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TELLUS

Atmospheric new particle formation at Utö, Baltic Sea 2003–2005

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ABSTRACT

Nearly 3 yr (March 2003–December 2005) of continuous particle number size distribution measurements have been conducted at the island of Utö in the Baltic Sea. The measured particle size range was from 7 to 530 nm. During the measurement period, a total of 103 regional new-particle formation events were observed. The characteristics of the nucleation events at Utö were similar to those reported in the literature in other Nordic sites, though measured condensation sinks were rather high (geometric mean of 3.8×10^{-3} s^{-1}) during event days. Clear evidence was found that new particles nucleate regionally near Utö, rather than are transported from greater distances. However, the Baltic Sea seems to have an inhibiting effect on new-particle formation. The boreal forest areas in the continental Finland were found to have an enhancing effect on the nucleation probability in Utö, suggesting that at least some of the precursor gases for nucleation and/or condensational growth of particles originate from these forests. In addition to regional new-particle formation events, a total of 94 local events were observed in Utö. These are short-lived events with a small footprint area, and can at least partly be tracked down to the emissions of ship traffic operating at Utö.

1. Introduction

Formation of new aerosol particles by nucleation and subsequent growth has been observed to take place in almost all atmospheric compartments ranging from clean polar areas to heavily polluted urban environments (see Kulmala et al., 2004, and references therein). Such aerosol formation events are likely to give a significant contribution to the global budget of the total particle number concentration (Spracklen et al., 2006). Regionally, atmospheric aerosol formation has been shown to enhance cloud condensation nuclei concentrations and to be involved in the cloud droplet activation process (Lihavainen et al., 2003; Kerminen et al., 2005; Laaksonen et al., 2005).

The climatic and other effects of aerosol particles formed in the atmosphere are tied closely to the frequency and overall magnitude of aerosol formation events in the system concerned. Boreal forests constitute probably the most widely studied aerosol system with regard to atmospheric aerosol formation (e.g. Kulmala et al., 2001a; Komppula et al., 2003a; Vehkamäki et al., 2004; Sogacheva et al., 2005; Tunved et al., 2006; Dal Maso et al., 2007; Kristensson et al., 2008; Svenningsson et al., 2008). However, all of the studies conducted so far have been based on measurements inside the boreal forest zone itself. Practically nothing is known about how boreal forest emissions influence aerosol formation downwind of the forest zone where fresh terpene emissions are lacking.

In this manuscript, we will analyse aerosol formations events taking place at Utö, a small island in the Baltic Sea tens of kilometres away from the boreal forest zone. In addition to being downwind of the boreal forest zone, the site offers a good possibility to investigate the potential influence of other emission types on atmospheric aerosol formation, such as aged industrial emission from central and eastern Europe, marine emissions from the Baltic sea, and ship emissions. Our analysis is based on almost 3 yr of measurement data recorded between March 2003 and December 2005. From the analysis, event day distribution will be obtained, and event characteristics will be presented from the event days. An air mass back trajectory analysis will be made in order to identify the air masses favourable to aerosol formation and to estimate the role of the Baltic Sea on new-particle formation in Utö. A comparison with similar measurements in the other Nordic stations will also be performed.

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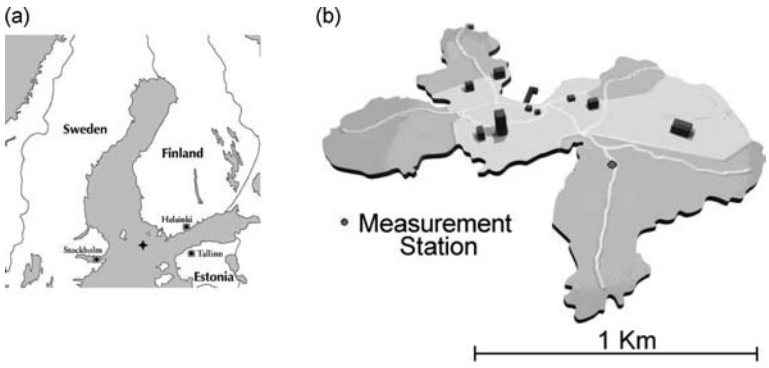

Fig. 1. Left-hand figure: Utö is situated at the Baltic Sea, south west of the Finnish coast. Right-hand figure: Location of the measurement station in Utö.

2. Methods

2.1. Measurement site

The location of the measurement site has been described in detail by Engler et al. (2007), so only a short overview is given here. Measurements were conducted at the EMEP European Monitoring and Evaluation Programme-station Utö (59°47′N, 21°23′E), located in the Baltic Sea (Fig. 1). The station is situated 8 m above the sea level some 60 km from the Finnish southwest coast. The closest city, Turku, is situated about 90 km to the northeast of the island. The wind direction is dominantly southwesterly in fall and winter and north or southwesterly during spring and summer times. The wind speed is highest in winter. Local pollutant sources include passing ships and tractor activities close to the station. These sources can be identified easily from the measurement data, since they produce exceptionally high, yet short-lived, particle concentrations in certain size classes.

2.2. Instrumentation

Several different measurements are conducted continuously at the Utö EMEP station. The measurements include basic meteorological observations, irradiance measurements, and measurements of trace gases, volatile organic compounds (VOC's) and aerosols. In particular, particle size distributions have been measured continuously since March 2003 using a Differential Mobility Particle Sizer (DMPS) system, described in detail by Engler et al. (2007). The measurements cover a size range of 7–530 nm which is divided into 30 discrete size bins.

2.3. Trajectory analysis

Trajectory analysis is a useful method of determining source regions of atmospheric aerosols and different pollutants (e.g. Birmili et al., 2001; Sciare et al., 2003; Sogacheva et al., 2005; Tunved et al., 2005; Kristensson et al., 2008). In this work, trajectory analysis was used to identify the air masses leading to nucleation at Utö. Five-day back trajectories were calculated with the FLEXTRA-model (Stohl et al., 1995), based on the wind fields available from the European Center for Medium-Range Weather Forecasts (ECMWF). The trajectories were calculated eight times per day (arrival times of 00, 03, 06, 09, 12, 15, 18 and 21 UTC at the station) for the 950 hPa level. The arrival time closest to the nucleation event starting time was used while evaluating the event time trajectories.

2.4. Classification of nucleation events

The new particle formation events, observed from the particle size distribution measurements, were classified by applying the methodology described by Dal Maso et al. (2005). In this classification, measurement days are divided into particle formation event days, non-events days and undefined days. Event days are those during which a new particle mode appears below 25 nm and subsequently grows into larger sizes. Depending on their intensity, the event days are further divided between the 'Class I' (clear event) and 'Class II' (fluctuation in number concentration). Undefined days are those from which it is impossible to state whether they are event or non-event days. In general, event days are indicative of so-called regional nucleation events when new-particle formation is taking place over scales of tens to hundreds of kilometres. During some of the undefined days, so-called 'local' particle formation events were observed in this study. These local events were short-lived with no clear particle growth. Such events are expected to takes place in plumes having a relatively small footprint area.

3. Regional nucleation events

3.1. General character of the nucleation events

The frequency of nucleation event days in Utö is presented in Fig. 2. It can be seen that new-particle formation was most probable during the spring and autumn and quite variable between the different summer months. A clear minimum in the event frequency could be seen during the winter. This kind of seasonal pattern reminds those observed at many other Nordic sites (Mäkelä et al., 2000; Komppula et al., 2003a,b; Vehkamäki et al., 2004; Dal Maso et al., 2007; Kristensson et al., 2008). The total number of event, non-event and undefined days are presented in

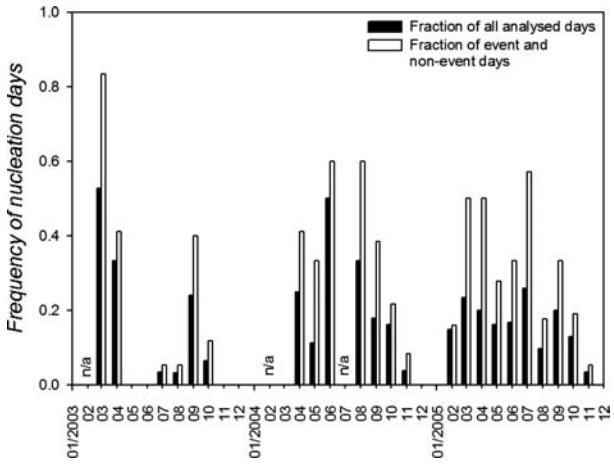

Fig. 2. The frequency of monthly Class I and II new particle formation events at Utö between 2003 and 2005. Black bars: fractions with all analysed days taken into account. White bars: fractions with only event and non-event days taken into account (no undefined days). In the *x*-axis, 01 denotes to January, 02 to February, etc.

Table 1. Number of event, non-event and undefined days at Utö between 2003 and 2005. Local event numbers are presented in brackets

Year	Event	Non-event	Undefined [Local]	Discarded
2003	27 (10.9%)	130 (52.4%)	91 (36.7%) [25]	117
2004	27 (12.6%)	97 (45.3%)	90 (42.1%) [20]	152
2005	49 (14.0%)	160 (45.8%)	140 (40.1%) [49]	16

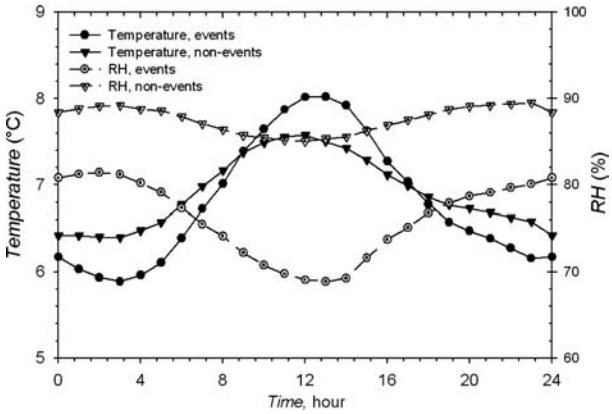

Fig. 3. Mean hourly temperature and relative humidity on event and non-event days.

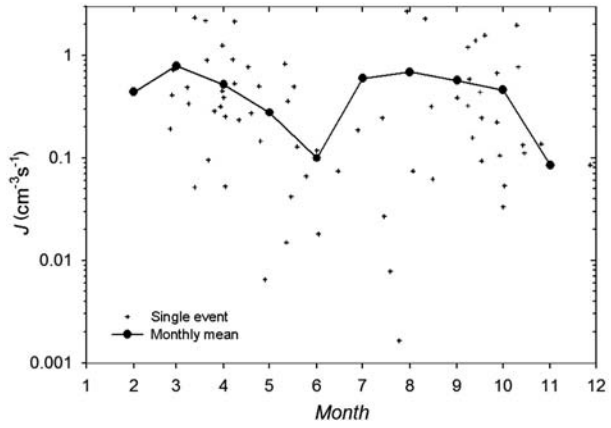

Fig. 4. Formation rates, *J*, of new 7 nm particles at Utö.

Table 1. Although the fraction of yearly event days seems to have increased since 2003, a longer set of continuous measurements is needed to draw any conclusions on trends. The event frequency is similar to those measured at other Nordic stations, with the exception that July 2005 was exceptionally rich in event days in Utö, showing no typical July minimum. The starting times of the events were also analysed, the average starting time being around 13:00. There were practically no events starting outside the daylight hours.

The diurnal variation of temperature, relative humidity and trace gas concentrations were compared between the event and non-event days. The event day mornings were, on average, about 0.2 °C colder than the non-event day mornings, but the highest temperatures during the event days were about 0.45 °C higher than those during the non-event days (Fig. 3). The high temperature difference between the morning and noon is typical for clear sky conditions. This is also backed up by the relative humidity data. On average, the relative humidity was about 16% lower on event-day noons compared with non-event-day noons. This is likely to be caused by dryer air masses during new-particle formation. In Värriö, the corresponding relative humidity difference was 25%, as air masses in Lapland can be very dry (Vehkamäki et al., 2004).

The particle formation and growth rates of the Class I nucleation events were analysed using method by Dal Maso et al. (2005). Formation rates ($J_{7\mathrm{nm}}$) were calculated from the observed formation rates $\mathrm{d}N/\mathrm{d}t_{7\mathrm{nm}}$ at 7 nm with coagulation loss rate taken into account. Class II nucleation events were not included because fluctuations in the growing nucleation mode interfere with the analysis, yielding often ambiguous results. The average particle formation rate observed in Utö was 0.45 cm^{-3} s^{-1} with a standard deviation of 0.61 cm^{-3} s^{-1}. A dip to lower rates was observed in June and in winter time (Fig. 4). The average particle formation rates were very similar to those found in Aspvreten (0.4), lower than those found in Hyytiälä (0.8) and Vavihill (1.54) and higher than those found in Lapland (Värriö 0.2 and Pallas 0.1) (Dal Maso et al., 2007; Kristensson et al., 2008). It should be noted that the particle formation rates measured in Hyytiälä and Vavihill are not totally comparable with those in the other stations due to the considerably lower cut-off diameter (3 nm) of the measurement system in these two locations. On the average, 3300 particles cm^{-3} were formed during Class I nucleation

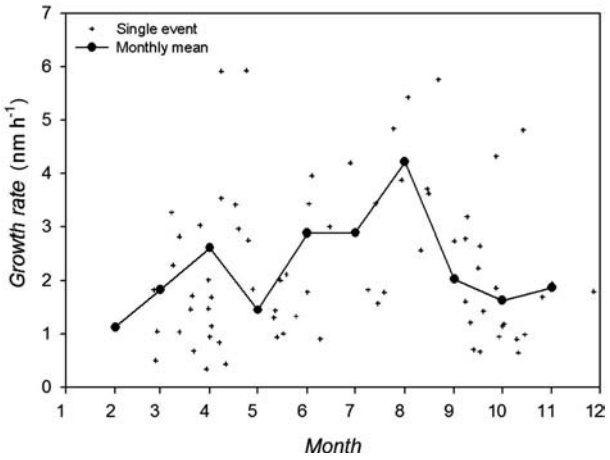

Fig. 5. Growth rates of nucleated particles at Utö.

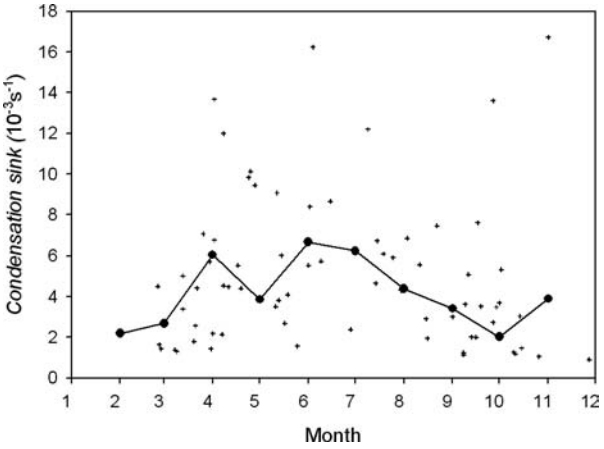

Fig. 6. Condensation sink during nucleation events at Utö.

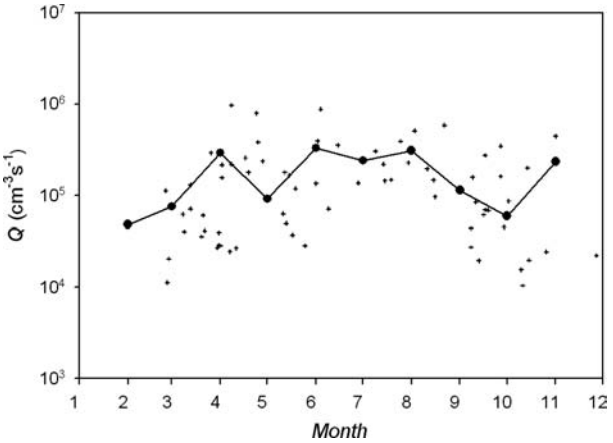

Fig. 7. Vapour production rate, Q, during nucleation events at Utö.

events. The mean increase in the total number of particles was about 120%, the maximum increase being as high as 1500%.

Growth rates of the nucleated 7 nm and larger particles are illustrated in Fig. 5. On average, the particle growth rate was 2.3 nm h^{-1} with a standard deviation of 1.0 nm h^{-1} and a clear peak value during August. The average growth rate was slightly lower in Utö than in other Nordic stations. The closest average growth rate is observed in Vavihill (2.5 nm h^{-1}; Kristensson et al., 2008). The annual behaviour of the growth rate differed slightly as well: in Utö there is no clear summer maximum in the growth rate as typically observed in the other Nordic stations. A more detailed analysis of vapours responsible for nuclei growth will be presented later in section 3.4.

3.2. Condensation sink and vapour source rate

The condensation sink (CS) is a measure of the rate at which vapour molecules are condensing from the gas phase into the pre-existing particle population. The value of CS has been found to be one of the most important parameters affecting the occurrence of new-particle formation (Hyvönen et al., 2005). A high surface area of the pre-existing particle population creates a large condensation sink which decreases the concentrations of vapours available for the growth of newly formed particles. The value of CS during the nucleation event days is presented in Fig. 6. The geometric mean of CS was about 3.8×10^{-3} s^{-1} with a standard deviation of 3.6×10^{-3} s^{-1}. This is substantially higher than those in other Nordic stations, except in Vavihill where the median value of CS was 3.5×10^{-3} s^{-1} (Kristensson et al., 2008). It seems apparent that the locations of Utö and Vavihill make these two stations perceptive to air masses from central Europe. Moreover, Utö is one of the furthermost Nordic stations for air masses originating from typically clean sectors (Atlantic and arctic). As a result, Utö is affected less by clean air masses than the other Nordic stations.

The vapour source rate was calculated from the vapour concentration needed to explain the observed particle growth, assuming a steady-state situation (Kulmala et al., 2001b). The vapour source rate is presented in Fig. 7. The mean vapour source rate was 1.8×10^5 cm^{-3} s^{-1} with a standard deviation of 1.6×10^5 cm^{-3} s^{-1} and a maximum during June–August. When compared against the Nordic stations, the source rate was closest to that in Aspvreten (2.2×10^5 cm^{-3} s^{-1}).

3.3. Relation to wind direction and air mass origin

The air mass origin in Utö was determined from back trajectory calculations. The trajectories were classified according to their azimuth angle at 24 h before arriving at the station. This classification method is similar to that presented by Dal Maso et al. (2007). Eight trajectories were calculated for each day, and the trajectory arriving closest to the starting time of a nucleation event was chosen. The trajectory directions for event and non-event times are presented in Fig. 8. When nucleation events were observed, about 21% of the trajectories arrived from the sector NNE. Sectors NNW and WWN were also important, with about 14% contribution from each. Only few nucleation events were

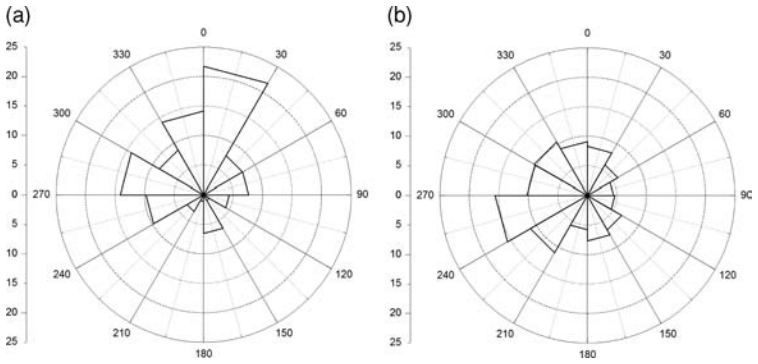

Fig. 8. Left-hand figure: regional nucleation event time trajectories. Right-hand figure: non-event time trajectories.

Table 2. Mean and median values of the nucleation rates classified according to the time the air masses have spent over the Baltic Sea prior to arrival at Utö

	Mean	Median ($cm^{-3}s^{-1}$)
≤2 h	0.778	0.400
>2 h	0.455	0.223
>5 h	0.409	0.146
>7 h	0.363	0.074
>10 h	0.093	0.066
All days	0.523	0.275

observed in southerly and easterly air masses. This pattern differs slightly from that observed in the other Nordic stations (Dal Maso et al., 2007), in which the most dominant air mass sector for nucleation events is NW–NNW. In Utö, the NNE sector points to the boreal forest areas in Finland. The WWS sector was the dominant one for non-event trajectories in Utö, with about 15% of non-event trajectories arriving from this direction.

Due to the marine location of Utö, measured air masses have spent some (varying) time over the Baltic Sea before reaching the site. The effects of this varying time on nucleation parameters were studied. On average, the nucleation rate was observed to decrease with increasing air mass transport time over the sea (see Table 2), even though no clear correlation between these two parameters could be drawn. This feature suggests that after a sufficient transport time over the sea, no nucleation events are expected to take place. The trajectory statistics give support to this finding. Of all trajectories, 32% had spent more than 10 h over the Baltic Sea before the arrival at Utö, while in only 19% of the nucleation days the air masses had spent more than 10 h over the Baltic Sea. The mean and median times of air masses time spent over sea were 9.7 and 7.0 h, respectively, for all the days and 8.0 and 5.5 h, respectively for the event days.

3.4. Vapours contributing to the particle growth

In order to get some insight on the vapours contributing to the growth of newly formed particles in Utö, sulphuric acid concentrations were estimated. The estimation was based on a pseudo-steady state situation and sulphuric acid closure presented by Boy et al. (2005) from measurements in Hyytiälä. Assuming a first-order reaction kinetics and condensation sink as the primary loss term for sulphuric acid, the following relationship can be derived:

$$[H_2SO_4] = \frac{k \times [OH] \times [SO_2]}{CS}, \quad (1)$$

where k is the reaction rate between the OH radical and SO_2. The concentration ratio for sulphuric acid between Utö and Hyytiälä follows directly from eq. (1):

$$\frac{[H_2SO_4]_{Utö}}{[H_2SO_4]_{Hyytiälä}} = \frac{[OH]_{Utö}\,[SO_2]_{Utö}}{[OH]_{Hyytiölö}\,[SO_2]_{Hyytiälä}} \times \frac{CS_{Hyytiälä}}{CS_{Utö}}. \quad (2)$$

Determining the OH concentration is a complicated process involving a large number of chemical reactions responsible for the production and loss of OH. The solar ultraviolet radiation intensity has been found to be the primary factor explaining the variability in the OH concentration (Rohrer and Berresheim, 2006). Since the average radiation intensity in Utö is probably very close to that in Hyytiälä, the OH concentrations were estimated to be roughly the same in these two locations. This approximation may cause some underestimation of the sulphuric acid concentration in Utö compared with Hyytiälä, since concentrations of volatile organic compounds (an effective loss term for OH) are very likely higher in Hyytiälä due to a stronger biogenic activity there. The sulphuric acid concentration ratios, presented in Fig. 9, were calculated from the monthly average values of $[SO_2]$ and CS for both stations. The resulting ratios ranged from 0.37 to 2.15, depending mainly on the levels of the SO_2 concentration. The average ratio was 1.12, which suggests that annually sulphuric acid has a slightly more important role in the particle growth in Utö compared with Hyytiälä. Boy et al. (2005) estimated that the contribution from sulphuric acid to particle growth was about 8.8% for March–April, 2003, in Hyytiälä. Based on monthly average condensation sinks, particle growth rates and sulphuric acid ratios, the contribution of sulphuric acid to growth rate in March–April would be about 5.5% in Utö (Fig. 9). We may conclude that similar to many other Nordic stations organic compounds are likely to play an important role in the growth of newly formed particles in Utö. The source of organics is most

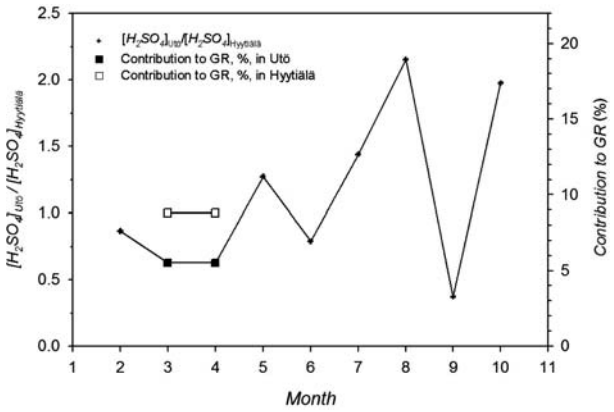

Fig. 9. Calculated ratio of H_2SO_4 concentrations at Utö and Hyytiälä, and H_2SO_4 contribution to particle growth in Hyytiälä (Boy et al. 2005) and Utö.

probably from boreal forest areas in Nordic countries, as suggested by the high fraction of nucleation events associated with trajectories from the direction of the Finnish forest areas.

4. Local events

A total of 94 undefined cases in the 3-yr period presented here were deemed as local nucleation events: short-lived events with no clear growth indicating plume like behaviour with a small footprint area. The yearly number of local event days is presented in Table 2 and an example of such an event is presented in Fig. 10. The clearest influencing parameter associated with these events was the wind direction (Fig. 11). The typical wind direction during local events was around 330°, which is where the ship traffic to and from Utö solely takes place. This differs dramatically from regional nucleation events for which no dominant wind direction could be identified.

Although the exact times of all ships trafficking at Utö cannot be checked, there is a ship connection to Utö that has a set timetable (Finnish Maritime administration, http://www.fma.fi/e/). This timetable was correlated with winds coming from the 300–360° sector. A total of 79 cases were found during the operational time of the DMPS when the connection ship was leaving from Utö, and when the winds were from the above sector. During 71% of these conditions a local new-particle formation event, or a less intense signal not qualified as local event, was observed in the particle size distribution data. Fig. 10 shows an example of such a day, with the connection ship scheduled to leave Utö at 12:00 local time. If the local event were due to aerosol formation in sulphur dioxide rich ship plumes, temporarily elevated SO_2 concentrations should coincide with the local event times. However, this was visible in only 40% of the cases. The average of the maximum SO_2 concentration during the local events was equal to 3.2 μg m^{-3}, as compared with the overall average SO_2 concentration of 0.9 μg m^{-3} for the wind sector of 300–360°. According to these evidences, we suggest that the 'local

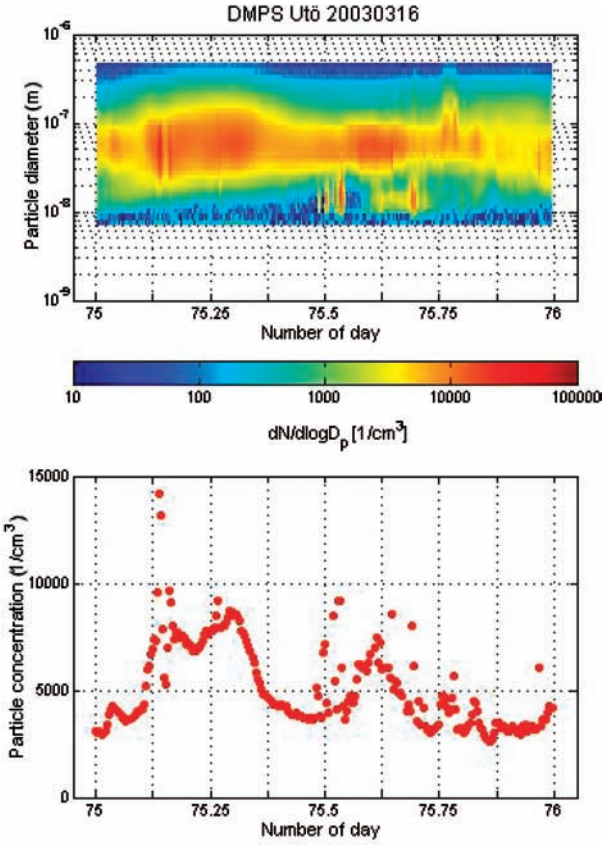

Fig. 10. Aerosol size distribution during a local nucleation event day. Local particle formation occurs around 12:00.

nucleation events' were at least partly related to emissions from diesel-powered ships operating at Utö. However, other mechanisms remain plausible.

The interest in ship emissions is tied usually to the so-called ship tracks which are regions of high cloud reflectivity associated with particulate emissions from ships (e.g. Devasthale et al., 2006). Practically all ship track studies have concentrated on primary soot particle emissions from ships and their further processing in the atmosphere by clouds and co-emitted pollutants such as sulphur dioxide (e.g. Russell et al., 1999; Durkee et al., 2000; Scheier et al., 2006). Very little emphasis has been put on aerosol formation taking place in ship plumes, even though indications of such a process can be seen from particle number size distribution measurements (Hobbs et al., 2000). The closest relatively well-characterized analogy to the observations made here comes from aerosol formation taking place in car exhaust plumes. Such a phenomenon seems to be very common for diesel-fuelled vehicles close to traffic lines and in urban environments (Kittelson et al., 2006; Rönkkö et al., 2006). Particles formed in car exhaust plumes grow usually very rapidly into the diameter range of 10–30 nm and, similar to our observations, appear frequently as if they were very small primary particles caused by some local source.

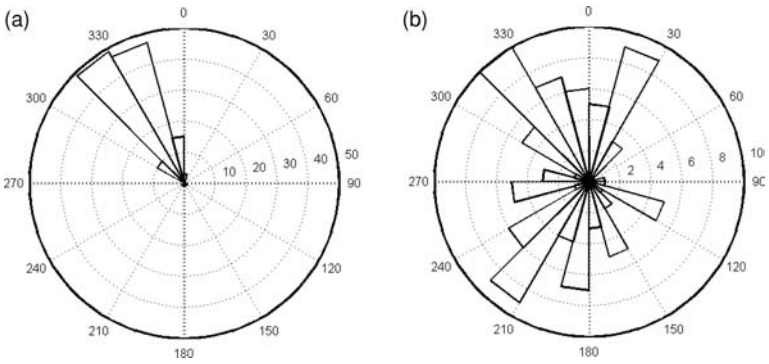

Fig. 11. Wind rose from local event times (left-hand figure) and from regional nucleation event times (right-hand figure).

5. Conclusions and discussion

Atmospheric nucleation events measured in Utö during 2003–2005 were studied in this work. The observed nucleation events could be divided into regional and local events according to their characteristics. The regional nucleation events seemed to be linked to the same phenomenon as Scandinavian inland-type events, that is, influenced strongly by natural emissions from boreal forest areas. Several pieces of evidence support this conclusion. First, the annual cycle of nucleation events looked very similar to that observed in the other Nordic stations in or near boreal forest areas. Second, trajectory analysis suggests that nucleation days occurred more often when trajectory path went over the boreal forest areas. Third, sulphuric acid was estimated to have less than 20% contribution to the nuclei growth, indicating a dominant contribution from organic compounds. Finally, nucleation rates were smaller for arriving air masses spent more time over the Baltic Sea, suggesting that the sea has an inhibiting role in triggering/maintaining new-particle formation. However, as also the very smallest particle sizes (<10 nm) could be observed at Utö, regional new-particle formation associated with boreal forest emissions seems to extend itself all the way to the Utö region.

The local nucleation events observed at Utö originated, at least partly, from local ship emissions. The wind direction during the local events was almost entirely from the direction of the entrance passage to the Utö pier, and under favourable wind conditions the local event starting times correlate with the departure times of the regular ship connection to Utö. Furthermore, the particle number size distributions during the local nucleation events were similar to those observed typically in car exhaust plumes near traffic lines and urban environments.

6. Acknowledgments

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