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Probability of nucleation events and aerosol particle concentration in different air mass types arriving at Hyytiälä, southern Finland, based on back trajectories analysis

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The probability of a new ultrafine particle formation as well as the spatial sources of the nucleation, Aitken and accumulation modes of particles measured in Hyytiälä, southern Finland, were investigated using air mass history analysis. The studied period covered the years 1997–2003. We estimated the probability of a new ultrafine particle formation for nine sub-areas inside a domain limited by 50°N–80°N in latitude and 30°W–60°E in longitude. For all the seasons North Atlantic and arctic regions dominated as an origin of air masses that accompany nucleation events. The probability of nucleation events in Hyytiälä reached its maximum in spring and summer with north and north-west air masses transfer directions. The highest concentration of nucleation mode particles was observed in arctic and polar air masses, whereas the highest concentration of accumulation mode particles was observed during intrusions of continental air masses.

Introduction

The formation of new ultrafine particles in the atmosphere is of growing interest due to their potential climatic and health effects. In recent years the formation and growth of ultrafine aerosol particles have been observed at many different sites around the world (Kulmala *et al*. 2004). However, the mechanism of new-particle formation is not completely clear. The nucleation processes depend strongly on meteorological properties and chemical composition of the air mass in which they occur (Nilsson *et al*. 2001, Kulmala *et al*. 2004). To investigate the conditions leading to nucleation, knowledge of the air mass origin and transport are of great importance.

Back-trajectory analysis is widely used in studying the long-range transport of atmospheric pollutants (Siebert *et al*. 1998), in source–receptor investigations (Hopke *et al*. 1995, Stohl 1996, Sciare *et al*. 2003), in probability analysis (Ashbaugh *et al*. 1985) as well as in characterization of properties of air masses arrived at a recipient site (e.g. Kulmala *et al*. 2000). Virkkula *et al*. (1997) estimated the number concentration of particles with diameters > 14 nm in marine and continental air and identified source areas of SO_2 concentration in Finnish Lapland for the period

of January 1992 to June 1994. Chemical reactions and deposition sink can also be estimated in Lagrangian (Bates *et al*. 1998) and pseudo-lagrangian (Nilsson and Leck 2002) studies. Using back trajectory data, Birmili *et al*. (2001) defined the role of air masses in particle size distribution. Tunved *et al*. (2003, 2004) used a cluster trajectory analysis for one-year aerosol size distribution data to explain changes in aerosol properties.

The objective of this study is to improve our understanding on the spatial and meteorological factors influencing nucleation processes. We aim to explain the results of long-term aerosol particle size distribution measurements at the SMEAR II station in Hyytiälä, southern Finland (Dal Maso *et al*. 2005), as a consequence of air mass transport. In this paper we present a spatial analysis of the history of air masses arrived at Hyytiälä during the period 1997–2003 to reveal regions that can be considered either as the sources of conditions leading to the new particle formation or as source areas for particles in the nucleation mode size range.

Material and methods

Site and instrumentation

The SMEAR II station (61°51´N, 24°17´E, 180 m above sea level) is located at the Hyytiälä Forestry Field Station of Helsinki University in a homogenous Scots pine stand (Vesala *et al*. 1998, Kulmala *et al*. 2001b), 220 km north-west of Helsinki. The natural managed stand was established in 1962 by sowing after the area had been treated with prescribed burning and light soil preparation. The station is located inside a boreal coniferous forest. The air quality at the site represents typical background conditions. The local pollution from the station buildings (0.5 km away from the measurement site) and the city of Tampere (60 km away), both located west-south-west of the station, affect occasionally the air quality at the SMEAR II station. The station consists of four operational blocks: a 73-m-high mast for atmospheric measurements, a 15-m-high tower for tree measurements, water catchments and aerosol instrumentation (Vesala *et al*. 1998).

Continuous measurements of aerosol number size distributions have been performed at the station since 1996 (Mäkelä *et al*. 1997). The sample air for aerosol measurements is taken from 2 m above the ground next to the measurement building. Aerosol particle size distributions have been measured with a DMPS system (Aalto *et al*. 2001), which consists of two DMA's producing overlapping size distributions, and two CPC's which are used to detect the selected particles. The system covers the particle size range of 3–600 nm and measures with a temporal resolution of about 10 minutes.

Trajectory analysis

To analyze the source and transport pathways of air masses arriving at Hyytiälä (SMEAR II), a trajectory analysis was done for the period 1997–2003. The back trajectories at 250-m arrival height above the ground level at 00:00, 06:00, 12:00 and 18:00 UTC were calculated using the HYSPLIT_4 model, developed by NOAA/ARL (Draxler and Hess 1998). The HYSPLIT_4 model is a single particle lagrangian trajectory dispersion model. The model runs were made using Global FNL meteorological archive with a spatial resolution of 191 × 191 km. Air parcel back trajectories were calculated typically 96 hours backwards in time. However, some trajectories were missing or shorter than 96 hours in duration, since the FNL archive data has some gaps when output meteorological data are not available. Current literature suggests that errors accompanying HYSPLIT-generated trajectories can be anywhere from 15% to 30% of the travel distance (Stohl 1998, Draxler and Hess 2004). In this paper we consider the sources of aerosol particles in a regional scale, so the accuracy of trajectory calculations is sufficient to reveal the differences in the particle concentration for different types of air masses.

To analyze the prevailing air masses transport direction, the territory around Hyytiälä was divided into six sectors starting from the north. However, in a complicated synoptic situation an air mass can change its direction several times on its way to the recipient. The mean position of the air parcel at reference time back steps –12,

–24, –48, –72 and –96 hours was calculated for each 60° sector and was presented as contour lines of distant points at different time steps. For each patch, limited by the direction and reference time, we calculated the frequency (in %) of trajectory passing through the patch.

Classification of nucleation events

Particle formation events were classified according to the particle number concentration and the particle formation and growth rate (Mäkelä *et al*. 2000, Dal Maso *et al*. 2005). Particle formation is registered when a mode of fresh particles appears into the measured size range. Class 1 includes data with a very strong new small particles mode appearing and growing sometimes for several days. The process of new particle formation and growth is also taking place in cases belonging to class 2 but less intensively than in class 1. Class 3 events are days with clear particle formation, but formation and growth rates cannot be obtained from the size distribution data. Besides the event days also days with no particle formation were classified as a separate class, termed the nonevent class. The other days, for which it was not clear whether some new particles had been formed or not, were not taken into the analysis in the present paper.

Probability of the aerosol particle formation events

The analysis of the nucleation event probability at Hyytiälä as a function of the air mass origin and history was done for the area limited by 50°N–80°N in latitude and 30°W–60°E in longitude. This area was divided into nine sub-areas of 10° × 30° in latitudinal and longitudinal directions, respectively (Fig. 1). Such a division corresponds to the one proposed by Kulmala *et al*. (2000), where the analysis of atmospheric trace gases and aerosol concentrations was done for the SMEAR II station for several periods during 1997–1999 using a TRADOS long-range transport model. Kulmala *et al*. (2000) used a similar scale to reveal the areas of the start point of a trajectory and carried out the meteorology and gas

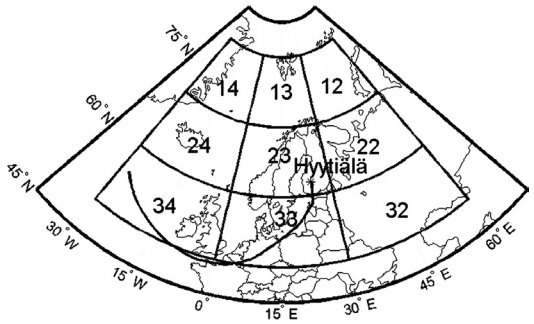

Fig. 1. Schematic figure of the sub-areas used in the trajectory analysis. The location of the SMEAR II station in Hyytiälä is marked with a star.

concentration analysis for five main classes (sub-areas 12, 13, 14 and 24 considered as the Arctic area, sub-areas 33 and 34: central Europe and Great Britain area). In the present investigation the air parcel passing over several sub-areas has been registered in each sub-area (for instance, the trajectory presented in Fig. 1 was counted in sub-areas 34, 33 and 23).

For each sub-area the probability P of event trajectories as a function of event and nonevent trajectories has been calculated as:

$$P = \frac{N_{ev}}{N_{ev} + N_{nev}} \quad (1)$$

where N_{ev} and N_{new} are the numbers of event and nonevent trajectories passed through the sub-area.

We also estimated quantity R given by:

$$R = \frac{N_{ev}}{N_{nev}} \quad (2)$$

which is the ration between the number of event trajectories to the number of nonevent trajectories for each 10° × 30° sub-area.

The effect of air masses history on aerosol particle number concentration

The number concentration of aerosol particles in Hyytiälä was calculated for three size ranges called the nucleation (< 25 nm), Aitken (25–90 nm) and accumulation (> 90 nm) modes. Values corresponding to the trajectory arrival time were assigned to the geographical position

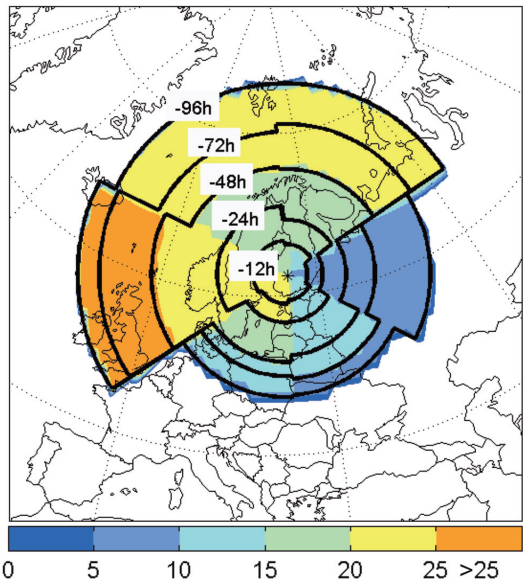

Fig. 2. Frequency (%) of the location of an air parcel in different direction sectors between the reference back time steps (contour line for –12 h, –24 h, –48 h, –72 h, –96 h) for the whole period.

of each one-hour back point of the path. Mean values of the number concentration for areas with a spatial resolution of 2.5° × 5.0° were calculated.

The atmospheric life time of particles varies from hours (nucleation mode) to days (accumulation mode). Recently Tunved *et al.* (2005) estimated the life, or turnover, time of particles of different sizes analysing the particle transport between two Scandinavian measurement stations. They found that during northerly transport the shortest turnover times of 1.6–1.7 days for particles in the nucleation mode and around 2.4 days for particles in the Aitken mode. Nilsson and Rannik (2001) observed a turnover time of around 3.5 days for the Aitken mode in the central Arctic on the basis of eddy correlation aerosol deposition. Thus, in the case of the nucleation mode it is not correct to talk about source regions at distances of several thousands of kilometres from the recipient. However, the origin and transfer direction determine the common properties (temperature, water content, chemical composition, precursor sources, etc.) of air masses that may affect new particle formation.

Results and discussion

Direction of air masses passage

As shown by Tunved *et al.* (2004), new particle formation depends on the properties of air masses determined mainly by their source region. However, moving air masses are transformed with regard to their temperature, humidity and/or stability. This transformation occurs primarily due to the exchange of heat and moisture with the surface over which the air mass travels, due to radiative cooling, and due to processes associated with large-scale vertical motions.

The prevailing atmospheric currents over Scandinavia depend on the location of two quasi-stationary baric systems in the northern hemisphere, the Icelandic Low and Siberian High. As a consequence of the location of the pressure maximum and minimum, air masses originating from the North American continent go through North Atlantic to the region of Iceland, Norwegian Sea and Scandinavia, and finally to the Arctic. The Scandinavian region is characterized mainly by western and south-western atmospheric flows, bringing air masses from the Atlantic Ocean or from eastern and central Europe.

At Hyytiälä, located in southern Finland, the prevailing western atmospheric flows are clearly seen (Fig. 2). More than 25% of the air masses arriving at Hyytiälä have originated or passed over the Atlantic and Great Britain. The fraction of air flows originating from the North Atlantic and the Arctic was about 20%–25%. The frequency of trajectories coming from Russia and central Europe is much lower and the distance traveled is much shorter than those of air masses coming from the Atlantic Ocean. However, baric systems and consequently the transport directions at each specific moment can be essentially different from those estimated by averaging. Local wind directions can also differ from prevailing directions under the impact of the local features.

Although the pressure distributions during the different seasons retain many of the features of average annual conditions, there are some noteworthy departures. Pressure belts and cells shift southward in January and northward in July. In January a strong cell of high pressure

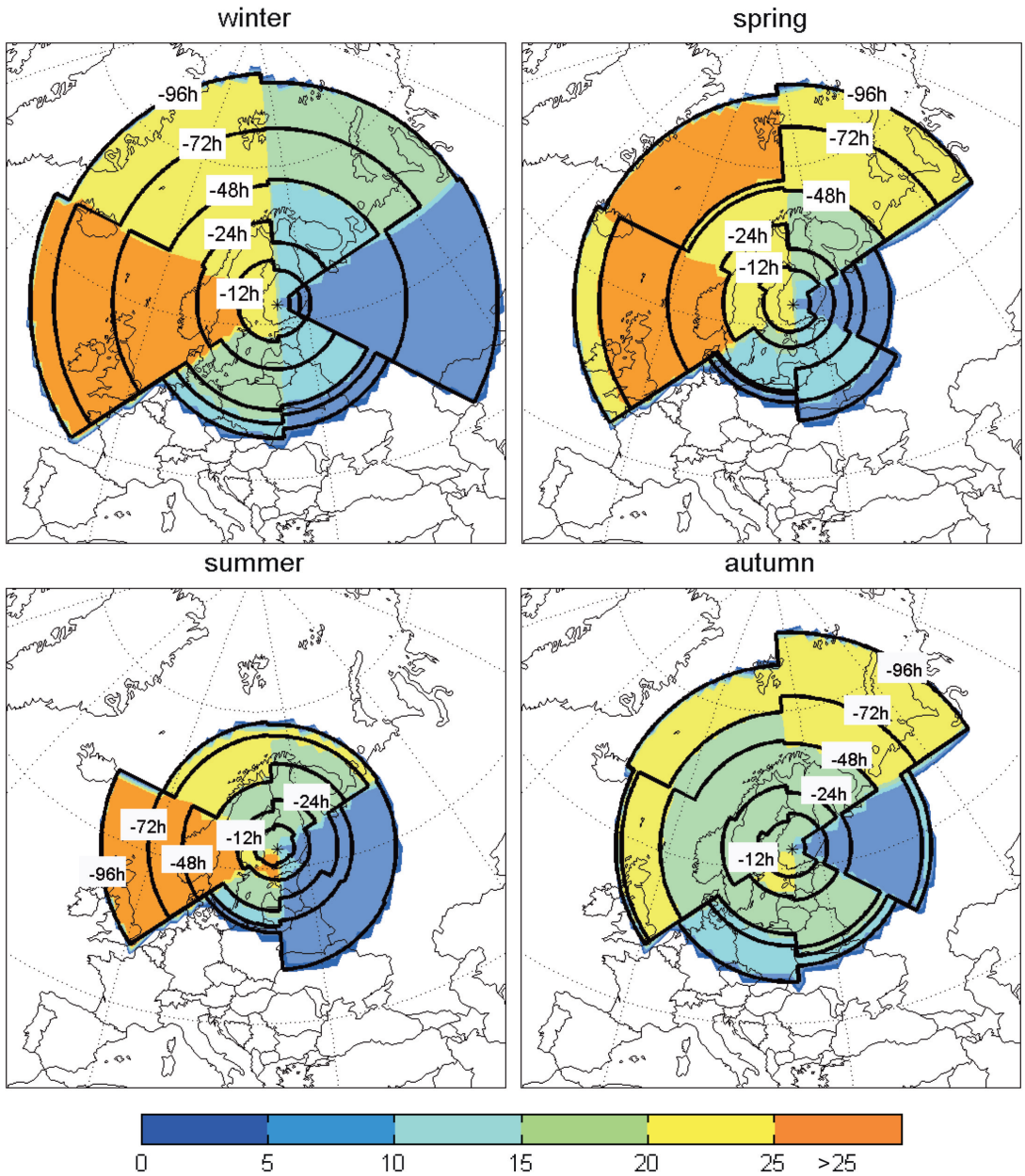

Fig. 3. Frequency (%) of the location of an air parcel in different direction sectors between the reference back time steps (contour line for –12 h, –24h, –48 h, –72 h, –96 h), for all the seasons.

develops over the higher mid-latitudes of the cold Eurasian continent. The pressure gradient is steeper in the winter season compared with other seasons. At Hyytiälä the most probable air flow directions in winter are between south-west and north (Fig. 3). The fraction of air masses originating from over the Eurasian continent is very low (5%–15%).

In July the Eurasian continent develops a weaker thermal low at lower latitudes, as a result of which the atmospheric circulation is less intensive. Since the horizontal pressure gradient is flatter in summer than in winter, the speed of air flow is lower and air masses overpass shorter distances during a given time period. The influence of underlying territories becomes more

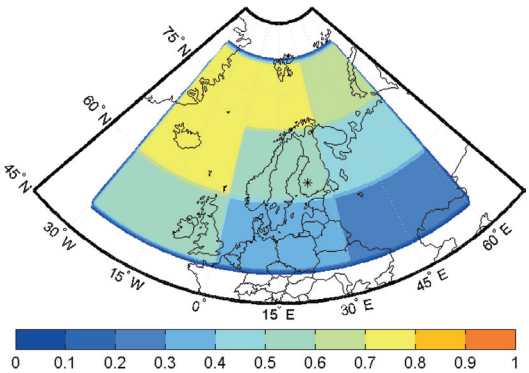

Fig. 4. Distribution of nucleation event probabilities in 1997–2003. The colours in grid cells describe the probability of nucleation event at the receptor location for corresponding trajectory direction.

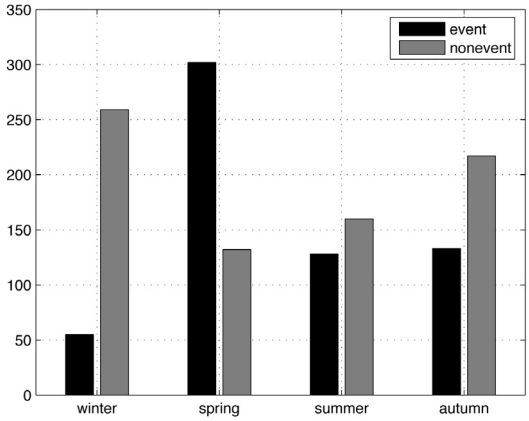

Fig. 5. Seasonal frequency of the days with nucleation events and nonevent days at SMEAR II (Hyytiälä) station in 1997–2003 (Dal Maso *et al.* 2005).

significant. However, the intensive development of a boundary layer in the warm season causes a strong mixing of the lower troposphere, which results in the "cleaning" of air masses from anthropogenic pollution.

Probability of particle formation events

We have investigated the probability of particle formation events as a function of the history of air masses arrived at Hyytiälä (Fig. 4). The highest (0.7–0.8) probability of new particle formation events in the SMEAR II station was observed in arctic and polar air masses coming from North Atlantic and Arctic regions (sub-areas 13, 14 and 24). This is consistent with the earlier findings that the role of cold and clean arctic and polar air masses in nucleation processes is important (Kulmala *et al.* 2001b, Nilsson *et al.* 2001). Arctic air masses originate from snow- and ice-covered regions. The specific moisture of these air masses is low, since sources of water vapour are limited and low temperatures restrict the amount of water vapour that the air mass can contain. The water vapour content remains low even when the air is saturated. Maritime polar air, which reaches Hyytiälä in western atmospheric flows, originates from over the North Atlantic and the Arctic. It starts as very cold and dry but has a long history over the ocean before reaching Scandinavia. In winter when water surfaces are covered by ice, the properties of maritime polar air do not differ a lot from those of arctic air. However, in other seasons the temperature and water content of polar air masses may increase noticeably when passing over water surfaces.

The smallest (0.2–0.4) probability of nucleation events is associated with continental air masses originating from Europe and central Russia (sub-areas 22, 32 and 33). Although continental air is usually drier it is also warmer as compared with maritime air. In addition, anthropogenic activities make it more polluted. New particle formation is suppressed by condensation and coagulation processes and occurs therefore rarely in continental air (Nilsson *et al.* 2001, Komppula *et al.* 2003).

Since nucleation events have a clear seasonal dependency in Hyytiälä (Fig. 5), and since the activity and structure of atmospheric circulation have a seasonal variability, the analysis of probability was also done for different seasons (Fig. 6). The dominant role of arctic and polar air masses in new particle formation can be seen during all the seasons. The seasonal distribution of nucleation events in Hyytiälä for prevailing west–north atmospheric flows is closely related to the increasing of the role of the Arctic and North Atlantic.

In winter, the highest probability of nucleation events (up to 0.5) was observed in air masses arriving from the Atlantic. Air masses coming from Europe and central Russia posed rarely conditions favorable to new-particle formation. In spring, nucleation events were observed with

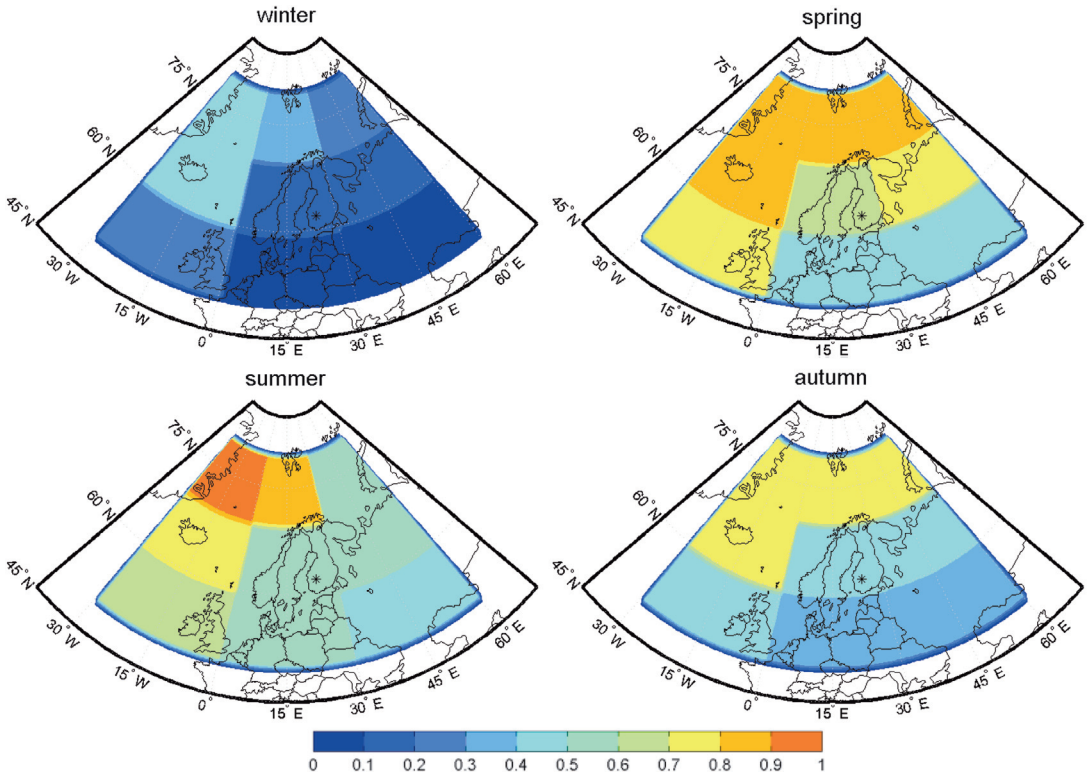

Fig. 6. Seasonal distribution of nucleation event probabilities in 1997–2003. The colours in grid cells describe the probability of nucleation event at the receptor location for corresponding trajectory direction.

a probability between 0.8 and 0.9 when air masses had originated or passed over the Arctic and the North Atlantic. Smaller (0.7–0.8) probabilities were observed in air masses coming from the Great Britain region or from the north of the European part of Russia, whereas probabilities between 0.4 and 0.5 were observed in air mass originating from Europe and the central European part of Russia.

In summer, when the frequency of arctic air masses outbreaks becomes rare, the role of north regions in nucleation was getting more diverse. Lower (in comparison with that in winter) temperature gradient is directed to the south-east. Smearing and displacement of the polar front zone reduces the differences of air masses properties, arrived at Hyytiälä from south-west and north-east directions. However, in comparison with that in spring the new particle formation probability became even higher in summer in the case of air masses originated over east Greenland (north-west Atlantic).

In autumn, the role of air masses coming from different sub-areas in formation of nucleation bursts was similar to that in spring. The autumn is also a transition period for the recombination of atmospheric pressure zones. However, in comparison with those in spring the reduced frequency of nucleation events in air masses coming from the Arctic and the North Atlantic may be related to the higher water content of these air masses. In spring, the Arctic and north regions of the Atlantic are covered by ice, whereas in autumn arctic and polar air masses pass over the water surface.

Ratio of event to nonevent trajectories

The ratio R of event to nonevent trajectories in each sub-area during the entire period and during different seasons follows the sectional distribution of the nucleation probability P (Table 1). However, this information gives us the proportion of

event to nonevent trajectories for each sub-area. Thus, the number of event trajectories originated over the north-west North Atlantic (sub-area 14) exceeded more than three times the number of nonevent trajectories originated in the same sub-area. The ratio of event to nonevent trajectories decreased in the south-east direction and reached a minimum in the area that covers central Russia.

The ratio of event to nonevent trajectories was highest in sub-area 14 during all the seasons, reaching its first maximum (15.50) in summer and its second maximum (8.5) in sub-areas 13 and 14 in spring (Table 2). Consequently, we can again emphasize the role of the North Atlantic and Arctic areas in the process of new particle formation in Hyytiälä.

Table 1. Number of event and nonevent trajectories, ratio *R* and probability *P* for each sub-area for the whole period.

Sub-area	Number of event trajectories	Number of nonevent trajectories	*R*	*P*
12	720	365	1.97	0.66
13	1018	365	2.79	0.74
14	434	141	3.08	0.75
22	922	984	0.94	0.48
23	3239	3039	1.07	0.52
24	701	264	2.66	0.73
32	322	732	0.44	0.31
33	1086	1807	0.60	0.38
34	509	417	1.22	0.55

Distribution of spatial sources of the particles

The spatial distribution of the origin of air masses with high concentrations of aerosol particles in specific size ranges differed greatly. High

Table 2. Number of event and nonevent trajectories, ratio *R* and probability *P* for each sub-area for all the seasons.

Winter					Spring				
Sub-area	Number of event trajectories	Number of nonevent trajectories	*R*	*P*	Sub-area	Number of event trajectories	Number of nonevent trajectories	*R*	*P*
12	49	149	0.33	0.25	12	324	48	6.75	0.87
13	99	173	0.57	0.36	13	424	50	8.48	0.89
14	66	79	0.84	0.46	14	169	20	8.45	0.89
22	56	356	0.16	0.14	22	393	134	2.93	0.75
23	252	1012	0.25	0.20	23	1256	527	2.38	0.70
24	82	113	0.73	0.42	24	271	38	7.13	0.88
32	18	247	0.07	0.07	32	110	147	0.75	0.43
33	68	556	0.12	0.11	33	361	353	1.02	0.51
34	49	146	0.34	0.25	34	187	65	2.88	0.74
Summer					**Autumn**				
Sub-area	Number of event trajectories	Number of nonevent trajectories	*R*	*P*	Sub-area	Number of event trajectories	Number of nonevent trajectories	*R*	*P*
12	131	86	1.52	0.60	12	216	82	2.63	0.72
13	217	39	5.56	0.85	13	278	103	2.70	0.73
14	93	6	15.50	0.94	14	106	36	2.94	0.75
22	255	204	1.25	0.56	22	218	290	0.75	0.43
23	970	634	1.53	0.60	23	761	866	0.88	0.47
24	199	54	3.69	0.79	24	149	59	2.53	0.72
32	63	72	0.88	0.47	32	131	266	0.49	0.33
33	389	370	1.05	0.51	33	268	528	0.51	0.34
34	170	96	1.77	0.64	34	103	110	0.94	0.48

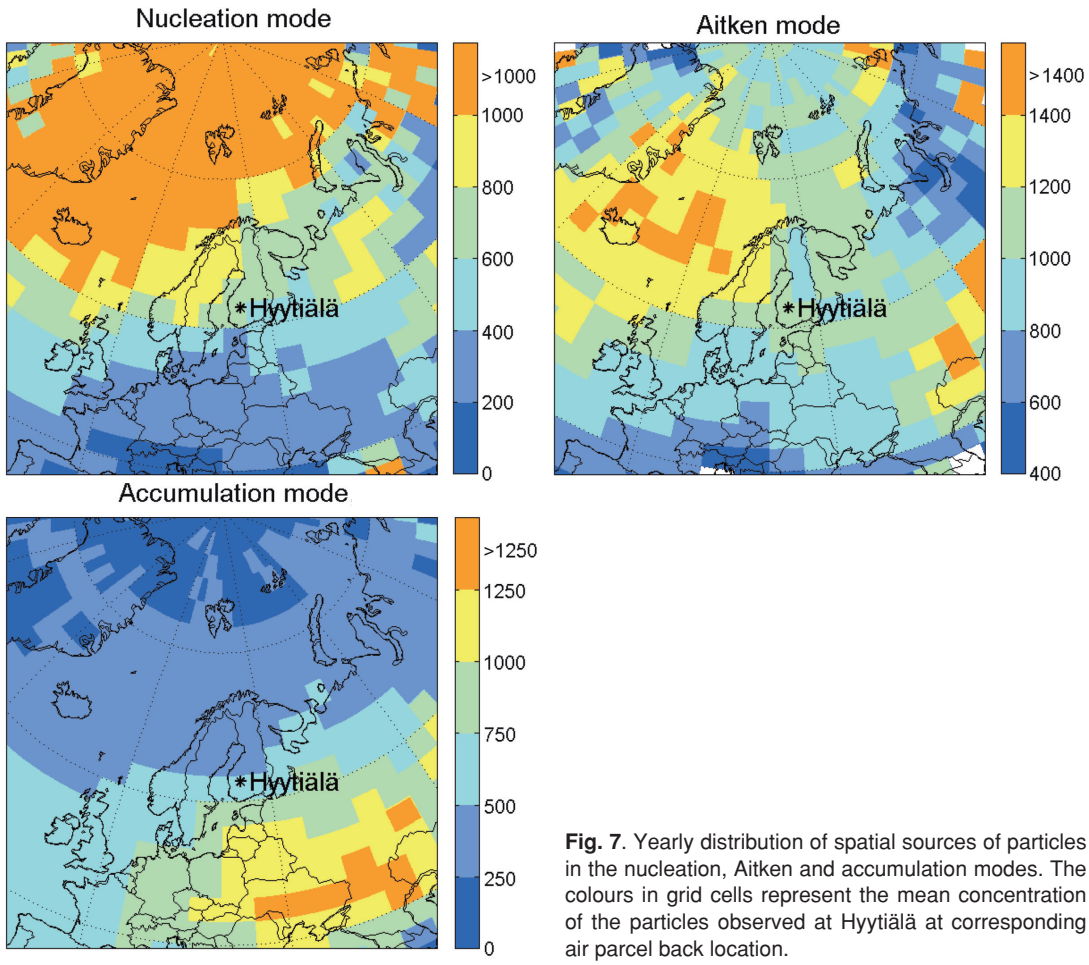

Fig. 7. Yearly distribution of spatial sources of particles in the nucleation, Aitken and accumulation modes. The colours in grid cells represent the mean concentration of the particles observed at Hyytiälä at corresponding air parcel back location.

(> 1000 cm^{-3}) concentrations of nucleation mode particles were observed in clean cold air when the predominant source regions for the air parcel arriving at the SMEAR II station were the Arctic and North Atlantic areas (Fig. 7), whereas low (< 200 cm^{-3}) concentrations corresponded to the south and south-west air flows. The location of sources for accumulation mode particles was clearly opposite. The concentrations of accumulation mode particles were typically highest when air masses originated from east to south from the experimental site. In these cases Hyytiälä was exposed to continental air, which is dry and usually warmer as compared with maritime air. Large concentrations of background aerosol particles in continental air masses may strongly decrease the number concentration of nucleation mode particles via removal processes (Korhonen *et al*. 2003, Kerminen *et al*. 2004). Condensation and coagulation sink can prevent the growth of newly formed particles to detectable sizes.

Contrary to the concentrations of the nucleation and accumulation mode particles, the concentrations of Aitken mode particles varied less with the upwind direction. On the other hand, the source areas seem to have higher geographical variation. Aitken mode particles have at least two evident source-regions: the North Atlantic which is also the source of nucleation mode particles, and Russia which is the main source of accumulation mode particles. From these results one can conclude that the north-western transport was associated with enhanced concentrations of particles with sizes in the lower end of the Aitken mode. These particles are formed by the nucleation and subsequent growth processes; however the enhanced concentrations in the Aitken mode can also be due to primary marine aerosol pro-

duction (Mårtenson *et al*. 2003, Geever *et al*. 2005). The North Atlantic source of aerosol particles might be originated by both primary and secondary (gas-to-particle conversion) aerosols. Gas-to-particle conversion via nucleation (homogeneous and heterogeneous) and condensation can also provide additional mass, and consequently, increase the size of the existing aerosol particles (O'Dowd *et al*. 1997, 2004) from nucleation to Aitken mode. Particles with sizes closer to the upper end of the Aitken mode are actually from the lower end of accumulation mode, and this can also be seen from the fact that they are most likely arrived from continental polluted regions, which are also the sources of accumulation mode particles.

The spatial distribution of sources of particles in different modes in all seasons reproduced relatively well the corresponding annual features (Fig. 8). For instance, the sources related to higher concentrations of nucleation mode particles were mainly located to the north and north-west of the recipient in all season. Sources for accumulation mode particles were located in the south and south-east in all the seasons. The behavior of sources for the Aitken mode particles was similar as well in all the seasons. However, the absolute values of particle concentrations in the different modes had a clear seasonal distribution.

Summary and conclusions

In this study, we considered the probability of new particle formation in different types of air masses arrived at Hyytiälä, southern Finland, and the spatial distribution of the sources of particles of different size ranges.

Prevailing air masses transport directions for Hyytiälä, southern Finland, are between south-west and north-west, whereas north and north-east transport is of secondary importance. Seasonal differences in prevailing flow directions exist but they are minor. In spring the frequency of south-west to north-west directions increases. In summer western flows prevail. In autumn the frequency distribution of flow directions is smoother as compared with that in other seasons. Since the horizontal pressure gradient is flatter in summer, the speed of air flows is lower and air masses overpass shorter distances. The speed of an air parcel is lower during that period and the influence of the local sources increases.

The dominant role of arctic and polar air masses causing new-particle formation is clearly seen during all the seasons. Nucleation events occurred more often in clean marine air originating from the North Atlantic and Arctic areas. In typically more polluted continental air masses, new-particle formation was very rarely observed. More explicitly, the role of arctic and polar air masses in the nucleation processes was expressed in spring when the frequency of new particle formation reached its maximum. The results allow us to conclude that a lower temperature, low amount of anthropogenic pollution, and in some cases lower humidity, are the necessary factors associated with new-particle formation events.

Air masses with high concentration of Aitken and accumulation mode particles present a high condensation and coagulation sink, decreasing the lifetime of vapours participating in the new particle formation process and removing newly formed ultrafine particles (Kulmala *et al*. 2001a, 2005). This may be a key factor in the observed tendency of northerly, cleaner air masses producing new particle formation events.

Clear differences in sources of the particles of different sizes were observed. In all the seasons the Arctic and North Atlantic areas were the sources of nucleation mode particles. The influence of that region on new-particle formation reached a maximum in spring when highest concentrations for all the modes are observed in Hyytiälä. Being an industrial region, the south-west of Russia and central Europe were the main sources of the accumulation mode particles during all the seasons. The formation of the accumulation mode particles over those regions may be connected with high concentrations of anthropogenic combustion products, including those from fossil fuel combustion.

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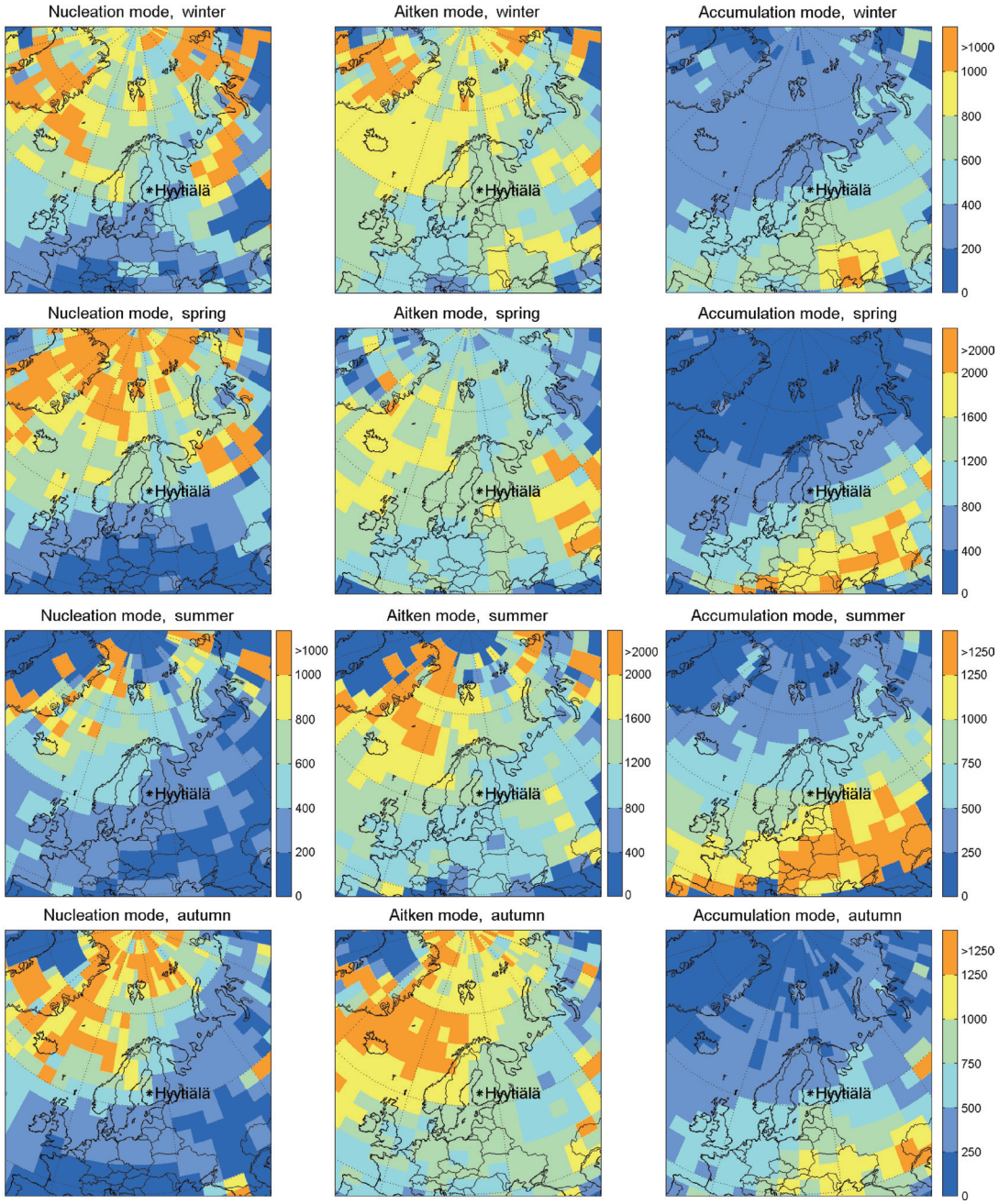

Fig. 8. Seasonal distribution of spatial sources of particles in nucleation, Aitken and accumulation modes. The colours in grid cells represent the mean concentration of the particles observed at Hyytiälä at corresponding air parcel back location.

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