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Time span and spatial scale of regional new particle formation events over Finland and Southern Sweden

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Abstract. We investigated the time span and spatial scale of regional new particle formation (NPF) events in Finland and Southern Sweden using measured particle number size distributions at five background stations. We define the time span of a NPF event as the time period from the first moment when the newly formed mode of aerosol particles is observable below 25 nm until the newly formed mode is not any more distinguishable from other background modes of aerosol particles after growing to bigger sizes. We identify the spatial scale of regional NPF events based on two independent approaches. The first approach is based on the observation within a network of stationary measurement stations and the second approach is based on the time span and the history of air masses back-trajectories. According to the second approach, about 60% and 28% of the events can be traced to distances longer than 220 km upwind from where the events were observed in Southern Finland (Hyytiälä) and Northern Finland (Värriö), respectively. The analysis also showed that the observed regional NPF events started over the continents but not over the Atlantic Ocean. The first approach showed that although large spatial scale NPF events are frequently observed at several locations simultaneously, they are rarely identical (similar characteristics and temporal variations) due to differences in the initial meteorological and geographical conditions between the stations. The growth of the newly formed particles during large spatial scale events can be followed for more than 30 h where the newly formed aerosol particles end up in the Aitken mode (diameter 25–100 nm) and accumulation mode size ranges (diameter 0.1–1 μm). This study showed clear evidence that regional NPF events can pose a significant source for accumulation mode particles over the Scandinavian continent provided that these findings can be generalized to many of the air masses traveling over the European continent.

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1 Introduction

Contrary to greenhouse gases that warm up the atmosphere, aerosol particles are capable of cooling down the climate. Aerosol particles have direct and indirect effects on the Earths' radiation balance. The direct effect includes scattering of incoming solar radiation that directly affects the radiative balance of the Earth (e.g. Haywood and Boucher, 2000). The indirect effect is a result of the interplay between aerosols and water vapor where aerosol particles play a role as cloud condensation nuclei (CCN) for the cloud formation that further affects the radiative balance of the Earth (e.g. Lohmann and Feichter, 2005). New particle formation (NPF) is possibly one of the key processes in producing high aerosol number concentration over large scales in both remote and urban areas as illustrated by several studies performed worldwide (e.g. Kulmala et al., 2004 and references therein).

NPF events can be identified when a distinct new mode of aerosol particles is observable in the nucleation mode size-range (mainly diameter $<$25 nm) for several hours (e.g. Dal Maso et al., 2005). In general, the newly formed particles

may find suitable ambient conditions to grow to larger sizes where they can act as cloud condensation nuclei (CCN). It has been noticed already earlier that the observed growth of aerosol particles following a continental NPF event suggests that the event has a spatial scale over large areas (e.g. Tunved et al., 2003; Mäkelä et al., 1997). Similar reasoning was used by Charron et al. (2007) to conclude that observed particle formation in southern England occurred over an area reaching 20–200 km from the measurement site. Tunved et al. (2003) investigated NPF events at four Nordic background stations (Värriö, Hyytiälä, Aspvreten, and Vavihill) during June 2000–May 2001; they revealed that NPF events may take place on a regional scale over Finland and Southern Sweden. Recently Dal Maso et al. (2007) also confirmed the occurrence of regional NPF events at four Nordic background stations (Värriö, Pallas, Hyytiälä, and Aspvreten) when they investigated the NPF events during a longer time period (2000–2004). Tunved et al. (2006a) concluded in a pseudo-Lagrangian study that regional-scale nucleation is a probable cause for the increased number of particles as one travels from north to south. One should notice that the continental particle formation studied here differs from the coastal NPF observed, e.g. in western Ireland, where the formation itself occurs on the coastline and the growth of the formed particles continues on a larger spatial and time scale (O'Dowd et al., 2002).

A typical continental regional NPF event consists of two stages: during the first stage the aerosol particles are formed and observed within the nucleation mode size-range and during the second stage the newly formed particles grow to bigger sizes. This introduces a new concept, "time span", that can be defined as the time period from the first moment when the newly formed mode of aerosol particles is observable below 25 nm until the newly formed mode is not any more distinguishable from other background modes of aerosol particles after growing to bigger sizes. It is, any way, expected that the longer the time span is, the larger the spatial scale of the regional NPF event. However, it has not been clear how the spatial scale is linked to the time span.

Aiming at linking the relationship between the time span and the spatial scale requires high quality aerosol measurements of particle number size distributions within a network of stationary measurement sites during a long-term period. Even though NPF events have been observed at many places regardless to the area type, location, or altitude e.g. Kulmala et al. (2004 and references therein), most of these observations have been limited to short time periods, single stationary location, or platforms not necessarily aligned on the same air mass trajectory. While trajectory studies of air mass origins leading to NPF have been performed earlier for both Boreal and Southern European NPF (e.g. Sogacheva et al., 2005 and 2007), such studies did not give information on the spatial scale of the formation itself as no information of the time span of events was used.

In this study we investigated the time span and the spatial scale of the observed NPF events at five monitoring stations in Finland (SMEAR I–III stations) and southern Sweden (Vavihill and Aspvreten). We did not aim at presenting a new classification for the observed NFP events; instead, we used previous classifications as reported by Hussein et al. (2008), Kristensson et al. (2008), and Dal Maso et al. (2005 and 2007) in order to select the dates of observed NPF events at these measurement stations. Contrary to previous studies, which were focused on the seasonal variation of the NPF events, in this study we considered event-to-event comparison at these stations. An attempt of this kind has rarely been tried before and it provided quantification for the occurrence of regional identical-NPF events. We also quantified the time span of the observed regional NPF events and how it is related to the spatial scale of the event. Thorough statistical data was provided for this analysis, which is not available for any other NPF event study. At the end of this study we present examples and detailed analysis for a regional identical-NPF event and a regional event with a long time span.

2 Materials and methods

In this study we utilized the continuous measurements of aerosol particle number size distributions at five background stations (Fig. 1): the SMEAR I–III stations (Station for Measuring forest Ecosystem-Atmosphere Relations), Vavihill, and Aspvreten. These stations have been in the focus of many studies, especially those related to NPF (e.g. Hussein et al., 2008; Kristensson et al., 2008; Dal Maso et al., 2005 and 2007; Vehkamäki et al., 2004; Tunved et al., 2003).

The selected stations are aligned along the north-south starting at Vavihill in Southern Sweden and ending at Värriö (SMEAR I) in the Finnish Lapland. This alignment along the north-south line is suitable for both long-range transport (LRT) analysis and regional NPF events. Vavihill and Aspvreten are the first to be influenced with LRT episodes from Central Europe (e.g. Tunved et al., 2005). NPF events at these stations are often observed during North-Western air masses originating from the Atlantic Ocean (e.g. Hussein et al., 2008; Dal Maso et al., 2007 and 2005).

Handling long-term data sets is a challenging task especially when it involves more than one site. Because the measurement period varied from site to site, we considered common time periods according to the availability of aerosol data, classification of NPF events, and spatial scale considered in this study; as will be shown later in the following sections (Table 1). We did not include the aerosol data after 31 December 2006 in our analysis mainly because some of the data has not been finally processed and quality checked after that date at some sites.

Table 1. A summary about the time periods of the available aerosol data and NPF classification.

Monitoring station	Start date of the measurement	NPF classification	Reference
Värriö (SMEAR I) [1,2]	1 January 1998	1 January 1998–31 December 2006	Dal Maso et al. (2007) and UHEL
Hyytiälä (SMEAR II)[3]	30 January 1996	20 January 1996–31 December 2007	Dal Maso et al. (2005) and Riipinen (2008)
Helsinki (SMEAR III)	5 May 1997	5 May 1996–31 December 2006	Hussein et al. (2008)
Aspvreten	31 May 2000	31 May 2000–5 September 2004	Dal Maso et al. (2007)
Vavihill	10 February 2001	21 February 2001–20 May 2004	Kristensson et al. (2008)

[1] The classification of NPF events at SMEAR I was available by Dal Maso et al. (2007) until the end of 2004. The rest of the period was available from unpublished analysis at the University of Helsinki.
[2] Dal Maso et al. (2007) reported 535 NPF events (226 Class I) at Värriö during 1998–2006.
[3] Riipinen (2008) reported 850 NPF events (425 Class I) in Hyytiälä during 5 May 1997–31 December 2006.

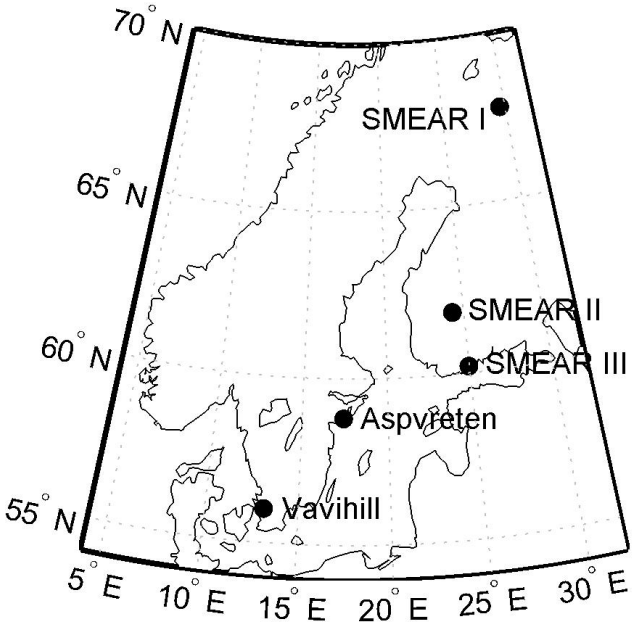

Fig. 1. A map showing the measurement stations considered in this study.

2.1 Description of stationary measurement stations

The *SMEAR I* station is situated at Värriö (67°46′ N, 29°35′ E) in Eastern Lapland close to the Russian border (e.g. Hari et al., 1994). Värriö is a background remote continental site far from pollution sources, although emissions from the Kola Peninsula give rather strong influence when winds transport air from this region. Also, winds coming from the St. Petersburg area as well as Russia in general may bring elevated concentrations of acidifying gases and particulate pollution. The particle number size distributions have been measured at the SMEAR I station since year 1998 and the station has been operated by the University of Helsinki.

The *SMEAR II* station is situated at Hyytiälä (61°51′ N, 24°17′ E) in Southern Finland (e.g. Hari and Kulmala 2005). Hyytiälä is a remote boreal forest site dominated by Scots Pine trees. The station is located fairly far from urban pollution sites; the nearest urban sites are Tampere (∼50 km to the southwest) and Jyväskylä (∼100 km to the northeast). The particle number size distributions have been measured at the SMEAR II station since January 30, 1996 and the station has been operated by the University of Helsinki.

The *SMEAR III* station officially started in year 2004 on the Kumpula campus (60°12′ N, 24°57′ E) of the University of Helsinki located in Helsinki. Before that, the measurements of the particle number size distributions started on May 5, 1997 at the old location of the Department of Physics on the Siltavuorenpenger campus. In March 2001, the Department of Physics was re-located to its current location on the Kumpula campus (∼5 km north of the old location). The first re-location from Siltavuorinpenger to Kumpula did not show significant differences on the physical characteristics of the measured particle number size distribution. During the whole measurement period since 5 May 1997 the aerosol particles have shown clear urban background characteristics (Hussein et al., 2004). The SMEAR III station has been operated by the University of Helsinki.

The background monitoring station *Aspvreten* (58°48′ N, 17°22′ E) is located in Sörmland about 70 km south west of Stockholm. The station is situated about 2 km from the coast in a rural area covered by mixed coniferous and deciduous forest with some meadows. The area around the station is sparsely populated and the influence of anthropogenic activities is considered minor (e.g. Tunved et al., 2003). The particle number size distributions have been measured since June 2000 and the station has been operated by the Institute for Applied Environmental Research. This station is a part of the European Monitoring and Evaluation Programme network (EMEP).

Vavihill (56°01′ N, 13°09′ E) is a background site at the top of Söderåsen in Skåne. The surroundings are dominated by grasslands and deciduous forest (e.g. Kristensson et al., 2008). The monitoring station itself is situated about 10 km away from the closest small villages and about 20 km from the city Helsingborg. The area of Malmö and Copenhagen,

with about 2 million inhabitants, is situated about 60–70 km to the SSW. The measurement site is not influenced by local anthropogenic activities. The particle number size distributions have been measured at Vavihill since February 2001 and the station has been operated by Lund University. This station is a part of the European Monitoring and Evaluation Programme network (EMEP).

2.2 Aerosol particle measurements and data extraction

The particle number size distributions have been measured with Differential Mobility Particle Sizer (DMPS) at the five monitoring stations. The differential mobility analysis (e.g. Aalto et al., 2001; Winklmayr et al., 1991) of aerosol particles relies on bipolar charging of aerosol particles followed by classification of the particles based on their electrical mobility by using a differential mobility analyzer (DMA) and then counting the particles with a condensation particle counter (CPC). In general, the measured diameter of aerosol particles with a DMPS is limited to a size-range from several nanometers up to several hundreds of nanometers (dry diameters). A system with two DMPS units (a twin-DMPS system) can measure the particle number size distribution within a wide size-range from 3 nm up to 1000 nm.

The particle number size distributions are extracted from a DMPS by data inversion. The transfer function of the DMA is implemented in the inversion procedure according to Stolzenburg (1988) and the particle charging probability according to the semi-empirical functions by Wiedensohler (1989). The detection efficiency and the losses in the transport lines and inside the DMA can be estimated from the laminar flow tube diffusion loss equations. The time resolution of the measured particle number size distribution varies from 5 to 10 min and the number of measured size bins varies from 24–48.

2.3 Aerosol data handling and validation

The aerosol data was first quality checked and prepared for this analysis individually by the operator of the measurement at each monitoring station. The DMPS setup was slightly different at different sites; however, laboratory calibration and data quality assurance provide reliable data for further processing and analysis. The measured size-range of the particle number size distribution varied from site to site and also from time to time on the same site due to improvements in the measurement setup. The lower limit of the measured size-range varied between 3–10 nm whereas the upper limit varied between 320–1000 nm. The upper limit of the measured size-range should not have a significant influence on the comparison among the sites because particles larger than 320 nm have a minor fraction of the total particle number concentration. Even though the lower limit is the critical matter here we can validate our comparison among different sites by fitting the measured particle number size distributions with the multi-lognormal distribution function. The multi-lognormal distribution function has been used to parameterize the particle number size distribution (e.g. Whitby 1978). Each lognormal mode is defined by a geometric mean diameter (D_{pg}), a geometric standard deviation (σ_g), and a number concentration (N).

In this study, we used our automatic algorithm (DO-FIT, Hussein et al., 2005) to find the suitable number of modes and their parameters ($D_{pg,i}$, $\sigma_{g,i}$, $N_{,i}$) needed to best-fit the measured particle number size distribution. We used the lognormal parameters to describe the dynamic behavior of aerosol particles during and after NPF events. With the help of such parameterization we can also identify and distinguish a growing mode of aerosol particles from other pre-existing aerosol particles within the same size-range.

2.4 Air mass back-trajectory

Trajectory analysis of air masses is a useful tool to investigate the origin of air masses arriving at the location of interest. We used the HYSPLIT_4 model (Draxler and Hess 1998) that was developed by NOAA/ARL. HYSPLIT_4 is a single particle lagrangian trajectory dispersion model. We made the model runs by using the Global FNL meteorological archive with 190×190 km^2 spatial resolution and 96 h back in time at 100 m arrival height. Current literature suggests that the error in a trajectory is within 15–30% of the travel distance (Stohl 1998, Draxler and Hess 2004). In this paper we considered the sources of aerosol particles in a local (arrival point) and regional scale (2–4 days back).

2.5 Analysis of new particle formation events: time span and spatial scale

In this study we did not aim to re-classify NPF events again; instead, we used the classification results for the five sites from our previous studies (Table 1); the classification scheme is summarized in Appendix A.

Our analysis included the "time span" and the "spatial scale" of the reported NPF events. As defined in the introduction section, we quantified the time span of an event by visually inspecting the temporal variation of the particle number size distribution from the start time of the event (when the newly formed mode is observable below 25 nm) until the newly formed mode is not any more distinguishable in the atmosphere after growing to bigger sizes. We quantified the spatial scale according to two independent approaches as will be summarized in the following subsections.

2.6 Approach I: Spatial scale based on a network of stationary measurement stations

In this approach we considered an NPF event occurred over a small spatial scale (more or less local event) if the event was observed at one site only. In a similar manner, an NPF event has a large spatial scale if the event was observed at

more than one site starting by considering Hyytiälä (SMEAR II) as the central site in this study. Here the term "small spatial scale" refers to a limited area that does not exceed $200 \times 200\,km^2$. On the other hand, the term "regional" refers to a spatial scale larger than several hundreds of kilometers in each horizontal direction. This approach is straight forward and it requires a network of stationary stations to identify the spatial scale of NPF events. We should keep in mind that this approach has limitations. For example, it is still possible that a small spatial scale event can be in fact regional (or occurring on a larger spatial scale) but due to the limitations on the number of measurement sites within our domain it was identified as a small spatial scale event. This is especially true for the sites such as Värriö and Vavihill that are located on the edge of the domain. We, therefore, expect this procedure to be more accurate for the most central sites such as Hyytiälä, Aspvreten, and Helsinki according to the domain in this study.

According to this approach we defined three spatial scales for the observed regional NPF events:

1. "Southern Finland" that includes Hyytiälä and Helsinki only.
2. "Finland" that includes the SMEAR stations: Hyytiälä, Helsinki, and Värriö.
3. "Finland and Southern Sweden" that includes the SMEAR stations, Aspvreten, and Vavihill.

We classified the regional NPF events observed on the same day at several sites as "simultaneous NPF events". Because we were interested in the so called "regional identical-NPF events" which we identified according to the event-to-event comparison among all sites within each spatial scale. "Identical" here refers to the similarity in the start time and temporal behavior of the event at all sites regardless to the absolute value of the number concentration.

According to this approach, identical-NPF events are a subset of the simultaneous events. Furthermore, the identified events over Finland and Southern Sweden are a subset of the events over Finland, which are in turn a subset of the events identified over Southern Finland.

2.7 Approach II: Spatial scale based on the time span and back-trajectories

Since the data acquisition is Eularian and NPF events take place within individual air masses, which is Lagrangian, certain conditions must apply in the analysis of regional NPF events. From the fact that a regional NPF can be followed by a growth pattern as observed at a single location, it follows that such events must have taken place over large areas. As a result, the growth and formation rates are expected to be similar on that spatial scale. This is especially true for well defined events such as class I events. In that since, the time span of an event holds information about its spatial scale.

Let us consider a regional NPF event that has a time span of 24 h where the mean wind speed is 5 m/s at the station. Assuming that the growth rate of the particles is similar on the spatial scale of interest (i.e. homogeneous characteristics of aerosols), it follows that the extent of the event is at least 400 km upwind. This, in principle, leads to the conclusion that if we know the time span and the wind speed we can determine the minimum spatial scale of the event in at least one dimension. Since it is possible to follow regional NPF events during the course of hours up to days, it is clear that the events start over large spatial scales simultaneously. This means that following the back-trajectories for every time-step during the time span of the event we can trace back the minimum distance, and thus the minimum spatial scale upwind, from the observation location where the air is homogeneous with respect to the characteristics of aerosols.

We have to be aware that the only possibility why this approach would work is that meteorology and source distribution is similar (i.e. homogeneous) over the spatial scale on which the event takes place. The fact that NPF events should in general be a regional phenomenon and that the sources over the boreal forest are homogenous was previously demonstrated by Tunved et al. (2006b) by correlating the time spent over the forest with the evolution of the size distribution.

In details, this approach consists of the following steps:

1. We assumed that all events started at 10:00, which is the typical start time of NPF events observed at Hyytiälä and Värriö.
2. We divided the time span of the event into 1-h time-steps, and then we calculated back-trajectories for each time-step throughout the time span of the event.
3. The length of the individual trajectories (in hours) is determined by the number of time steps taken. For example, assuming an event started at 10:00 everywhere, we calculated one trajectory for 10:00 with 1-h duration (i.e. 10:00 + 1-h). This was followed by trajectories calculated for the next time-step at 11:00 (+ 2-h long trajectories), 12:00 (+ 3-h) and so on.
4. For every trajectory, which corresponds to a single time-step in the time span of the event, the last endpoint was registered as the minimum distance of the event from the observation point. This yielded the minimum spatial scale associated with the event in the air masses that the trajectories represent.
5. In order to better describe the spatial scale of NPF events according to this approach, a polar grid was set up for the receptor station. In the evaluation, the North Pole was transferred to the receptor, and the polar grid surrounding was made up of 29×29 grids (between

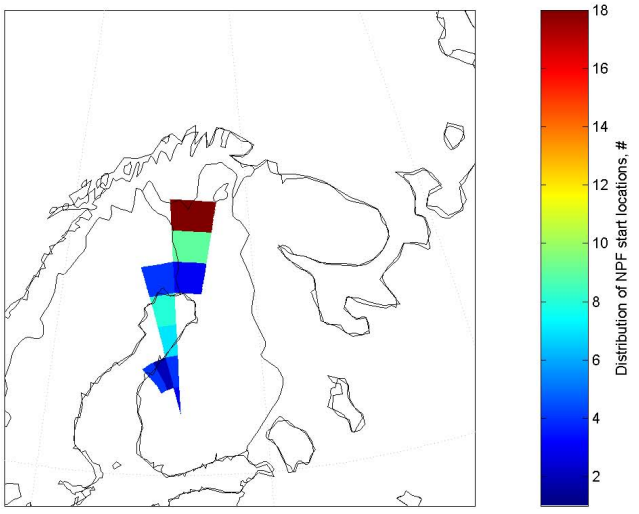

Fig. 2. The minimum spatial scale of the regional NPF event observed on 19 October 2001 at Hyytiälä. The time span of this event was 65 hours and the figure is created from the endpoints of 65 trajectories calculated through the duration of the event.

90° N–60° N and 180° E–180° W) each one with a spatial extent of 1°×12° (lat, long) grids. This type of analysis provided the origin of aerosol particles observed at Hyytiälä, for instance, during a NPF event.

As an example, consider the regional event that occurred on 19 October 2001 in Hyytiälä: according to this analysis, the time span was 65 h. The length of the back-trajectories considered for this event varied from 1-h (at the start time of the event) to 65-h (at the end time of the event). The minimum spatial scale of this event is then presented in Fig. 2 which illustrates the distribution of locations (spatial scale) at which the NPF takes place simultaneously where aerosol particles are expected to have homogeneous regional characteristics. These locations do only represent the upwind spatial scale of the regional NPF events, and from this it follows that the true extent of the event is likely larger, i.e. downwind of the receptor station.

3 Results and discussion

3.1 Time span of NPF events

The Swedish sites were excluded from the time span analysis because they often experienced strong long-range transport events that prevented us from accurately observing the end time of NPF events. Helsinki was also excluded from this analysis for the same reason and additionally because of the high concentrations of ultrafine particles in the urban background. The majority of NPF events observed at Hyytiälä (∼62%) and Värriö (∼72%) had a time span between 6 and 24 h (Fig. 3). The fraction of events with a time span between 6 and 24 h observed at Värriö was bigger than that for Hyytiälä. This was because Värriö is a cleaner background station compared to Hyytiälä, and thus, it was more accurate to observe the end time of the event when the newly formed aerosol particles had grown to the accumulation mode size range.

A reasonable fraction of NPF events observed at Hyytiälä (∼34%) and Värriö (∼22%) had a time span longer than 24 h. Furthermore, about 3% and 1.1% of the events observed at Hyytiälä and Värriö, respectively, had a time span longer than 3 days. This is indeed a small fraction, but this accounts for 25 and 6 events observed at Hyytiälä and Värriö respectively. In this study we will present detailed analysis for a special cases when the time span of NPF events exceeded five days.

The median value of the time span was 23 and 16 h respectively for Class I and Class II NPF events observed at Hyytiälä (Fig. 4). The time span of Class Ia events (median value 27 h) was found to be longer than that of Class Ib events (median value 21 h) at Hyytiälä. Similarly, the event time span was also the longest for Class Ia events at Värriö (Fig. 4). This finding is expected because Class Ia events were classified as such because are the best quality events with clear growth pattern and high formation rate; they are also suitable for model studies. Being clear event with the best quality among other classes implies that the event remained for a long time period.

It is important to mention here that NPF events often occurred on a daily sequence for several days; each event started between 10:00 and 12:00 and lasted for about one day. The observation of sequence events can be explained as follows: (1) these events were observed in arctic and polar air masses during cold air outbreaks (e.g. Nilsson et al., 2001a). (2) The observation of NPF was also associated with the onset of strong vertical mixing that often results in significant decrease in particle number concentrations prior observation of recently formed particles (Nilsson et al., 2001b). The disappearance of the aerosol particles originated from NPF events can also happen via one or more of the following: coagulation with pre-existing particles, dilution and convection with incoming air, and a sudden change in the air masses. The modal structure of the particle number size distribution changes its characteristics immediately when the air mass changes suddenly.

3.2 Spatial scale of regional NPF events

3.2.1 Observations within a network of stationary measurement stations

Even though we observed 137 simultaneous NPF events at Helsinki and Hyytiälä during 1998–2006, only 60 of these events were regional identical-NPF events in Southern Finland. Extending the spatial scale to cover Finland by

Table 2. Occurrence of simultaneous and identical regional NPF events. Simultaneous NPF events are those observed on the same day whereas identical-NPF events are those simultaneous events started at the same time at all sites and showed similar characteristics.

Time period	Regional scale	Simultaneous events	Identical events
5 May 1997–31 December 2006	Southern Finland(1)	142	61
1 January 1998–31 December 2006	Southern Finland	137	59
	Finland(2)	50	9
21 February 2001–20 May 2004	Southern Finland	31	20
	Finland	22	3
	Finland and Southern Sweden(3)	9	2

(1) The regional scale "Southern Finland" includes Helsinki (SMEAR III) and Hyytiälä (SMEAR II). The total number of NPF events was respectively 184 and 850 observed at Helsinki and Hyytiälä.
(2) The regional scale "Finland" includes Helsinki (SMEAR III), Hyytiälä (SMEAR II), and Värriö (SMEAR I). The total number of NPF events was respectively 175, 823, and 535 observed at Helsinki, Hyytiälä, and Värriö.
(3) The regional scale "Finland and Southern Sweden" includes the SMEAR stations, Aspvreten, and Vavihill. The total number of NPF events was respectively 56, 380, 220, 164, and 245 observed at Helsinki, Hyytiälä, Värriö, Aspvreten, and Vavihill.

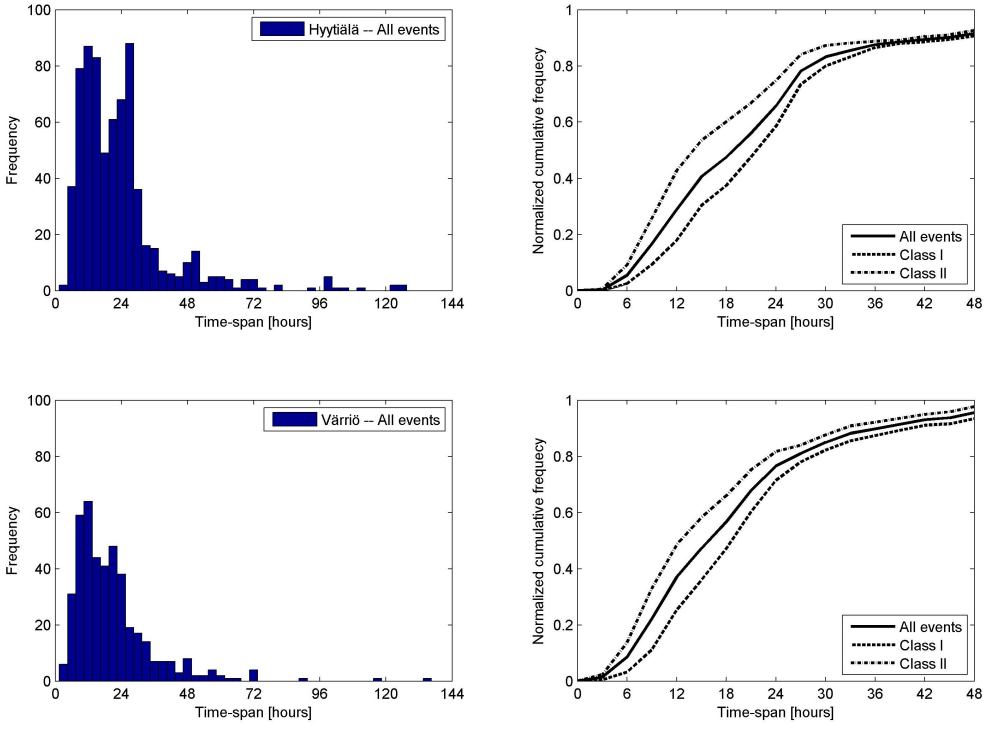

Fig. 3. Time span of NPF events observed at Hyytiälä (upper) and Värriö (lower).

including Värriö, the number of simultaneous NPF events decreased to 50 of which only 9 events were regional identical-NPF events in Finland (Table 2). In order to expand our regional scale to include Southern Sweden we considered Aspvreten and Vavihill during the time period 21 February 2001–20 May 2004, which is the period of available information about NPF events classification at these two sites. During this time period we observed only 9 simultaneous NPF events in Finland and Southern Sweden; we considered only 2 cases as regional identical-NPF events (Table 2). Two cases out of nine clearly implies that it is very rare to have similar local meteorology over a large spatial scale that the simultaneous NPF events are considered identical; keeping in mind that NPF events observed at these measurement stations have the same predominant air masses.

Table 3. Time span of local and regional identical-NPF events over different spatial scales.

Spatial scale	Location	Time span of NPF event [hours]				
		Mean	Std	25%	Med.	75%
Local(1)	Hyytiälä	19	6	14	19	21
	Värriö	17	2	17	17	20
Southern Finland(2)	Helsinki	27	31	18	27	45
	Hyytiälä	30	30	23	30	47
Southern Finland(3)	Helsinki	28	40	18	28	48
	Hyytiälä	30	39	24	30	47
Finland(4)	Helsinki	25	30	17	25	55
	Hyytiälä	30	33	26	30	55
	Värriö	25	17	21	25	36
Finland & Southern Sweden(5)	Helsinki	63	47	30	63	96
	Hyytiälä	76	65	30	76	122
Värriö	23	7	18	23	29	

(1) Local NPF events at a location were identified by including the SMEAR stations and Aspvreten during 31 May 2000–5 September 2004, which is the time period of available NPF classification at Aspvreten.
(2) The event-to-event comparison was performed for Helsinki (SMEAR III) and Hyytiälä (SMEAR II) during 1998–2006.
(3) The event-to-event comparison was performed for Helsinki (SMEAR III) and Hyytiälä (SMEAR II) during 5 May 1997–31 December 2006.
(4) The event-to-event comparison was performed for the SMEAR stations during 1998–2006.
(5) The event-to-event comparison was performed for the SMEAR stations, Aspvreten, and Vavihill during 21 February 2001–20 May 2004.

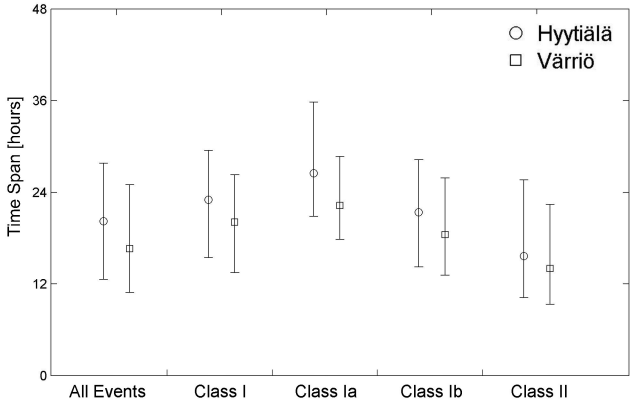

Fig. 4. Time span (median and quartiles) of NPF events with different classes observed at Hyytiälä and Värriö.

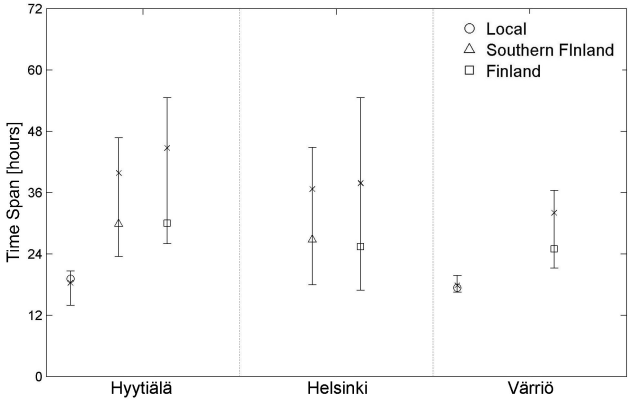

Fig. 5. Time span of identical-NPF events over different spatial scales; see also Table 3. The figure illustrates median and quartiles; the mean value is indicated with (x).

Aiming at observing small spatial scale NPF events we focused on the time period 31 May 2000–5 September 2004. This is the time period for which simultaneous aerosol size distribution data and NPF classifications were available for the Aspvreten, Värriö, Hyytiälä, and Helsinki stations. During this time period we found 10 small spatial scale NPF events at Hyytiälä and only 5 small spatial scale events at Värriö (Table 2). Note that these small spatial scale events were identified based on the fact that on the same day there were no events or indication of events at other sites included in this study. Even though we identified these events as small spatial scale, we believe they occurred over a certain area smaller than the spatial scales assumed in this study.

According to observations on the time span of regional NPF events occurred on a large spatial scale (e.g. over Finland) the newly formed mode of aerosol particles could grow to the accumulation mode size-range (diameter between 0.1 and 1.0 μm) and remained distinguishable in the atmosphere for a long time; from a day to several days (Table 3 and Fig. 5). Coagulation (in the early stage of the event) and condensation (also after few hours of the start of the event)

processes are expected to be pronounced during such events and it takes a long time before the newly formed aerosol particles are removed by the incoming air masses because aerosol particles retain similar physical characteristics over a large spatial scale. It is therefore expected that the growth pattern of aerosol particles continues further if the air masses remain similar or have a chance to circulate over the same area where aerosol particles are formed on a regional scale. Tunved et al. (2003) also confirmed that the clear growth pattern after a NPF event is an indication that it occurs over a large area; i.e. a regional event. According to Dal Maso et al. (2007) the newly formed aerosol particles have a chance to grow if the concentrations of pre-existing aerosol particles are low enough resulting in low condensation and coagulation sinks.

When the spatial scale of NPF events was small the time span of the event was shorter than 20 h; as clearly observed at Hyytiälä and Värriö (Table 3 and Fig. 5). Furthermore, the number concentration of the newly formed mode declined rapidly because such events were localized within a small area where the newly formed particles could be easily removed with the incoming air which was likely having different characteristics of aerosol particles. Because the time span of small spatial scale events is short due to convection and dilution with the incoming air, the newly formed mode is not expected to have the opportunity to grow to bigger sizes beyond the Aitken mode size-range; i.e. bigger than 65 nm in diameter.

Some studies comparing particle formation at several stations have been carried out by using less number of stationary measurement stations or shorter time period of measurements than what was used here in this study. For example, Komppula et al. (2006) compared stations in Northern Finland and reported that even though local meteorology affected the NPF characteristics the NPF event itself remained to hold clear regional characteristics. Stanier et al. (2004) compared simultaneous NPF events at an urban background site and a rural site (38 km away) in Pittsburgh, USA during two months. They showed that most of the simultaneous NPF events were similar at both sites. In Leipzig, Germany and at 3 rural stations, which were maximum 50 km apart, there were three days with homogeneous NPF events recorded as well as one inhomogeneous (Wehner et al., 2007). However, a thorough statistical analysis of the frequency of both types of events has not been attempted except for a study by Birmili et al. (2003) that considered two locations with 3 km horizontal distance in between and 300 m vertically apart; obviously the observed events were strongly homogeneous.

3.2.2 Analysis based on the time span and back-trajectories

Based on the time span and the history of back-trajectories we estimated the minimum spatial scale of each event at Hyytiälä (850 events) and Värriö (535 events) separately. We

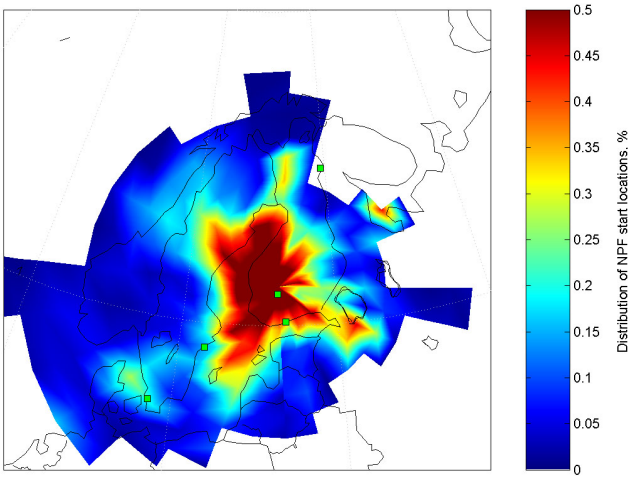

Fig. 6. The normalized distribution of spatial scale of regional NPF events observed at Hyytiälä during 1997–2006. This figure was created from more than 16 000 trajectories.

then estimated the probability of the minimum spatial scale of all events as illustrated in Figs. 6 and 7. This kind of analysis revealed that most of the regional NPF events observed at Hyytiälä started in a sector N-SW. The likely-hood probability for larger spatial scale NPF events taking place SW of Hyytiälä appears to be small (Fig. 6). Our analysis also showed that regional NPF events are often confined east of the Norwegian coast. This makes sense because a number of studies have emphasized the role of the forest in NPF over Scandinavia (e.g. Tunved et al., 2006b). We estimated that about 60% of the NPF events can be traced to locations beyond 220 km and only 10% of the NPF events can be traced beyond 550 km from Hyytiälä. Another interesting result is that a significant number of NPF started at locations over the Baltic Sea and the strait between Denmark and Sweden. This suggests that sulfur rich sea-traffic emission along the busy transport routes in the Baltic Sea might have a role in these events. This finding has also been observed by Kristensson et al. (2008) for the Vavihill station by employing a similar method with back trajectories.

Similar analysis applied to Värriö revealed that the spatial scale of the regional NPF events does not exceed the coast line. This behavior strongly supports that land-based emissions are required for the NPF events to occur. This also suggests that the forest emissions are a likely candidate to support the NPF events. Some of the NPF events can likely be attributed to the heavy sulfur emitters located on the Kola Peninsula, e.g. Monchegorsk. At Värriö, about 40% of the regional NPF events had a spatial scale of 110 km from Värriö and roughly about 28% of the events were actually formed farther away than 220 km. This means that the typical spatial scale of the events observed at Värriö is somewhere between 110–220 km (for Hyytiälä it was 200–300 km).

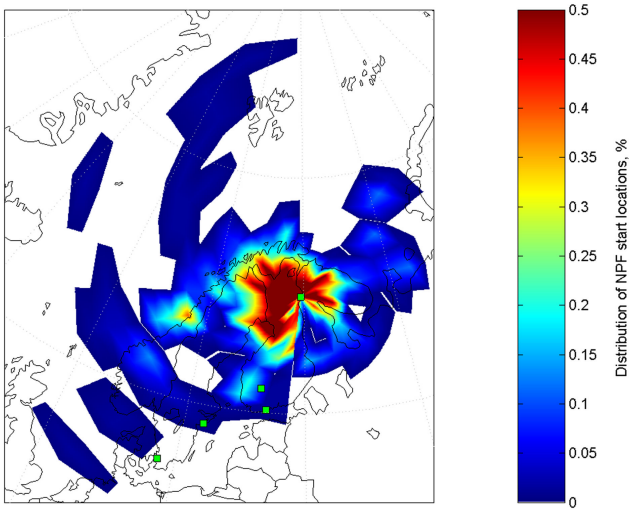

Fig. 7. The normalized distribution of spatial scale of regional NPF events observed at Värriö during 1998–2006. This figure was created from more than 7000 trajectories.

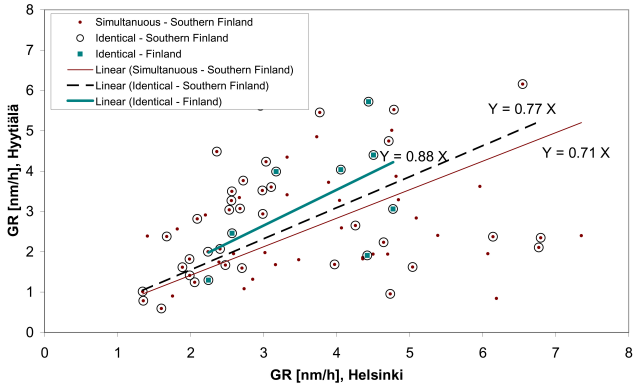

Fig. 8. The growth rate of aerosol particles observed at Hyytiälä versus that at Helsinki during the same events over different spatial scales.

The difference between Värriö and Hyytiälä regarding the spatial scale of NPF events may be explained by the fact that Värriö is located relatively close to the ocean, and is situated at the northern rim of the Boreal forest zone. This means that the time the air is allowed to spend over the sources of nucleating material is limited, and thus the spatial scale of the NPF events will become smaller. Of course, different meteorology and different emission patterns in the typical source areas of Hyytiälä vis-à-vis Värriö may add to the difference.

Alike the situation with the spatial scale calculated using observations within a network of stationary measurement stations, there are few international studies providing data for the spatial scale using the analysis of time span and the history of back trajectories. The two studies available for Leipzig and a German mountain site (Wehner et al., 2007 and Birmili et al., 2003 respectively) showed that the spatial scale can be as high as a few hundred km for those NPF events that show clear growth. There were too few data available in these studies, to provide enough statistical data or to be able to plot the starting locations of the NPF events as in this study.

According to our scale, local events have very short time span and the newly formed aerosol particles are dispersed in the background atmosphere quickly. A good example of such events is secondary particle formation due traffic emissions in cities. On the other hand, regional events tend to have long time span because the atmospheric composition is homogeneous over a large area where the event takes place. However, analysis of air masses at a single observation point only provides information on the minimum spatial scale of the event. For example, if the air masses circulate within a certain region, the atmospheric composition is expected to be homogeneous and thus the newly formed aerosol particles are expected to experience continuous growth pattern as long as the air masses remain within the same region (a good example is the case study presented in Sect. 3.3.2); though, the atmospheric composition is homogeneous over a larger area than the circulation region of air masses. On the other hand, prevailing air masses from the same direction during a regional NPF event also provides information on the spatial scale of the event along the path of the incoming air masses only. In that sense, multiple sites analysis provides more accurate estimation on the spatial scale of an event (a good example is the case study presented in Sect. 3.3.1). When considering multiple sites, the occurrence of a simultaneous regional NPF event at different locations requires homogeneous composition of the atmosphere as well as similar local meteorological conditions. Furthermore, the occurrence of simultaneous regional identical-NPF events requires similar concentrations (or at least the minimum threshold of concentration) of precursors at the different sites.

We further investigated the growth rates as estimated for Hyytiälä and Helsinki (data taken from previous studies cited in this article) during simultaneous regional events over Southern Finland (Observed at Hyytiälä and Helsinki) and over Finland (Hyytiälä, Helsinki, and Värriö). In general, the estimated growth rate at Helsinki is bigger than that at Hyytiälä as clearly shown in Fig. 8 for the simultaneous events – Southern Finland. This is mainly due to the higher concentrations of condensable vapors as a result of the urban sources in Helsinki. However, the proportional relationship becomes closer to 1:1 between the two sites (Helsinki and Hyytiälä) when we select the identical events over Southern Finland; . and it further improves when selecting the identical events over Finland (Fig. 8; identical events – Southern Finland). The proportional relationship even improves when considering larger scale of events (Fig. 8; identical – Finland). This can be explained as follows: (1) the regional characteristics of aerosols and precursors become significant during regional identical events and (2) local sources, which

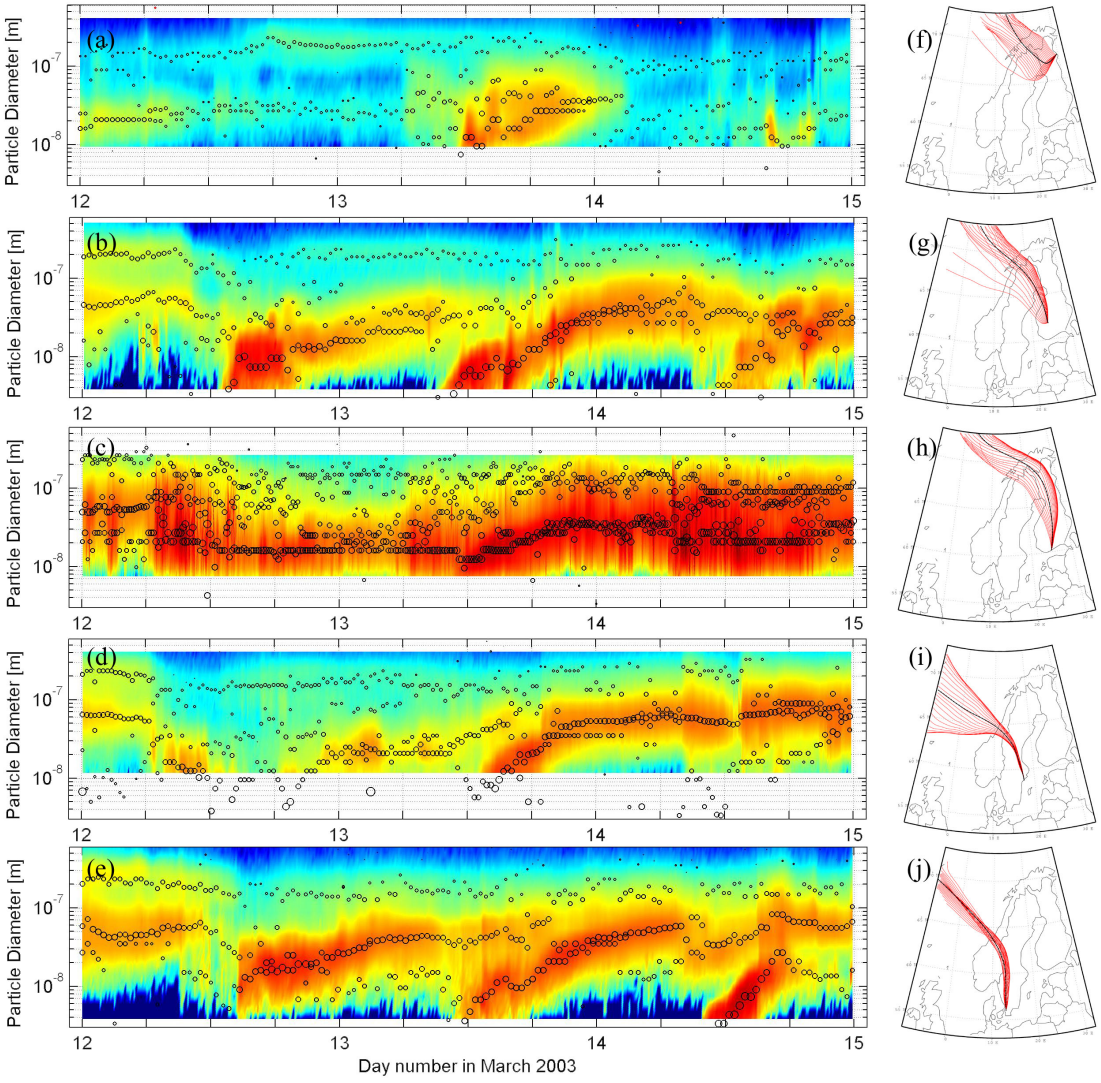

Fig. 9. Regional identical new particle formation event observed on March 13, 2003 at **(a)** Värriö, **(b)** Hyytiälä, **(c)** Helsinki, **(d)** Aspvreten, and **(e)** Vavihill. **(f–j)** corresponding 2-days back-trajectory for each hour in these sites. The circles on the particle number size distribution spectrum represent the mode geometric mean diameter ($D_{pg,i}$) and the size of the circles represents the mode number concentration (N_i).

are the sources of other condensable material, become less significant during regional events.

3.3 Case studies of regional NPF events

3.3.1 A regional identical-NPF event over Finland and Southern Sweden

In this study we observed only two regional identical-NPF events over Finland and Southern Sweden: 13 March 2003 and 5 September 2003. Here we presented only one of them (13 March 2003) with a detailed analysis. The event started around noon at all sites as clearly seen from the geometric mean diameter (D_{pg}) that was obtained by the multi-lognormal fitting (Fig. 9). Even though the measured size-range of the particle number size distribution was not the same at all sites, the temporal behavior of the D_{pg} indicates that the event started at the same time at all sites, which qualifies it as an identical regional NPF event. The event time span was similar (around 20–24 h) at the background stations Hyytiälä, Aspvreten, and Vavihill but it was the shortest at Värriö (around 14 h).

The newly formed aerosol particles grew to the Aitken mode size-range (25–100 nm) during the night (Fig. 10). The estimated growth rate of the newly formed aerosol particles varied between 2 and 3.5 nm/h and the observed formation rate was about 1.2, 1.3, and 0.5 particles/cm^{3}s respectively at Helsinki, Hyytiälä, and all other sites (Aspvreten, Värriö, and Vavihill). The observed differences between the events at the five sites could be due to (1) aerosol data quality and

processing, including mainly multi-lognormal fitting and the lower limit of the measured size distribution, and (2) local meteorology.

The reason for observing these events at the same time at all sites is mainly related to the history of the air masses. The air masses that arrived in Helsinki, Hyytiälä, and Värriö were from the Atlantic Ocean and they crossed over North Norway and Sweden whereas the air masses that arrived in Aspvreten and Vavihill crossed over middle Norway and Sweden and they were also originated from the Atlantic Ocean. The air masses relatively spent similar time over land (Fig. 9). On the following day the air masses remained rather similar for all sites except for Värriö, and that explains the short time span of the event at Värriö. As all sites were influenced by similar air masses, they received similar gaseous precursors and most likely the regional NPF events are expected at all sites when suitable ambient conditions are available for gas-to-particle conversion. It is, therefore, expected that the newly formed aerosol particles due to such regional NPF events have similar chemical and physical properties in the initial steps of the formation, and thus they are identical.

3.4 A regional NPF event over Southern Finland with a long time span

We observed 25 regional identical-NPF events over Southern Finland where the time span of the regional event was longer than 3 days. The four most interesting cases were the events with time span longer than 6 days: 4 June 1997 (about 10 days), 30 July 1999 (about 6 days), 9 May 2002 (about 6.5 days), and 5 August 2002 (about 8 days). These events were observed during the summer season (May–August). According to our analysis, these events had a long time span mainly because of two reasons: (1) the air masses were continuously from the Atlantic Ocean crossing over the northern part of Scandinavia during the first couple of days and (2) the air masses were confined within Southern Finland during the following days after the newly particles were formed. In this study we will present detailed analysis on the event that started on 4 June 1997, though other events can be as important as this one.

The event on 4 June 1997 was followed by three regional events during 5–7 June 1997 (Fig. 11a). In fact, daily NPF events were also observed in Hyytiälä during 25 May–7 June, 1997 but the strongest event was on 4 June 1997 with an observed formation rate (J_{nuc}) of about $1.03\,\mathrm{cm^{-3}s^{-1}}$ in Hyytiälä and $4.77\,\mathrm{cm^{-3}\,s^{-1}}$ in Helsinki. By the beginning of June 6 the newly formed mode of aerosol particles had grown to a D_{pg} of about 100 nm and after that it continued growing slowly to about 250 nm by mid June 14; i.e. the span time of the event was about 10 days. The growing mode was also observable in Helsinki where it also showed very similar dynamic behavior as that in Hyytiälä (Fig. 11b).

According to the 2-days back-trajectory analysis for Hyytiälä, during 5–14 June the air masses had a mixed origin within Southern and middle Finland (9 and 11–13 June), from the Baltic Sea and Southern Sweden (6–8 June), and from Russia and Eastern Finland (10 and 14 June); see for example Fig. 11d–g.

According to our observation for this long-term data set, the air masses originating from the Baltic Sea and Nordic countries are typically not loaded with high concentrations of accumulation mode and Aitken mode particles when arriving at Hyytiälä; the concentration might vary between 1000–$2000\,\mathrm{cm^{-1}}$. The number concentration of the accumulation mode (diameter between 0.1–1.0 μm) was about $100\,\mathrm{cm^{-3}}$ during 4 June 1997 and started to increase steadily to about $1000\,\mathrm{cm^{-3}}$ during 5 and 6 June 1997 as a result of the growing aerosol particles that originated from the regional NPF events on 4 and 5 June. After 7 June the accumulation mode concentration varied between 1000 and $2000\,\mathrm{cm^{-3}}$ as a result of the already existing mode from the previous regional NPF events and also due to transported aerosol particles within South and middle Finland (Fig. 11). It is pronounced here that the growing mode of aerosol particles originated from the regional NPF events on 4 and 5 June remained distinguishable from other modes originated from other sources in the region. For example, during 10 June (air masses from Russia and east Finland) we could distinguish two modes: a clearly growing mode with D_{pg} between 150 and 200 nm, which is the one from the regional NPF events, and another mode with rather variable D_{pg} that is most likely due to the anthropogenic emissions.

One interesting feature of this event covering a long time span is how it illustrates the suppression of particle formation on the following days. As the first nucleated particle mode grows, it forms a condensation sink, which grows together with the increasing particle diameter, despite the slow decrease of the particle number. This sink possibly acts as a sink for both nucleating vapors and fresh particles or clusters, and therefore particle formation on subsequent days is progressively weaker and finally ceases altogether. Despite the lack of particle number production, the aerosol mass continues to grow, demonstrating the availability of condensable vapors.

As a simple exercise, we estimated the mass concentration of each size-fraction by assuming the particles are spherical with unit density (Fig. 12). The ultrafine fraction contributed 30–35% of the total mass between 3–400 nm during the first three days of this NPF event. The mass concentration of the accumulation mode particles varied from $\sim 1\,\mu\mathrm{g/m^3}$ to $\sim 15\,\mu\mathrm{g/m^3}$ during 5–14 June. In fact, after the fourth day the mass concentration was more than $5\,\mu\mathrm{g/m^3}$. According to Laakso et al. (2003) the annual mean PM_1 concentration at Hyytiälä is $\sim 4.3\,\mu\mathrm{g/m^3}$, and our estimation for the total mass of the fine fraction is a clear indication that regional NPF events can have significant impacts on the aerosol particle burden over large areas during and after the event. This fact should be taken into account in regional models because we believe that a significant fraction of regional aerosol particles

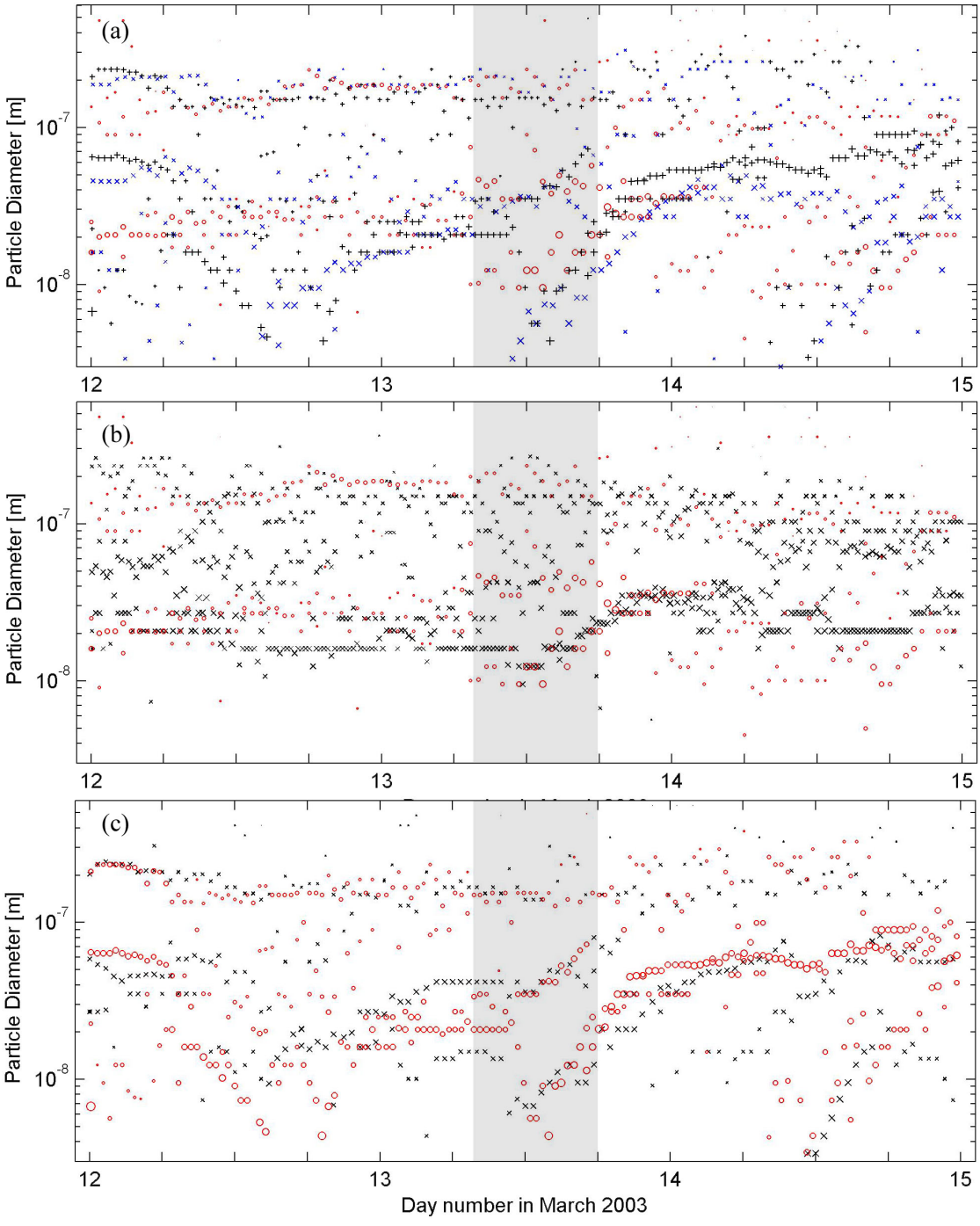

Fig. 10. The temporal variation of the geometric mean diameter ($D_{pg,i}$) during the regional new particle events presented in Fig. 9. **(a)** Remote background sites: Värriö (circles), Hyytiälä (x), Aspvreten (+). **(b)** Remote background site Värriö (circles) and urban site Helsinki (x). **(c)** Remote background sites in Southern Sweden Aspvreten (circles) and Vavihill (x). The shaded area represents the initial stage of the NPF event.

is produced during regional NPF events and not only transported over long distances.

Here we should point out the difference between regional aerosol particles produced during regional NPF events and those from LRT. It is expected that the temporal variation of regional aerosol particles originated during regional NPF events is rather similar within the region of interest; i.e. they should have similar temporal variation without time-lag from

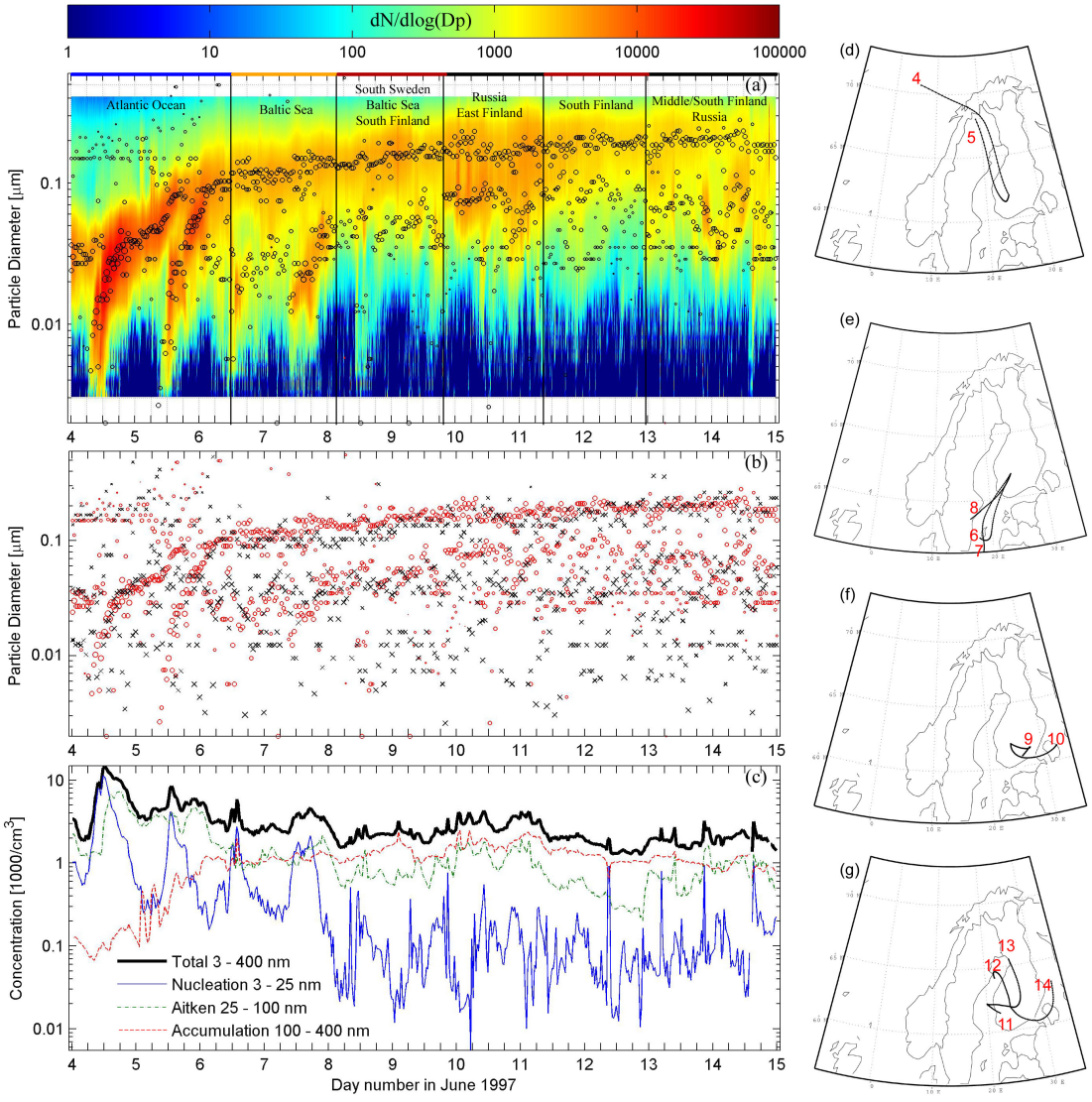

Fig. 11. The dynamic behavior of aerosol particles during 4–14 June 1997 after several intensive regional new particle formation events observed in Southern Finland: **(a)** particle number size distribution; the circles represent the mode geometric mean diameter ($D_{pg,i}$) and the size of the circles represents the mode number concentration (N_i). **(b)** Comparison between the dynamic behavior of aerosol particles in Helsinki (x) and Hyytiälä (circles). **(c)** Particle number concentrations of different fine modes. **(d–g)** 2-days back-trajectories; the number represent the day number in June 1997. The air mass origin is also marked on (a).

area to another within the same region. On the other hand, aerosol particles transported from another region outside the area of interest (i.e. LRT episodes) appear with similar temporal variation but with time-lag over different areas in the region of interest depending on the traveled distance of the air masses from the source origin. The concentration of aerosol particles during a LRT episode also decreases while the air masses move away from the source.

4 Summary and conclusions

We investigated the time span and spatial scales of new particle formation (NPF) events in Finland and Southern Sweden. Our analysis consisted of two novel approaches, high quality measurements of submicron particle number size distribution at five stationary measurement stations, back-trajectories, and observation of NPF events during long-term periods. The first approach was based on observation within a network of measurement stations and the second approach was based on the time span and the history of back trajectories. These two approaches enabled us to quantify the spatial

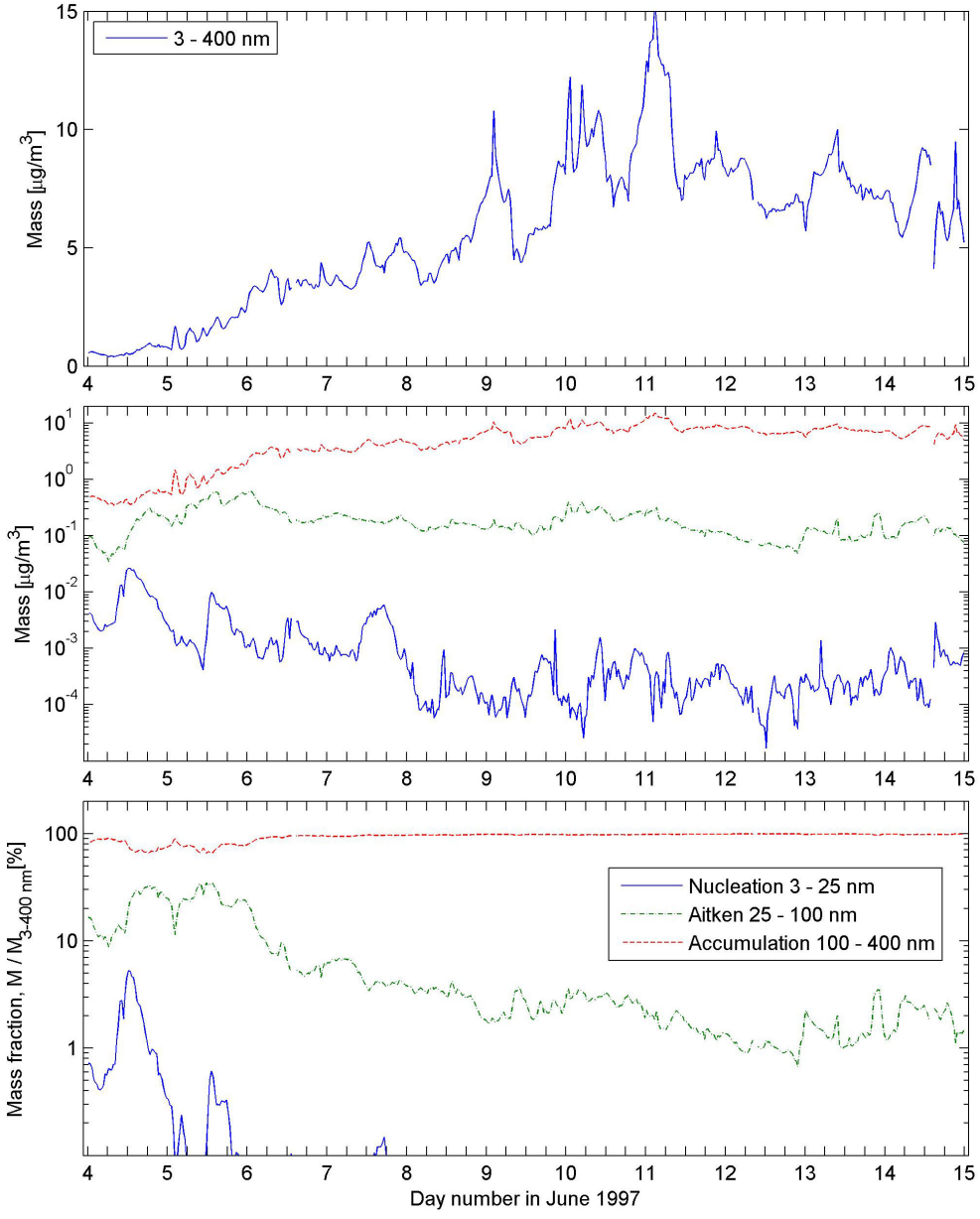

Fig. 12. Estimated mass concentrations within **(a)** the measured size-range 3–400 nm and **(b)** the size-fractions of nucleation, Aitken, and accumulation modes. **(c)** The mass fraction of each size-fraction of the nucleation, Aitken, and accumulation modes. See also Fig. 11. Note that (b) and (c) share the same legend.

scale of regional NPF events and the occurance of regional identical-NPF events. According to this study the time span is defined as the time period from the first moment when the newly formed mode of aerosol particles is observable below 25 nm until the newly formed mode is not any more distinguishable from other background modes of aerosol particles after growing to bigger sizes. During large spatial scale events, the time span of the event can be followed for more than 30 h.

According to the first approach which was based on the observations at the five Nordic stations, NPF events are often observed at several locations simultaneously, meaning there were favourable conditions for NPF over a large domain and the events cover a large spatial scale. However, the events were rarely identical in terms of the start time of the events and their temporal variation, which means that differences in local meteorology, air masses, source strengths and geography drives a local temporal behaviour for these events.

According to the second approach, about 60% and 28% of the observed NPF events could be traced back to locations beyond 220 km at Hyytiälä and Värriö respectively. This study agreed with our previous observations that NPF events started over the continents but not over the Atlantic Ocean (Tunved et al., 2006b). On the other hand, the sulfur rich sea-traffic emissions along the busy transport routes in the Baltic Sea probably has a role in the events observed at Hyytiälä as previously suggested for Vavihill in southern Sweden (Kristensson et al., 2008).

In this study we reported empirical evidence that the newly formed aerosol particles may remain distinguishable for a long time (more than 3 days) in the atmosphere. According to the Nordic conditions, such events had a long time span mainly because the air masses were first continuously from the Atlantic Ocean, which is highly propable for successive daily events, and the air masses were confined within a certain region during the following days after the newly particles were formed. As illustrated by a case study, which is representative for such events, the newly formed aerosol particles can grow to 0.25 μm in diameter. This is clear evidence that regional aerosol particles originating from such regional NPF events have a significant fraction in the particulate mass concentration of fine particles (PM_1). In the same case study, the ultrafine fraction of aerosol particles contributed to about 30% of the PM_1 concentration during the initial stage of such intensive regional NPF events.

The results obtained in our study holds valuable information to be used in the development of regional and global aerosol models. The estimate for the spatial scale of the particle formation process can, for example, be used to test whether the particle formation mechanisms used by such models can reproduce the correct area where such formation is taking place. The observation of a new mode growing far into accumulation mode sizes can also be used to compare the modeled growth process of new particles to measurements over almost the whole lifetime of such particles; this is often not possible with fixed measurements.

While the Eularian approach from this study provides initial evidence that regional NPF events can have a large impact on the accumulation mode population of particles, only a limited amount of data can be used where we can follow the air masses for several consecutive days. As a next step in the development to better understand the impact of NPF events, a Lagrangian type of study is recommended using the densely spaced EUSAAR European size distribution data. Especially the air movement along a line extending from Italy across Germany and Southern Sweden all the way to Hytiäälä and Värriö in Finland can be examined as the air moves both in the north-south as well as in the south-north direction. Several years of data should be used to provide a statistically sound analysis of the north-south and south-north transport.

Appendix A

Classification scheme for new particle formation events

Dal Maso et al. (2005) developed a classification scheme for identifying new particle formation (NPF) events in remote regions. An NPF event is identified if a distinctly new mode of aerosol particles is observable in the nucleation mode size-range (diameter $<$25 nm) for at least several hours and it must show a growth pattern. An additional criterion is the possibility to quantify basic characteristics such as the particle growth rate (GR) and formation rate (J_{nuc}). Therefore, the evolving nucleation mode should be clearly distinguishable for a sufficient time period to ensure that we have enough data points for the quantitative analysis. Based on the accuracy of the quantitative analysis there are two main classes of NPF events: Class I when both the growth rate and formation rate are determined with a good confidence level. Class II when the derivation of the growth rate and/or the formation rate was not possible or the accuracy of the results was questionable. Class I events are subdivided into two subclasses according to the possibility of modeling the event or not. In general, Class Ia events are clearer than Class Ib events.

A *"non-events"* class is identified whenever it is evident that no new particles are formed. However, many days do not fulfill the criteria for either an event or a non-event; instead an *"undefined"* class is introduced. In other words, the *undefined* class contains cases, for example, when newly formed particles occur sporadically in the nucleation mode size-range or when we clearly see the later phase of a mode growing in the Aitken mode size-range.

Even though this classification scheme was developed for remote sites, we believe that a modified scheme should be available for urban sites. As reported by Hussein et al. (2008), the observation of weak NPF events in Helsinki was not possible due to high concentrations of pre-existing ultrafine particles (UFP, diameter $<$100 nm) and also aerosol particles from long-range transport (LRT) origin.

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