewhere. The picture, however,

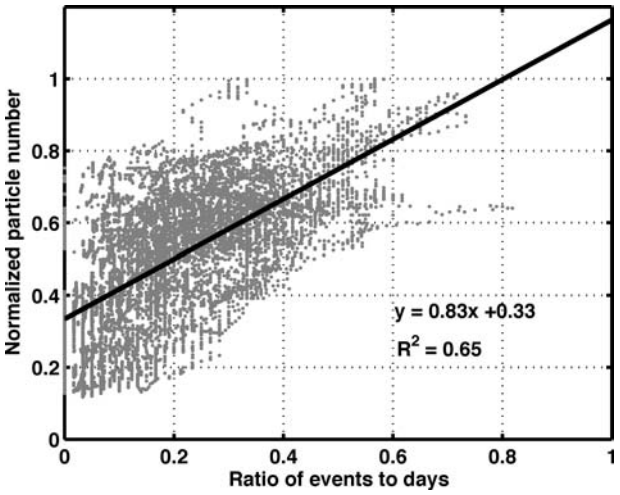

Fig. 6. The 2-month sliding average particle concentration (normalized to the maximum concentration) plotted versus the 2-month particle formation activity (number of event days in sliding 2-month window), along with a least-square fit to the data.

signals clearly that the importance of new-particle formation to the boreal aerosol number concentration is significant.

When investigating the seasonal variation of events it was found that the ratio of particle formation days to days with no particle formation was much greater at the southern stations (Hyytiälä and Aspvreten) than at the Lapland stations. Furthermore, is was found that although there was a summer minimum in the annual variation of the formation event frequency, the minimum was much less pronounced at the Lapland stations than at the Southern stations. Actually, in the summer months of June and July, the formation frequency is quite uniform over the whole Nordic area. The spring and autumn nucleation peak make up most of the difference in the nucleation event ratio to non-event days.

Despite this, the annual two-peak shape of the nucleation event fraction is still seen at the Lapland stations, but this feature is seen only very weakly in the number concentration at these stations, and only in the nucleation mode.

Considering these observations, it looks like the spring- and autumn-time particle production by nucleation is observed strongly only at the southern stations, and that it is much weaker in Lapland. The particle growth has been found to be seasonal, probably depending on both light as well as concentrations of condensable vapours, which are thought to be emitted by vegetation. This would explain the seasonal cycle of the particle volume concentration, with its summer peak, as the volume is increased by condensation. The nucleation cycle, however, remains partly unexplained.

Sogacheva et al. (2008) investigated the occurrence of new-particle formation events in Hyytiälä according to the synoptic situation and found that, at least for the three years investigated, cloudiness influenced the number of new-particle formation events. Nilsson et al. (2001) and Nilsson and Kulmala (2006) stressed the importance of both air mass origin and low pre-existing particle concentration, with Polar and Arctic air masses inducing most particle formation events. In Dal Maso et al. (2007) it was noted that particle formation was favoured in air masses advected from northwest, from the North Atlantic and/or Arctic Ocean. Hyvönen et al. (2005) also stressed the importance of low *CS* to particle formation. These explanations might work to explain the annual pattern in both the particle formation and the particle concentration at the southern stations, but at the Lapland stations they are not sufficient, especially regarding the condensation sink. While the origin plays a role by defining the properties of the air mass, it is these properties that define whether new particles are formed. One of the reasons that air masses from the NW have been found favourable for particle formation has been the low pre-existing aerosol concentration, characterized best by the condensation sink. The sink, however, is lower at the Lapland stations than at the sourthern stations, and therefore cannot be the limiting factor for the ultrafine particle source.

3.3. Interannual trends

In addition to studying the variation as a function of the time of the year, we also studied the variation of the particle number and volume concentrations between different years to find out whether the data set shows signs of a positive or negative trend in submicron aerosol concentration. This was done by calculating the arithmetic mean concentration as well as the 25th, 50th and 75th percentile for each year and comparing them to each other. The results are shown in Figs. 7 and 8, where box plots of the annual average concentrations are shown for all the stations. The line in the box shows the median concentration during the year, while the box covers the interquartile range of the yearly data.

We found that the average annual total concentration was variable, but not greatly so. In Hyytiälä, there is a minor rise in the particle concentrations from the year 2000 to 2003, after which the concentration steadily falls to 2006. The concentration variation is 400 $cm^{-3,}$ which is ca. 25% of the total concentration. In Aspvreten or Utö this rise is not observable. The Lapland stations follow each other, as expected.

The total volume concentration is shown on the right-hand panels of Fig. 7. In Hyytiälä, the total volume concentration seems to oscillate slightly, with a maximum in 1998 and a minimum in 2003–2004, after which the volume concentration rises again. This rise is seen also in the Utö data set. The volume concentration in Värriö and Pallas seems to replicate this oscillation, having a minimum in the same years; the oscillation, however, is quite small. The Aspvreten data does not show the same pattern.

A breakdown of the number concentration into the three different size ranges is shown in Fig. 8. The nucleation mode number concentration variation is very similar to the total number

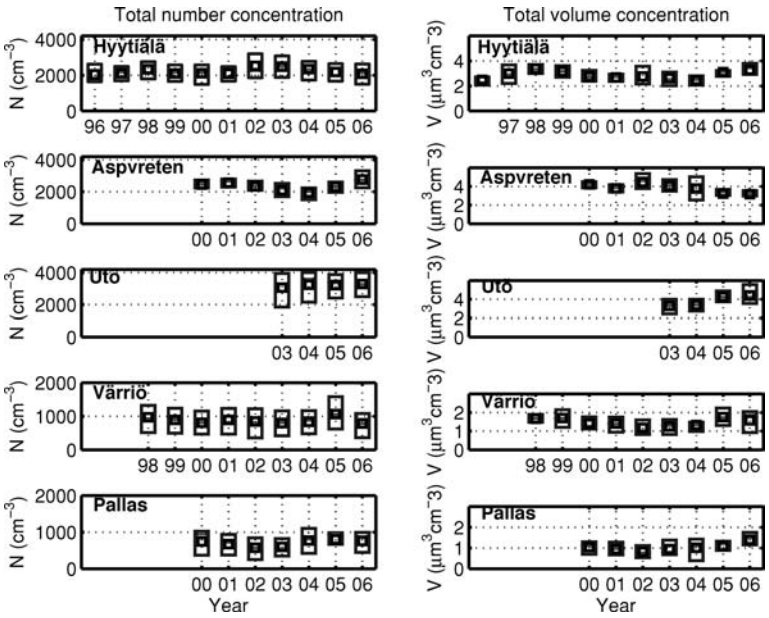

Fig. 7. Boxplots of the annual particle number and volume concentrations at the five studied stations. The boxes represent the interquartile (P_{25}–P_{75}) range, the line in the box is the median and the square marker stands for the arithmetic mean value.

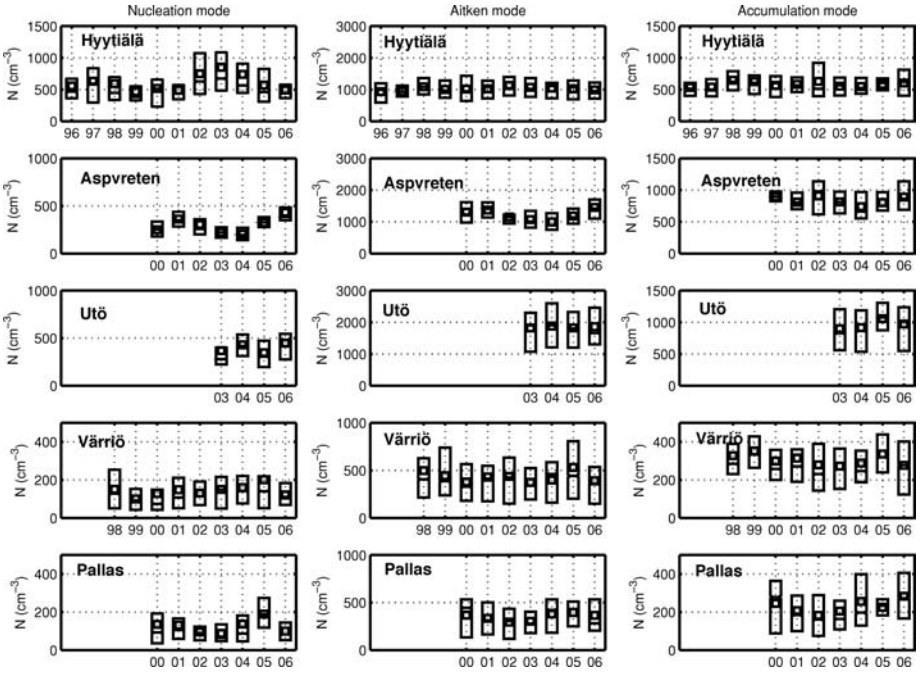

Fig. 8. Boxplots of the annual particle number concentrations in the nucleation, Aitken and accumulation size ranges at the five studied stations. The boxes represent the interquartile (P_{25}–P_{75}) range, the line in the box is the median and the square marker stands for the arithmetic mean value.

concentration at the Hyytiälä and Aspvreten stations. At the Utö station, the nucleation mode concentration does not follow the variation in the total number concentration. Similar behaviour can be seen for the Lapland stations, although not as clearly. At all stations, the nucleation mode concentration is clearly positively correlated to the total number; the correlation is the highest at the Hyytiälä station (0.77) and lowest at the Utö station (0.28). At the other three stations the correlation coefficient was 0.5–0.6. The number concentration in the Aitken mode also follows the total number concentration. The correlation coefficient between the Aitken mode and total number was very high at all stations, being the lowest at Hyytiälä (0.77) while at the other stations it was 0.88–0.91. This shows that the total number variation is mostly governed by the variation in the Aitken mode, as is expected. The accumulation mode concentrations are less well correlated to the total number. The best correlation

between total and accumulation mode numbers can be observed at the Aspvreten and Utö stations ($R^2 = 0.62$ and 0.67), while the lowest is observed in Hyytiälä (0.30).

Again, no clear trends could be found in the annual averages. The annual mean nucleation mode concentrations showed the largest variation between the years, but we could detect no systematic increase or decrease. The variation in the Aitken and accumulation mode concentration was small, especially the accumulation mode concentration of particles did not show any signs of long-term variation.

3.4. Comparison to other time series

There are few longer time series observations of continental submicron aerosol number concentrations available in the literature. One source for comparison is the paper of Sheridan et al. (2001), where a 4-yr data set is presented from continental USA. The mean number concentration observed by them was 4500 cm^{-3} (10 nm–3 μm), which is clearly higher than in our study. What is noteworthy is that the annual variation of the total concentration replicates the pattern observed in our study, with maximum number concentrations found in March–April and October. When only larger particles (100 nm–10 μm) are counted, the pattern changes to one with a maximum in summer, July–August. The authors attribute this to wind-blown dust episodes or agricultural burning; however, in the light of our data one might present the hypothesis that also here particle formation events might have an influence on the particulate number below 100 nm, while the enchanced condensation in summer increases the number of >100 nm particles.

Birmili et al. (2001) report in their 1.5 yr study total number concentrations between 4000 and 5000 cm^{-3}, with nucleation mode concentrations of 630–2010 cm^{-3}, Aitken mode concentrations of 2300–4900 cm^{-3}, and accumulation mode concentrations 500–1200 cm^{-3}. These are all again clearly higher than in this study, which is natural considering their environment of semi-rural central European boundary layer, which is much more polluted than any of the sites studied here.

One interesting comparison can be done with the study of Ström et al. (2003). They measured particle size distributions for one year at Svalbarg, an archipelago situated north of Scandinavia. They found an annual variation in the size distribution, with a quite sudden decrease of the particle size in summertime. The particle concentration varies with incoming solar radiation, being ca. 50 cm^{-3} in the dark winter and rising to ca. 1500 cm^{-3} in high summer. They also reject the condensation sink as a controlling factor of particle formation and rule out anthropogenic influence. They propose the Siberian tundra as a source for precursor gases for particle formation. Our annual cycle of particle number and size is similar to the Ström data, with the exception that our data shows a local particle size maximum in midsummer, which is not observable (or much smaller) in the Ström data. This could be caused by the Lapland stations' closer proximity to the precursor source, which could be either the boreal forest or the Siberian tundra.

It is also of interest to compare our concentration annual patterns to the Arctic haze, the distinctive Arctic aerosol which has a clear annual cycle (Law and Stohl, 2007). It has a maximum in late winter–early spring (February–April) and a strong minimum in summer (June–October) (Sharma et al., 2006). This is almost exact opposite of the seasonality of our Lapland data. Arctic haze is well-aged aerosol, conserved in the atmosphere due to slow removal processes. Our data, in contrast, describes a relatively fresh aerosol produced with the aid of photochemistry. The interaction of the Arctic haze with particle formation should be investigated in more detail.

4. Conclusions

We investigated five time series of boreal rural submicron particle concentrations and size distribution parameters for Nordic background stations with the objective of finding possible trends in the particle number and volume concentrations as well as establish a quantitative picture of the aerosol number and mass concentrations and other parameters of the aerosol size distributions. The time series covered the years 1996–2006, with the period 2003–2006 covered by all stations and 2000–2006 by four out of the five stations. The stations consist of two southern stations at the Baltic Sea (Aspvreten and Utö), one in southern mainland Finland (Hyytiälä) and two in the Finnish Lapland (Värriö and Pallas).

The size distribution characteristics investigated were particle total number and volume concentration as well as concentrations in the nucleation, Aitken and accumulation size ranges. In addition, we also presented time series of geometric mean size as well as aerosol dynamic parameters such as the condensation sink, condensation sink diameter, and the coagulation loss power law exponent m.

Statistics of these parameters, which are key aerosol dynamic characteristics covering the condensational growth of particles as well as the coagulational loss of freshly nucleated particles, are expected to be of interest in studies attempting to model the formation of aerosol number and mass over the Nordic area. We used long averaging windows in our analysis to reduce the influence of small-scale variations and sources, in order to obtain data representing Nordic background aerosol concentrations. The condensation sink diameter was found to be significantly higher than the geometric mean diameter of the measured aerosol, of the order of 100–120 nm compared to the geometric mean diameters of 50–80 nm. Therefore we suggest that models that represent submicron aerosol with a single monodisperse mode should use the condensation sink diameter if condensation is the main process being studied. The power law exponent m was found to be quite close to the value 1.7, with little variation. This result makes estimating coagulation losses of small particles easier, because the coagulation loss rates of particles can

be estimated simply by calculating only the condensation sink value and assuming m to be constant.

The time series data investigated can be used describe a 'typical' Nordic boreal aerosol, averaged over longer time periods. The main features found in this study are a north–south concentration difference in both particle number and volume. At the southern stations of Hyytiälä and Aspvreten, the typical aerosol concentration is 1500–3000 cm^{-3}, while in Lapland, the typical aerosol number is much lower, about 700–900 cm^{-3}. In the south, he particle volume is on average 3–4 $\mu m^3 m^{-3}$, peaking in summer with ca. 4 $\mu m^3 m^{-3}$ and a minimum in winter. In Lapland, the particle volume is on average 1–1.5 $\mu m^3 m^{-3}$.

The annual pattern for the particle number also exhibited a north–south difference. At the southern stations, the number concentration showed clear and strong peaks in spring and autumn, with a minimum in winter and a local minimum in summer. At the Lapland stations, the total number concentration showed only one minimum and maximum, in winter and summer, respectively. We presume this difference to be a result of much stronger particle formation during the spring and autumn nucleation maxima at the southern stations. Generally, the particle number concentration was strongly correlated to particle formation frequency, with indications that particle formation events are the main source of submicron aerosol number over Nordic background areas.

The clear variation in found in the annual pattern of the volume concentration with a peak in July and a minor minimum or plateau in late May is caused by the growth of the average diameter of the particles, which offsets the decrease of the particle number at the southern stations. This effect can be interpreted as a shift between two aerosol dynamic processes, nucleation and condensation. Nucleation activity is known to be reduced in the summer (Dal Maso et al., 2007) while the condensational growth of particles is stronger. This has been known to be the case for freshly nucleated particles; our results show that it applies more generally. The condensational growth is linked to available condensable vapours; Tunved et al. (2006a,b) showed that particle volume in summertime is linked to forest monoterpene emissions accumulated into an air parcel. In this light, the annual pattern of particle volume can easily be understood to be an end result of the annual pattern of biogenic volatile organic compound emissions, which are light- and temperature-dependent. The stronger growth is also reflected in the concentration of accumumulation mode particles (>90 nm) which also peaks in summertime. As these particles are considered to be large enough to be activated as cloud droplets, this has implications also for CCN concentrations.

We investigated the interannual variation of particle number in different size ranges and also the volume concentration. The interannual variation of all these parameters was found to be significant. The variation was, however, site-specific; for example the Hyytiälä number concentrations showed similar variations as the nucleation event numbers. No general trend of increasing or decreasing particle numbers could be found over this network of Nordic background stations, even though we were specifically searching for one. The decrease of sulphate emissions in Europe is, according to our observations, not reflected in the submicron particle number and volume. The particle number and volume seem to be influenced rather by the regional, 'natural' particle formation and growth activity.

References

Aalto, P., Hämeri, K., Becker, E., Weber, R., Salm, J. and co-authors. 2001. Physical characterization of aerosol particles during nucleation events. *Tellus* **53B**, 344–358.

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

Andreae, M. O. 2007a. Atmospheric aerosols versus greenhouse gases in the twenty-first century. *Philos. Trans. Roy. Soc. A: Math., Phys. Eng. Sci.* **365**(1856), 1915–1923

Andreae, M. O. 2007b. Aerosols before pollution. *Science* **315**, 50–51.

Berglen, T. F., Myhre, G., Isaksen, I. S. A., Vestreng, V. and Smith, S. J. 2007. Sulphate trends in Europe: are we able to model the recent observed decrease? *Tellus* **59B**, 773–786.

Birmili, W., Wiedensohler, A., Heintzenberg, J. and Lehmann, K. 2001. Atmospheric particle number size distribution in central Europe: statistical relations to air masses and meteorology. *J. Geophys. Res Atmos.* **106**(D23), 32005–32018.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T. and co-authors. 2005. Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä. *Finland Boreal Environ. Res.* **10**, 323–336.

Dal Maso, M., Sogacheva, L., Aalto, P. P., Riipinen, I., Komppula, M. and co-authors. 2007. Aerosol size distribution measurements at four Nordic field stations: identification, analysis and trajectory analysis of new particle formation bursts. *Tellus* **59B**, 350–361.

Engler, C., Lihavainen, H., Komppula, M., Kerminen, V.-M., Kulmala, M. and co-authors. 2007. Continuous measurements of aerosol properties at the Baltic Sea. *Tellus* **59B**, 728–741.

Garrett, T. J., Radke, L. F. and Hobbs, P. V. 2002. Aerosol effects on cloud emissivity and surface longwave heating in the arctic. *J. Atmos. Sci.* 59, 769–778.

Goldberg, M. S., Burnett, R. T., Bailar, J. C., Brook, J., Bonvalot, Y. and co-authors. 2001. The association between daily mortality and ambient air particle pollution in Montreal, Quebec. 2. cause-specific mortality. *Environ. Res.* **86**, 26–36.

Hari, P., Kulmala, M., Pohja, T., Lahti, T., Siivola, E. and co-authors. 1994. Air pollution in Eastern Lapland: challenge for an environmental measurement station. Silva Fennica. 29–39.

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

Hatakka, J., Aalto, T., Aaltonen, V., Aurela, M., Hakola, H. and co-authors. 2003. Overview of the atmospheric research activities and results at Pallas GAWstation. *Boreal Environ. Res.* **8**(4), 365–384.

Hyvärinen, A.-P., Komppula, M., Engler, C., Kivekäs, N., Kerminen, V.-M. and co-authors. 2008. Atmospheric new particle formation at Utö, Baltic Sea 2003–2005. *Tellus B*, in press.

Hyvönen, S., Junninen, H., Laakso, L., Dal Maso, M., Grönholm, T. and co-authors. 2005. A look at aerosol formation using data mining techniques. *Atmos. Chem. Phys.* **5**, 3345–3356.

IPCC. 2007. Summary for policymakers. In: *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change* (eds S. Solomon, D. M. Qin, Z. Manning, M. Chen, K. B. Marquis, M. T. Averyt and H. L. Miller). Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Kerminen, V.-M., Lihavainen, H., Komppula, M., Viisanen, Y., Kulmala, M. 2005. Direct observational evidence linking atmospheric aerosol formation and cloud droplet activation. *Geophys. Res. Lett.* **32**, L14803, doi:10.1029/2005GL023130.

Komppula, M., Dal Maso, M., Lihavainen, H., Aalto, P. P., Kulmala, M. and co-authors. 2003. Comparison of new particle formation events at two locations in northern Finland. *Boreal Environ. Res.* **8**, 395–404.

Kulmala M., Rannik, Ü., Pirjola, L., Dal Maso, M., Karimäki, J. and co-authors. 2000. Characterization of atmospheric trace gas and aerosol concentrations at forest sites in southern and northern Finland using back trajectories. *Boreal Environ. Res.* **5**, 315–336.

Kulmala, M., Dal Maso, M., Mäkelä, J. M., Pirjola, L., Väkevä, M. and co-authors. 2001. On the formation, growth and composition of nucleation mode particles. *Tellus B* **53**, 479–490.

Kurtén, T., Kulmala, M., Dal Maso, M., Suni, T., Reissell, A. and co-authors. 2003. Estimation of different forest-related contributions to the radiative balance using observations in southern Finland. *Boreal Environ. Res.* **8**, 275–285.

Laakso, L., Hussein, T., Aarnio, P., Komppula, M., Hiltunen, V. and co-authors. 2003. Diurnal and annual characteristics of particle mass and number concentrations in urban, rural and Arctic environments in Finland. *Atmos. Environ.* **37**(9), 2629–2641.

Law, K. S. and Stohl, A. 2007. Arctic air pollution: origins and impacts. *Science* 315(5818), 1537–1540.

Lehtinen, K. E. J., Korhonen, H., Dal Maso, M. and Kulmala, M. 2003. On the concept of condensation sink diameter. *Boreal Environ. Res.* **8**, 405–411.

Lehtinen, K. E. J., Dal Maso, M., Kulmala, M. and Kerminen, V.-M. 2007. Estimating nucleation rates from apparent particle formation rates and vice versa: Revised formulation of the Kerminen-Kulmala equation. *J. Aerosol Sci.* **38**, 988–994.

Lihavainen, H., Kerminen, V.-M., Komppula, M., Hatakka, J., Aaltonen, V. and co-authors. 2003. Production of "potential" cloud condensation nuclei associated with atmospheric new-particle formation in northern Finland. *J. Geophys. Res.* **108**(D24), 4782.

Mäkelä, J. M., Aalto, P., Jokinen, V., Pohja, T., Nissinen, A. and co-authors. 1997. Observations of ultrafine aerosol particle formationand growth in boreal forest. *Geophys. Res. Lett.* **24**, 1219–1222.

Nilsson E. D. and Kulmala, M. 2006. Aerosol formation over the Boreal forest in Hyytiälä, Finland: monthly frequency and annual cycles—the roles of air mass characteristics and synoptic scale meteorology. *Atmos. Chem. Phys. Discuss.* 6, 10425–10462.

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

Sharma, S., Andrews, E., Barrie, L. A., Ogren, J. A. and Lavoué, D. 2006. Pan-Arctic enhancements of light absorbing aerosol concentrations due to North American boreal forest fires during summer 2004. *J. Geophys. Res.* **111**, doi:10.1029/2005JD006581.

Sheridan P. J., Delene, D. J. and Ogren, J. A. 2001. Four years of continuous surface aerosol measurements from the Department of energy's atmospheric radiation measurement program southern great plains cloud and radiation testbed site. *J Geophys. Res. Atmos.* **106**(D18), 20735–20747.

Slaughter, J. C., Lumley, T., Sheppard, L., Koenig, J. Q. and Shapiro, G. G. 2003. Effects of ambient air pollution on symptom severity ond medication use in children with asthma. *Ann. Allergy Asthma Immunol.* **91**, 346–353.

Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V.-M., Mann, G. W. and co-authors. 2006. The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales. *Atmos. Chem. Phys.* **6**, 5631–5648.

Sogacheva, L., Saukkonen, L., Nilsson, E. D., Dal Maso, M., Schultz, D. M. and co-authors. 2008. New aerosol particle formation in different synoptic situations at Hyytiälä, Southern Finland. *Tellus B*, doi: 10.1111/j.1600-0889.2008.00364.x.

Stolzenburg, M. R. 1988. An ultrafine aerosol size distribution measuring system. PhD Thesis, University of minnesota, minneapolis MN, USA.

Ström, J., Umegard, J., Torseth, K., Tunved, P., Hansson, H.-C. and co-authors. 2003. One year of particle size distribution and aerosol chemical composition measurements at the Zeppelin Station, Svalbard, March 2000-March 2001. *Phys. Chem. Earth* **28**, 1181–1190.

Tunved, P., Hansson, H.-C., Kulmala, M., Aalto, P., Viisanen, Y. and co-authors. 2003. One year boundary layer aerosol size distribution data from five nordic background stations. *Atmos. Chem. Phys.* **3**, 2183–2205.

Tunved, P., Nilsson, E. D., Hansson, H.-C., Ström, J., Kulmala, M. and co-authors. 2005. Aerosol characteristics of air masses in northern Europe: Influences of location, transport, sinks, and sources. *J. Geophys. Res.* **110**, D07201, doi:10.1029/2004JD005085.

Tunved, P., Hansson, H.-C., Kerminen, V.-M., Ström, J., Dal Maso, M. and co-authors. 2006a. High natural aerosol loading over boreal forests. *Science* **312**(5771), 261–263.

Tunved, P., Korhonen, H., Ström, J., Hansson, H.-C., Lehtinen, K. E. J. and co-authors. 2006b. Is nucleation capable of explaining observed aerosol integral number increase during southerly transport over Scandinavia? *Tellus* **58B**, 129–140

Twomey, S. 1991. Aerosols, clouds and radiation. *Atmos. Environ.* **25A**, 2435–2442.

Wiedensohler, A. 1988. An approximation of the bipolarcharge distribution for the particles in the submicron size range. *J. Aerosol. Sci.* **19**, 387–389.