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Aerosol size distribution measurements at four Nordic field stations: identification, analysis and trajectory analysis of new particle formation bursts

By MIIKKA DAL MASO[1*], LARISA SOGACHEVA[1], PASI P. AALTO[1], ILONA RIIPINEN[1], MIKA KOMPPULA[2], PETER TUNVED[2], LAURA KORHONEN[1], VILLE SUUR-USKI[1], ANNE HIRSIKKO[1], THEO KURTÉN[1], VELI-MATTI KERMINEN[2], HEIKKI LIHAVAINEN[2], YRJÖ VIISANEN[2], HANS-CHRISTEN HANSSON[3] and MARKKU KULMALA[1], [1]*Department of Physical Sciences, University of Helsinki, P.O. Box 64, 00014 Helsinki, Finland;* [2]*Finnish Meteorological Institute, P.O. Box 503, 00101 Helsinki, Finland;* [3]*Department of Applied Environmental Science, Stockholm University, SE-10691 Stockholm, Sweden*

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ABSTRACT

We analyzed aerosol size distributions from the Finnish measuring stations at Hyytiälä, Värriö and Pallas and the Swedish station at Aspvreten over a period of several years. We identified occurrences of new particle formation bursts and obtained characteristics for the bursts from the size distribution data. In addition, we analyzed the directions from which air masses leading to new particle formation arrived. We found that new particle formation occurs over the whole area covered by the measurement stations. The Northern Atlantic is dominating as a source for air leading to new particle formation at all of the analyzed stations. The formation occurrence had a similar annual variation at all the stations, with peaks in springtime and autumn and minima in winter and summer. The ratio of event days to non-event days had a North-South dependence, with northern stations having lower event ratios. Particle growth rates ranged from 0.5 to 15 nm/h, with the mean growth rate being slightly higher at the southern stations. Southern stations also had a stronger particle source, on average 0.5 cm^{-3} s^{-1}, compared to the northern stations (0.1 cm^{-3} s^{-1}). Based on our analysis, it is evident that new particle formation occurs often in whole Nordic boreal forest area when air is transported from the North Atlantic, and that the same process or processes are very probably responsible for the formation over the whole area.

1. Introduction

Measurements of submicrometer aerosol size distributions have shown that new tropospheric aerosol formation and growth bursts are widespread, and at least at some sites a frequent phenomenon in the Earth's atmosphere (Kulmala et al., 2004a). The particles formed by this process have the potential to grow to sizes where they can activate as cloud condensation nuclei (CCN, see Charlson et al., 1992). This has prompted increasing activity in this field of study, both in trying to understand the physical and chemical pathways causing the formation of new particles (Kulmala et al., 2000, 2004b; McMurry et al., 2005), and assessing the climatic importance of the particles formed (see for example Kurten et al., 2003; Kerminen et al., 2005). One difficulty in these studies has been that most reports of new particle formation (NPF) in the literature cover quite short time periods and/or limited geographical areas. Because new particle formation is a process strongly dependent on local and regional meteorological conditions, measurements that cover a sufficiently long time and larger areas are needed, so that features emerging from local conditions can be separated from the larger-scale phenomena.

In this study we analyze continuous size distribution data measured at four different stations in Finland and Sweden, all situated within the boreal forest. The time series obtained range from five to ten years in length. The boreal forest has been shown by Tunved et al. (2006) to participate in the growth of submicrometer particles by producing volatile organic compounds (VOC) which in turn can oxidize to form low vapour pressure species which condense on the aerosol. We analyze the measured size distributions to find frequency of NPF events in the boreal forest and also the basic characteristics of these bursts. Reports of NPF frequency and characteristics in the boreal forest are

*Corresponding author.
e-mail: miikka.dal@helsinki.fi
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Table 1. An overview of the data used for this study.

Site	Latitude	Longitude	DMPS range*	Start
SMEAR I, Värriö	67°46'N	29°35'E	8–460 nm	1.1.1998
SMEAR II, Hyytiälä	61°51'N	24°17'E	3–500 nm	30.1.1996
Sammaltunturi, Pallas	67°58'N	24°07'E	10–490 nm	12.4.2000
Aspvreten,Sweden	58°46'N	17°24'E	10–452 nm	31.5.2000

*Narrowest range covered during analysis period

available (Mäkelä et al., 1997; Dal Maso et al., 2005; Vehkamäki et al., 2004), for two of the stations (Hyytiälä and Värriö) studied here, and Komppula et al. (2005) reported size distribution data with regard to cloud droplet activation. Tunved et al. (2003) reported characteristics of the observed aerosol characteristics for five Nordic stations for one year. However, no direct comparison or NPF climatology exists yet for this Nordic network of stations over an extended time period. Here we also report additional data added to the time series since the publication of the above-mentioned reports.

Using robust methods for identifying NPF days and obtaining the characteristics on long time series of data we aim to find the similarities and differences of NPF over the Nordic boreal area. We also take a look at the air masses arriving at the stations on NPF burst days. By doing so, we also aim to find whether the same processes can be assumed to be responsible for the appearance of the particles at all stations.

The work presented here consists of three main parts: first, identifying days containing a NPF burst or no burst; second, obtaining the characteristics of the identified bursts; and third, investigating the main directions of air mass advection that characterize days with NPF or no NPF. The methodology used here for NPF event identification and analysis is the same as presented in Dal Maso et al. (2005), and it is only summarized here, the main focus being on the results of the analysis and the comparison of the different stations.

2. Materials and methods

2.1. Measurement sites

In this paper we present an analysis of aerosol size distribution data measured at four field stations, all situated inside the boreal forest: 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.

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. An overview of the stations is given in Table 1.

2.1.1. Hyytiälä. The SMEAR II station at Hyytiälä (61°51′N 24°17′E, 180 m a.s.l.) has extensive facilities for measuring forest-atmosphere relations and has been doing so since 1996 (see for example Hari and Kulmala, 2005). Aerosol size distribution measurements were started with a twin-DMPS system at the end of January 1996, and have been ongoing since then. The DMPS measures submicrometer particles of sizes larger than 3 nm. The sampling is done at 2 m above ground inside the forest canopy. The DMPS system was moved from its original location by *ca.* 100 m in 2004.

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).

2.1.2. Aspvreten. 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. Anthropogenic influences are considered small (Tunved et al., 2003) as the surrounding area is not densely populated. Aerosol size distribution measurements are performed with a single-DMA and -CPC DMPS system, resulting in size distribution of particles in the size range 10–450 nm every 6 min.

2.1.3. Värriö. 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 years old in the station's immediate vicinity. The measurements are performed on a hill top (Hari et al., 1994). There are no pollution sources nearby, but emissions from industrial activities (e.g., smelters) from the Kola Peninsula area may be carried to the station.

The aerosol size distribution measurement system consisted of a single DMA and CPC measuring particles from sizes 8–460 nm until April 2003, when a twin-DMPS system was installed, decreasing the minimum observable particle size to 3 nm. The sampling is done at 2 m above ground inside the forest canopy. In addition to these measurements the station records a range of atmospheric parameters, including trace gas concentrations along with temperature, relative humidity (RH), solar radiation and wind speed. The station has been measuring size distributions since December 1997; in this study we analyze data starting in January 1998.

2.1.4. *Pallas.* 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 top of a field in western Lapland (Hatakka et al., 2003). The vegetation in the immediate vicinity of the station consists of mixed pine, spruce and birch forest. The station itself is above the tree line and the sampling is done 7 m above ground. 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. The DMPS system measures particles with sizes from 7–500 nm since April 2000.

2.2. Data treatment

Our analysis of new particle formation consists of the identification of NPF bursts, obtaining characteristics like the growth and formation rates of new particles, and a basic trajectory analysis to find geographic directions that might favor NPF bursts. A breakdown of the performed analysis is given in the following sections.

2.2.1. Identification of NPF bursts. The identification of days containing NPF bursts was based on the principles and methods presented in Dal Maso et al. (2005), which we will now summarize.

The time series of the size distributions were inspected visually, and a classification of days was made based on this analysis. Days were classified either as event days, containing a clear new particle formation burst; non-events, where no sign of new ultrafine particle appearance was measured, or undefined days, where some ultrafine particles were present but it was not evident that new particle formation was taking place.

For a day to qualify as an event day, a new mode of particles had to appear in the size distribution in the nucleation mode (<25 nm) and prevail continuously for more than an hour. Additionally, the size of the new particles needed to increase over time; this was justified by our aim to identify particle formation occurring over a large region, and thus producing a significant amount of tropospheric aerosol. It is also a means of eliminating local pollution sources from the analysis. The growth requirement effectively increased the time scope for the prevailing new mode to significantly more than one hour, as it was difficult to detect growth at such a short timescale. These criteria are geared towards detection of new particle formation taking place in the whole sampled air mass, on a regional scale. Assuming a horizontal advection speed of 30–40 km/h, this scale could be up to hundreds of kilometers.

Event days were further classified into separate classes regarding the possibility to derive the characteristics from the size distribution; see explanation in section 2.2.2.

If no mode of <25 nm particles was detected during a day, the day was classified as a non-event day. Short bursts of ultrafine particles lasting 10–30 min did not remove a day from this category.

A day where nucleation particles were present but other criteria for a NPF event were not fulfilled (for example no growth or continuity, or in some cases an existing nucleation mode decreasing in size and then growing again) were classified as 'undefined'. The reason for this classification is to remove these days for future analysis where the event-nonevent distinction is crucial. Days where we suspected that new particle formation had been taking place in the air mass prior to its arrival to the station, characterized by a growing tail of a former nucleation mode, were also classified as undefined.

The classification was performed in groups of three people or more, two of which participated in the analysis of all four stations. The classification was done in two stages: first in a day-by-day manner, and in the second phase the classes were checked separately for consistency. For a day to be classified as an event or non-event, all group members had to agree on the classification; otherwise, the day was marked as undefined.

2.2.2. Burst analysis. Obtaining the formation and growth rates and other characteristics of the new particle formation bursts was also done following the treatment in Dal Maso et al. (2005), summarized below. The size distributions were parametrized by a least-square log-normal fitting method, yielding parameters of 2–3 log-normal modes. The growth rate (GR $= \frac{dD_{p,g}}{dt}$, given in nm/h) was obtained by least-square fitting of a first-order polynomial to the geometric mean diameter $D_{p,g}$ time series of the new mode.

The observed change in new particle concentration ($\frac{dN_{nuc}}{dt}$) was obtained in two ways: a) as the time change of <25 nm particle concentration (N_{min-25}) between the start of particle appearance and the peak in N_{min-25} and b) as the time change of the modal concentration (N_{fit}) obtained by the log-normal fitting. While the fitting method performed very well with respect to $D_{p,g}$, the fitted number concentrations were sometimes grossly overestimated. This was the case especially at the very start of a nucleation burst, when only the edge of a new mode could be measured. Method b) with $N_{nuc} = N_{fit}$ is more exact as one does not have to worry of the loss of particles due to growth out of the size range; however, due to the limitations described above we predominantly used method a). Only on days where growth clearly diminished N_{min-25} with respect to N_{fit} we used method b). We would also like to note that the results obtained by the different methods were within *ca.* 10% of each other when both of the above-mentioned conditions were absent. The observed formation rate of new particles (J_{obs}) was calculated according to

$$J_{obs} = \frac{dN_{nuc}}{dt} + F_{coag} \tag{1}$$

where F_{coag} is the coagulation loss rate of the new particles, given by $F_{coag} = N_{nuc}K_{nuc}$, where K_{nuc} is the coagulation sink for nucleation mode particles caused by pre-existing aerosol (Kulmala et al., 2001). This could be calculated from the size distribution data, using data corrected for particle hygroscopicity (Laakso et al., 2004). The hygroscopicity correction was applied because the ambient air is dried before the DMPS systems measures the size distributions.

Because the minimum detectable particle size is different for different stations, the formation rates cannot be considered equivalent. This is because as the particles grow, a part of the population is lost due to coagulation (see e.g. Kerminen and Kulmala (2002)). The difference of detection size is quite small for the Värriö, Pallas and Aspvreten stations; the Hyytiälä station, however, has a significantly lower detection cut-off. To compare the formation rates, we calculated the coagulation loss rate of particles from between 3 and 10 nm for the Hyytiälä data. This could then be used to estimate equivalent formation rates with the other stations.

With some assumptions of the aerosol dynamics the concentration and source rate of condensing vapour can be estimated provided that the condensation sink (Kulmala et al., 2001) and growth rate are known. Here the condensation sink was calculated using the same hygroscopicity correction as for the coagulation sink.

Even though the methods to obtain the formation and growth rates are quite robust, the fluctuating nature of the atmosphere made it impossible to reliably derive these parameters for all classified NPF days. Therefore, days where fluctuations or pre-existing aerosols interfered with the analysis were classified as class II event days, whereas days where the parameters could be obtained were classified as class I. The results for NPF characteristics presented here therefore only include class I days.

2.2.3. Trajectories. Ninety six-hour back trajectories at 100-m arrival height above ground level at 10.00 UTC (12.00 Finnish winter time) were calculated using the HYSPLIT_4 model, developed by NOAA/ARL (Draxler and Hess, 1998). The model runs were made using Global FNL meteorological archive with a spatial resolution of 191 × 191 km. While the trajectory calculation may include some error (Stohl, 1998), the accuracy of the model to capture the regional features of the process investigated was deemed sufficient.

3. Results and discussion

3.1. *Occurrence of NPF bursts*

An overview of the results of the identification and classification of NPF bursts at the four stations under investigation is shown in Table 2. In the following we will take a closer look at the results of each station.

3.1.1. Hyytiälä. Our analysis here includes one more year than the analysis published by Dal Maso et al. (2005). The results did not change significantly, but they are summarized here for comparison purposes.

Of all data analyzed, 27% are days with new particle formation, while slightly more, 31% were non-event days. The rest, 41% of the days, were classified as undefined. If undefined days are not taken into account, the ratio of events is close to half, 46%. There was a steady increase in the number of event days from 1998 to 2003, but the number dropped in 2004 (see Fig. 1a). The annual variation in event numbers (Fig. 1b) shows a peak in event frequency in spring around April and another peak around September. Summer and especially winter are NPF frequency minima.

3.1.2. Aspvreten. The Aspvreten dataset proved to be difficult to analyze especially in terms of finding non-event days, due to rather strict criteria for such days. Aerosol concentrations in the nucleation mode fluctuated strongly, which resulted in a large number of undefined days: 71% of all analysed days. This causes a large discrepancy in the ratio of events depending on whether these undefined days are included or not. Of all analyzed days, 15% were classified as event days and 13% as non-events. If undefined days were excluded, these numbers changed to 54% and 46% events and non-events, respectively. The variability in the nucleation mode might be caused by anthropogenic activity but also the quite coastal location of the station. If particle formation is assumed to occur only over land, one needs a long period of advection from land to observe a regional particle formation event. If the period is interrupted by advection from sea, the day would be classified as undefined as no continuous nucleation mode could be seen. This problem is not encountered at stations which are uniformly surrounded by forest.

As seen in Fig. 2, the event fraction was largest in 2002 and then decreased. The spring and autumn peaks in NPF occurrence

Table 2. Statistics of the classification of new particle formation bursts at the four stations. ([A]analyzed = sum of event, non-event and undefined days; [C]lassified = sum of event and non-event days)

	Events	Nonevents	Undefined	Discarded	Ev/A, %	Ev/C, %
Hyytiälä	809	933	1224	291	27.3	46.4
Värriö	444	1198	644	271	19.4	27.0
Aspvreten	201	172	950	504	15.2	53.9
Pallas	283	777	455	312	18.7	26.7

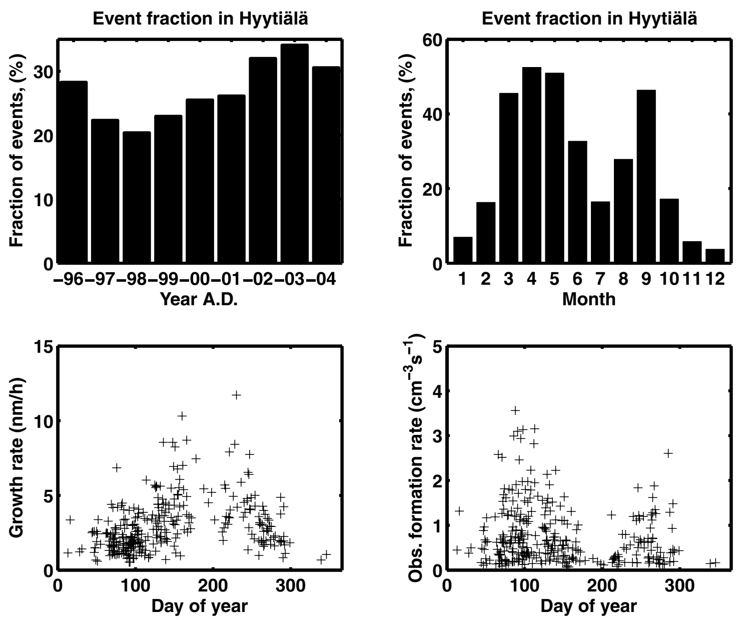

Fig. 1. Results of the analysis of new particle formation bursts at Hyytiälä. (a) fraction of event days of all analyzed days each year (%) (b) the same fraction for each month (%) (c) the growth rate of new particles (nm/h) as a function of the day of year (d) the appearance rate (J_{obs}, see eq. 1).

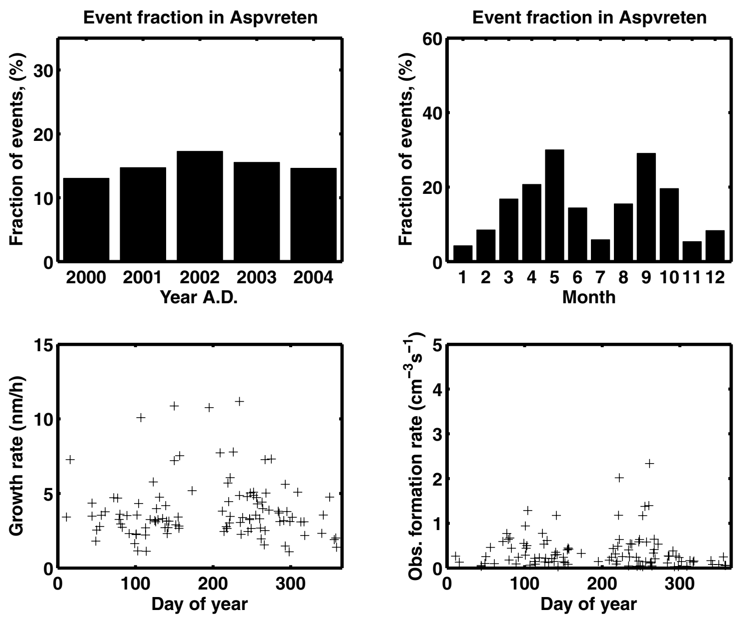

Fig. 2. Results of the analysis of new particle formation bursts at Aspvreten. (a) fraction of event days of all analyzed days each year (%) Note: measurements started in May, so the value for 2000 is not comparable to other stations. (b) the same fraction for each month (%) (c) the growth rate of new particles (nm/h) as a function of the day of year (d) the appearance rate (J_{obs}, see eq. 1).

are clearly seen, and the spring peak is clearer in May. The summer minimum is the most pronounced of all the stations.

3.1.3. Värriö. Our analysis of the Värriö data adds two more years to the analysis performed by Vehkamäki et al. (2004). Additionally, the methodology used in identifying NPF events differs somewhat, making our new dataset more suited for inter-station comparisons.

In Värriö, 19% of all analysed days were NPF days (27% with undefined days ignored) , and 52% (73%) were non-event days. Undefined days amounted to 28% of the data. The annual variation also shows peaks around April and September, but they are less pronounced than in the Hyytiälä case (see Fig. 3). The event numbers were at maximum during 2001 and have been decreasing since.

3.1.4. Pallas. The Pallas analysis yielded very similar results to the Värriö data. Again, 19% (27%, if no undefined) of all days were event days, whereas 51% (73%) were non-event days. 30% of days were undefined. The fraction of events of all days again peaks in the year 2001. The spring and autumn peaks were present also at this station, even though the maximum in September is even less pronounced than in Värriö (see Fig. 4).

3.1.5. Comparison of the stations. If we compare the ratio of events to all analyzed days, the Hyytiälä station stands out with

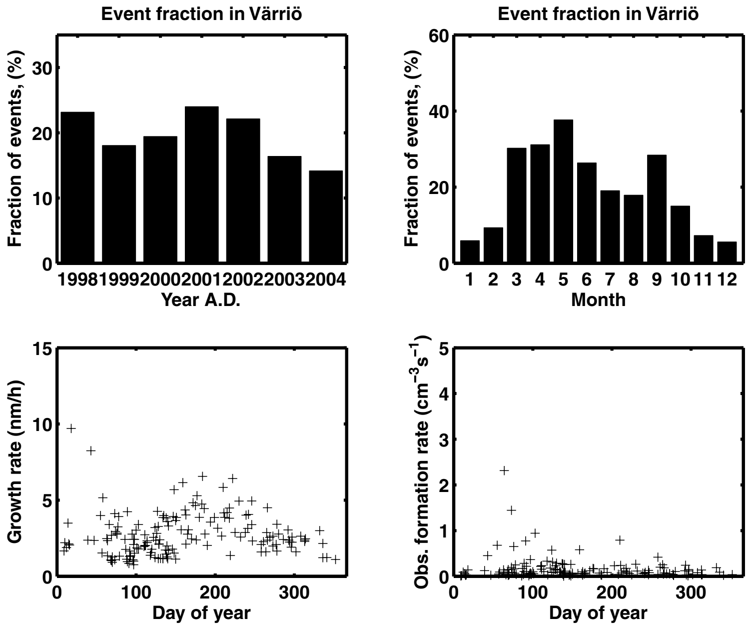

Fig. 3. Results of the analysis of new particle formation bursts at Värriö. (a) fraction of event days of all analyzed days each year (%) (b) the same fraction for each month (%) (c) the growth rate of new particles (nm/h) as a function of the day of year (d) the appearance rate (J_{obs}, see eq. 1).

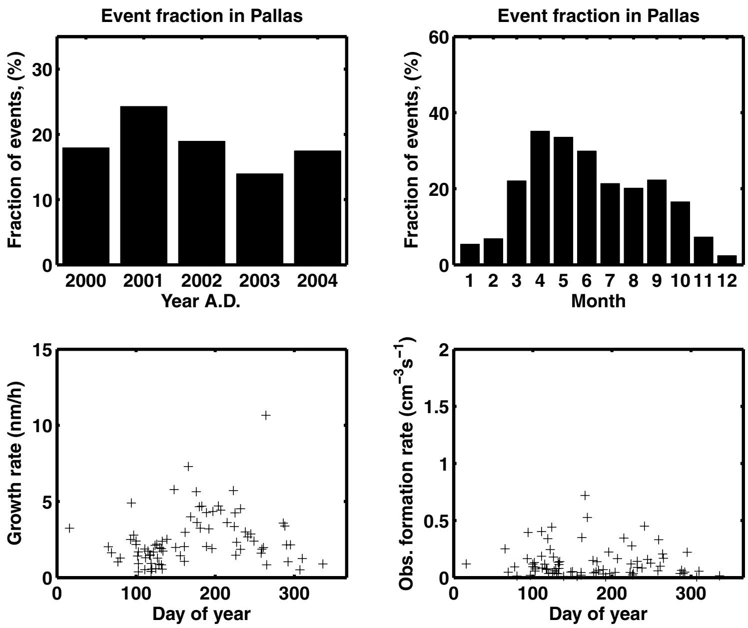

Fig. 4. Results of the analysis of new particle formation bursts at Pallas. (a) fraction of event days of all analyzed days each year (%) Note: measurements started in April 2000, so the value for 2000 is not comparable to other stations. (b) the same fraction for each month (%) (c) the growth rate of new particles (nm/h) as a function of the day of year (d) the appearance rate (J_{obs}, see eq. 1).

clearly more events than the other stations, whereas Aspvreten has distinctly more undefined days than the other stations. The Värriö and Pallas stations are very similar to each other.

If we look at the event fraction of classified days (see Fig. 5 and 6), we see that the Lapland stations, Värriö and Pallas, are still very similar in terms of event occurrence. As we are looking for regional-scale particle formation, and the two stations lie relatively close to each other, this is expected.

On the other hand, the two southern stations, Hyytiälä and Aspvreten, have also very similar event fractions. The yearly numbers follow the same trend and the annual variation is similar at both stations, except that the Aspvreten spring and autumn peaks seem to occur later in the year.

The ratio of events – non-events is clearly higher at the southern stations. Looking at the annual variation (Figure 6) we see that the difference is made during the spring and autumn peaks; during summer the event ratio is similar at all stations, and in winter only Aspvreten stands out with a higher event ratio.

We also looked into NPF days occurring on the same day on two or more stations. The period for this analysis started covering the period of measurements at the Aspvreten station, as measurements there were started last. During these 1673 days,

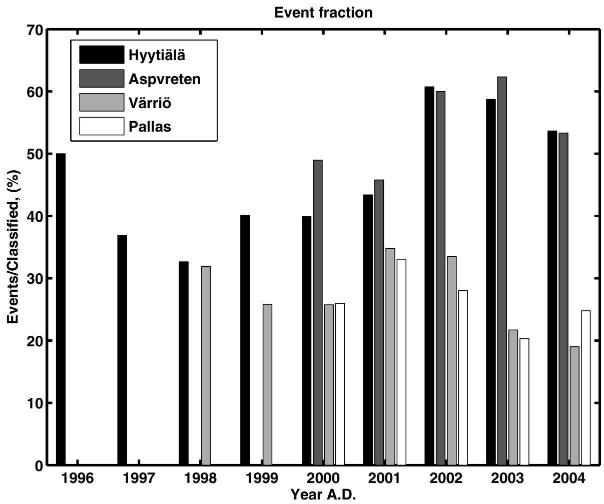

Fig. 5. The ratio of event days to the number of classified days (either event or non-event days), meaning that days of undefined status are ignored. The ratio is shown for all 4 stations as a function of the year the measurement is made. Note that measurements in Pallas and Aspvreten did not start until late spring 2000, so those numbers may not be directly comparable with other sites.

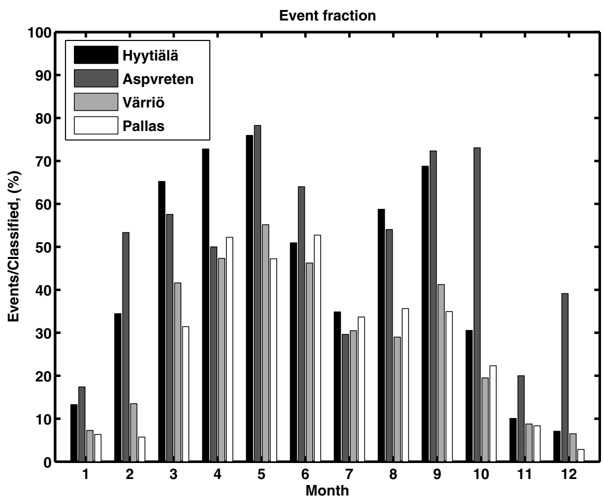

Fig. 6. The ratio of event days to the number of classified days (either event or non-event days), meaning that days of undefined status are ignored. The ratio is shown for all 4 stations as a function of the month the measurement is made. Note that measurements in Pallas and Aspvreten did not start until late spring 2000, so the first three months may not be directly comparable with other sites.

an event was observed on 782 (47%) days at least at one station. On 325 (19%) of the days, an event was seen at two or more stations and there were even 13 days where an event could be seen at all the four stations.

In Table 3 we show a breakdown of the occurrence of simultaneous events. On each row is the fraction of event days that was an event day also at another station. Comparing these numbers

Table 3. Simultaneous occurrence of events at the different stations. Each row gives the percentage of events at each station that are event days also on other stations (given in columns). For example for Hyytiälä, 21% of the 523 event days were also event days in Aspvreten, and so on. The numbers in brackets give the total event day number included in the analysis. The last row gives the fraction of events of all analyzed days at each station.

Station	Hyy	Asp	Var	Pal
Hyytiälä	(523)	21.0%	29.1%	25.2%
Aspvreten	54.7%	(201)	23.4%	21.9%
Värriö	48.3%	14.9%	(315)	38.4%
Pallas	46.6%	15.5%	42.8%	(283)
Overall	27.3%	15.2%	19.4%	18.7%

with the overall occurrence frequency (bottom line), we can get an idea whether there is some connection between the events of two stations. The most notable feature here is that over half of the event days at Aspvreten are also event days at Hyytiälä; this is double the fraction than would be expected from the Hyytiälä event frequency. The coincidence numbers of Värriö and Pallas with Hyytiälä are almost as large. The coincident fractions between the Pallas and Värriö stations are again roughly double the overall fraction of event occurrence, which is expected considering the geographical situation of the stations. The coincidence numbers between Aspvreten and the Lapland stations are low. These coincidence numbers strengthen the hypothesis that particle formation is a regional-scale phenomenon that is triggered under similar, regional-scale meteorological conditions.

Considering these results one should keep in mind that the lowest detection size of the Hyytiälä station is lower than at the other stations. This might lead to differences in the classification of some days, lowering the event day count at the stations with higher detection limits. Measuring at the lowest possible particle size is very important when studying the early stages of the life of atmospheric aerosols.

3.2. *Burst characteristics*

An overview of calculated burst characteristics is given in Table 4. A station-by-station breakdown is given in the following sections.

3.2.1. Hyytiälä. The Hyytiälä event characteristics have again been covered in (Dal Maso et al., 2005). The main results are as follows: the mean growth rate is 3.0 nm/h (median 2.5 nm/h), the values ranging form 0.5 to 15.1 nm/h. The growth rate has annual variation with faster growth seen in summer (see Fig. 1c).

The mean formation rate (J_{obs}) of particles was 0.8 (median 0.6) cm^{-3} s^{-1}, ranging from 0.06 – 5.0 cm^{-3} s^{-1}. The annual variation was similar to the event occurrence frequency, evident in Fig. 1d. For comparison with the other stations, we calculated the coagulation loss rate between 3 and 10 nm for the formation period. The mean loss rate was 0.15 cm^{-3} s^{-1} with a median of

Table 4. Statistics of the characteristics (growth rate *GR*, observed formation rate J_{obs} and condensing vapour source rate *Q*) obtained of the analysis of the new particle bursts for all four stations.

GR (nm/h)	Mean	Median	Std	P_{10}	P_{90}
Hyytiälä	3.0	2.5	1.9	1.1	5.3
Värriö	2.7	2.4	1.4	1.2	4.5
Aspvreten	3.9	3.4	2.0	2.1	6.9
Pallas	2.5	2.0	1.7	0.8	4.7
J_{obs} (cm^{-3} s^{-1})					
Hyytiälä	0.8	0.6	0.7	0.2	1.6
Värriö	0.2	0.1	0.3	0.03	0.3
Aspvreten	0.4	0.2	0.4	0.06	0.8
Pallas	0.1	0.1	0.13	0.02	0.3
Q (10^4 cm^{-3} s^{-1})					
Hyytiälä	9.9	7.1	10	1.9	21
Värriö	4.8	2.9	5.3	0.6	12
Aspvreten	22	13	33	4.4	42
Pallas	3.3	1.5	3.9	0.04	8.8

0.07 cm^{-3} s^{-1}. This was on average 20% of the observed 3 nm formation rate. The equivalent observed formation rate mean with a 10 nm detection limit was thus 0.6 cm^{-3} s^{-1} (median 0.4 cm^{-3} s^{-1}).

The condensable vapour concentration needed to explain the observed growth rate is obtained simply by setting $C_{vap} = A \cdot GR$, with $A = 1.37 \cdot 10^7$ nm $h^{-1}cm^{-3}$ in the free molecular regime (Kulmala, 1988). As it is simply the growth rate multiplied by a constant, no figures for the condensing vapour concentration are shown. The values for the condensing vapour source rate *Q*, obtained assuming a steady state situation (Kulmala et al., 2001), are shown in Fig. 7. The values vary between 0.5–64 · 10^4 cm^{-3} s^{-1}, with a mean (median) of 9.9 (7.1) · 10^4 cm^{-3} s^{-1}. The source is clearly stronger during summer, while the few winter events found have a very low vapour source.

3.2.2. Aspvreten. The growth rates in Aspvreten are higher on average than at the other stations, with the mean/median being 3.9/3.4 nm/h. The observed formation rate was lower than in Hyytiälä but higher than at the Lapland stations, 0.4 cm^{-3} s^{-1} being the mean. The annual variation is very similar to the other stations; this is the case also for the source rate of the condensing vapour, event though the absolute values are clearly higher in Aspvreten than at the other stations.

3.2.3. Värriö. In Värriö the mean (median) growth rate of particles was 2.7 (2.5) nm/h, ranging from 0.8–9.7 nm/h. The annual variation again shows elevated growth in summer (Fig. 3). An interesting feature is that some of the highest growth rates are seen during early spring, around the end of February. It is possible that these days are affected by advection of air from the Kola Peninsula industrial area, which could affect the observed aerosol (Virkkula et al., 1997). The same days are even more clearly seen in the source rate behavior (Fig. 7b); polluted days highlighted). To see whether these days were affected by the anthropogenic sources at Kola we analyzed the trajectories of the days before DOY 100 with source rates above 5 · 10^4 cm^{-3} s^{-1}. We found that with the exception of two days, all trajectories were coming from the east or northeast, over the Kola area. As emissions from the Kola industrial areas include possible aerosol precursors, such as SO_2, we assume that these days do not represent the typical background new particle formation burst.

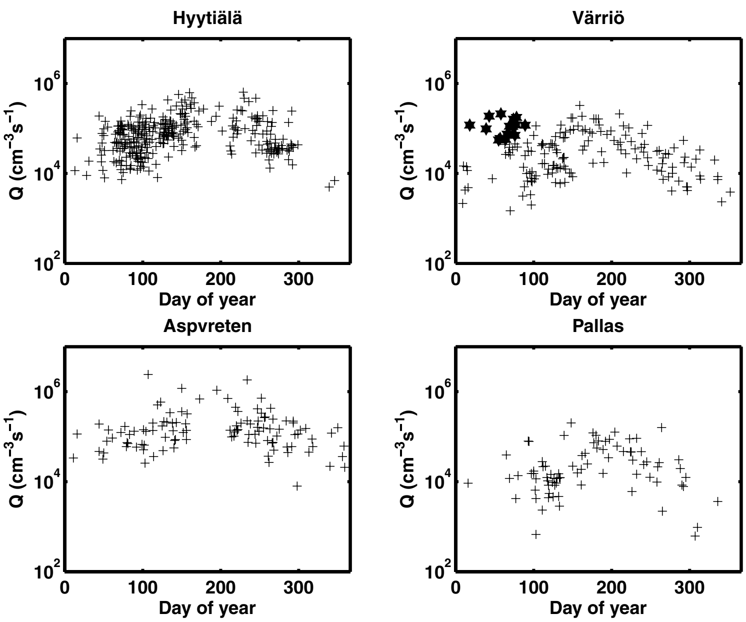

Fig. 7. The estimated source rate of condensible vapour needed to produce the observed growth rate of new particles for all four stations as a function of the day of year. In the Värriö panel the suspected polluted days are highlighted with asterisks.

The observed formation rate was on average 0.2 (median 0.1) $cm^{-3}\ s^{-1}$. Again, elevated formation rates are seen during the period of high event occurrence. The same polluted days that show exceptionally high growth also show higher than average formation rates.

3.2.4. Pallas. The Pallas results show on average the lowest growth rates, with 2.5 (2.0) nm/h being the mean (median) values. The annual variation shows the familiar peak in summer (Fig. 4), whereas the spring high values seen in Värriö are absent here. The formation rates are also low, with a mean (median) of 0.1 (0.1) $cm^{-3}\ s^{-1}$. There are some high values in the summer, but otherwise the same spring/autumn elevation as in the other stations can be seen. The vapour source rate is also low compared to the other stations.

3.2.5. Comparison of the stations. The growth rates obtained in the analysis are all of a very similar order, lying well inside the values found in literature for NPF at various sites around the world (Kulmala et al., 2004a). The annual variation shows consistently high growth rates in summer and lower rates in winter except for Värriö, where anthropogenic pollution probably caused some high growth rates in winter/early spring. This is consistent with the hypothesis that the growth after the initial few nanometers in diameter is mainly affected by (photo-)oxidation products of biogenically emitted organic vapours; the annual variation of both the emitted organics and solar radiation is similar to the one seen in our analysis. The growth rates are higher at the southern stations.

The condensing vapour source rates obtained also support the above-mentioned hypothesis. The vapour source rates at the southern stations are higher by a factor of 5–10; this would lead to higher growth rates (as observed) and also ultimately to more observed formation bursts. The effect of the source is somewhat countered by the low condensation and coagulation sinks at the northern stations, which allows particles with relatively low growth rates to reach detectable sizes.

The observed formation rates also consistently show similar annual variation with peaks in spring and autumn, coinciding with the maxima in event occurrence. The Hyytiälä formation bursts had the highest observed formation rates, even when the lower detection size was taken into account. Generally, formation rates are lower at the Lapland stations than at the southern ones, which may also be a factor leading to the differing event occurrence numbers.

3.3. *Condensation sink*

Hyvönen et al. (2005) showed, using data mining techniques, that the condensation sink caused by the pre-existing aerosol population was found to be a major parameter separating NPF event days from non-event days at the Hyytiälä station. To investigate whether this can be seen generally over the Nordic area, we calculated the daytime (9 am –15 pm) geometric mean of the condensation sink for each analyzed day. (The geometric mean was used because the distribution of the condensation sink values are log-normally distributed.) The data presented here are not corrected for hygroscopic growth, as the parametrization used is made for the Hyytiälä station; this also allows us to include more days as we did not have to care about RH data availability.

The geometric means of the daytime values are given in Table 5, broken down according to the classification of the days. Here we can clearly see a difference between the lower and higher latitude stations: the Lapland stations have on average 2–3 times lower *CS* compared to the southern stations, the averages being $0.98 \cdot 10^{-3}\ s^{-1}$ and $0.72 \cdot 10^{-3}\ s^{-1}$ for Värriö and Pallas, and $2.5 \cdot 10^{-3}\ s^{-1}$ and $3.3 \cdot 10^{-3}\ s^{-1}$ for Hyytiälä and Aspvreten, respectively. On event days, the condensation sink was significantly below average at all the stations; the average value of the sink was 70–80% of the overall average except at Aspvreten, where the event day average was *ca.* 50% of the overall average sink value. In contrast, non-event days had above average sinks at all stations except Pallas, where the nonevent days sink average was practically equal to the overall average. Undefined days were usually close to the overall average, the exception again

Table 5. The daytime condensation sink geometric means $\overline{CS}_g$ of event, non–event and undefined days analyzed at the four stations along with the number of days in each category. The differing numbers to Table 2 are caused by absent data during the averaging period.

Hyytiälä	N	$\overline{CS}_g$ $10^{-3}\ s^{-1}$
Events	791	1.7
Nonevents	912	2.9
Undefined	1210	2.5
Total	2913	2.4
Aspvreten		
Events	201	1.7
Nonevents	172	4.5
Undefined	948	3.3
Total	1321	3.1
Värriö		
Events	439	0.70
Nonevents	1160	1.2
Undefined	631	0.98
Total	2230	1.0
Pallas		
Events	279	0.48
Nonevents	772	0.59
Undefined	430	0.72
Total	1481	0.6

being Pallas where undefined days had an average *CS ca.* 30% higher than overall.

Considering these results, it seems that the condensation sink has a stronger effect on nucleation at the southern stations than at the northern ones. One possible reasons for this could be that at the southern stations the condensation sink, being a NPF inhibitor, prevents new particle formation and growth even though the precursors might be available. In the north, however, the sinks are generally so low due to low anthropogenic influence that their inhibiting power is diminished. This could indicate that particle formation at the northern stations is more source-regulated than at the southern stations.

3.4. *Trajectories*

To investigate the area from which the air leading to new particle formation originated the calculated back trajectories were analyzed with respect to NPF days. As analyzing each trajectory individually was impractical for the large number of days analyzed, we used the azimuth of the air parcel t_n hours before arrival at the measuring site as a parameter describing the air mass source direction. We used values $t_n = 6, 12, 24, 48, 72$ and 96 hr for our analysis, but the following results are computed using $t_n = 24$ h. The following results do not change if t_n is taken to be 12 or 6 hr; larger t_n:s lead to an averaging as the air mass history gets longer.

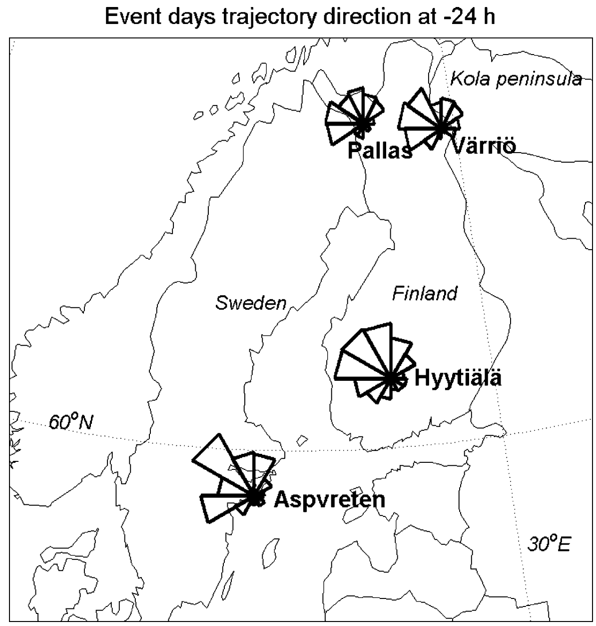

Fig. 8. The azimuth of the air masses 24 h prior to arriving at the measurement stations on event days.

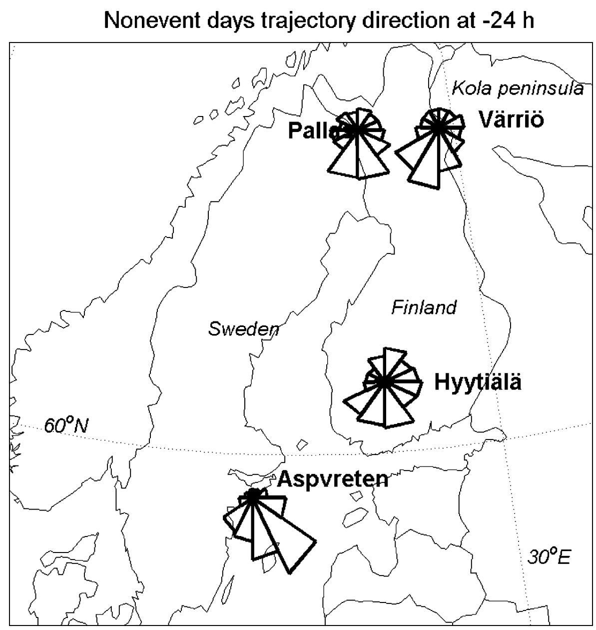

Fig. 9. The azimuth of the air masses 24 h prior to arriving at the measurement stations on non–event days.

Figure 8 shows the relative distribution of air mass azimuths on event days for all the four stations. From these it is clearly visible that air masses on event days are predominantly arriving from the northwest direction. If we draw an axis going from northwest–southwest, almost all event day trajectories arrived from the NW side of this axis.

In contrast, the relative distributions of the trajectories arriving on event days (see Fig. 9) show more variation. At the Finnish stations, nonevent air masses arrived predominantly from southern directions while for the Aspvreten station non–event days were observed when air masses were advected from SE.

Undefined days showed the most variation between the stations. In Pallas and Värriö the distribution of the transport direction of undefined days was quite flat, with no direction standing out. In contrast, the distribution at Hyytiälä is dominated by trajectories arriving from directions between west and south. The undefined events at Aspvreten are of special interest, considering their large number. We found that the direction between azimuths 210 and 270 (SW-S) was the main direction for undefined days to come from: 32% of all undefined days has air massed advecting from that direction. On the other hand, though, it has been shown previously by Tunved et al. (2003) that this direction is the prominent direction of any air mass arrival at Aspvreten; ca 30% of all trajectories come from this 60° range. This can be looked at also in another way: of all trajectories arriving from this direction, 81% percent are lead to undefined days. This, while being a larger fraction, is not significantly more than the 68% of undefined days for the [270 . . . 360] degrees range. Thus, we cannot attribute the large amount of undefined days to pollution transport from southern regions alone.

A coarse summary of the trajectory direction analysis is given in Table 6. Here we took into consideration the azimuth range that lead to NPF events (270–360 degrees, west - north) and the range leading to non–events (120–240 degrees, roughly southeast-southwest). The event, non–event and undefined day statistics for

Table 6. Statistics for different directions of air masses arriving at the stations. The azimuths 120..240 were found to be dominating as a source for non-event air masses, whereas the azimuth range 270..360 (W-N) lead often to new particle formation. $\overline{CS}_g$ is the geometric mean of the daytime condensation sink.

Sector	N total	Events	Non-event	Undef	$\overline{CS}_g$ 10^{-3} s^{-1}
Hyytiälä					
120..240	1081	159	415	507	3.9
270..360	797	353	148	296	1.4
Aspvreten					
120..240	506	30	117	359	4.5
270..360	311	91	9	211	1.8
Pallas					
120..240	561	41	381	139	0.98
270..360	369	117	134	118	0.35
Värriö					
120..240	876	60	616	200	1.6
270..360	524	177	186	161	0.45

trajectories arriving from these azimuth ranges are given along with the geometric mean of the daytime condensation sink of these days. The feature standing out of these statistics is the clear difference of the condensation sink averages between the north-west sector and the southeast-southwest sectors: the *CS* in air arriving from Northwest is lower by factor of 2.5–3 at all stations. The difference is markedly higher that in the comparison between only event and non-event days.

The results of the trajectory analysis are in agreement with results published by Sogacheva et al. (2005) for the Hyytiälä station, which established the Northern Atlantic area as the source of air masses leading to new particle formation. Coupled with the annual variation of formation occurrence, the results of Nilsson et al. (2001), which state that NPF occurs preferably in Arctic air masses, are also supported.

4. Conclusion

We have analyzed aerosol size distribution time series of four Nordic background stations with respect to identifying new particle formation events and deriving their characteristics. The total amount of data analyzed covers 8090 one-day periods; of these 1737 (21%) were new particle formation event days. The fraction of event days of the classified (undefined days are ignored) days is 36%. Considering these numbers, we are confident in saying that new particle formation in the lower troposphere over the boreal forest is a frequent phenomenon.

We also found that annual variation of NPF events, with maxima in spring and autumn, is a feature of all stations; however, it is clearly more distinctive at stations situated at lower latitudes. Event occurrence is less frequent at higher latitudes; the difference can be attributed mostly to less formation activity during the peak time periods.

The growth and formation rates obtained agree well with other observations found in the literature (see for example Kulmala et al. (2004a), Stolzenburg et al. (2005)). The annual variations of these parameters were as expected, the growth being faster with more intense solar radiation and the formation rate being higher with higher burst frequency. There were some anomalous bursts found, but they were explained by pollution from industrial areas. Lower-latitude burst were more intense measured by both the growth and formation rates of the newborn nuclei. We found that to explain the observed growth rates, southern stations should have a vapour source which is stronger by a factor of at least 5 compared to the northern stations. Northern NPF is characterized by a weaker source of particles and condensing vapour, but this is compensated by less pre-existing condensation and coagulation sink, enabling the new particles to survive for longer periods. The sink has a greater effect as a NPF inhibitor at the southern stations, whereas in the north the precursor source might be a more important limiting factor.

The source of the air masses leading to new particle formation was established to be towards the North Atlantic area for all the stations. Southern air transport rarely leads to new particle formation at any of the stations.

At all stations the data included bursts after which the particles grew to sizes where they could act as CCN. The similarity of our other results for all stations strengthen the argument that the same process or processes are producing particles over the whole area studied.

Summarizing, using long-term measurements over a geographically large area we were able to establish that new atmospheric particles produced by nucleation are being produced over the whole Nordic boreal forest area. The formation is characterized by low pre-existing condensation sink and advection of cooler air from the North Atlantic.

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