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Comparison of new particle formation events at two locations in northern Finland

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During 21 months (April 2000–December 2001) of continuous aerosol particle number size distribution measurements in a subarctic area in northern Finland over 90 new particle formation events were observed. Measurements were done at two stations separated from each other by 250 kilometres. The Pallas station is located in western Lapland and Värriö station in eastern Lapland. New particle formation events were compared between the stations. Trajectories, trace gas concentrations and various weather parameters were investigated to explain the reasons for observed differences. According to trajectory analyses, all events were associated with marine/polar air masses originating from the northern Atlantic or the Arctic Ocean. 55% of particle formation events were observed at the same day on both sites, while on 45% of the cases particle formation was observed only at one of the stations. Most of these differences were explained by air masses of different origin or rain/fog on the other station. Among events observed only at the Värriö site, experiencing air masses from Kola Peninsula industrial area, about 80% were associated with high SO_2 concentrations. At the Pallas site no increase in SO_2 concentrations was observed. This is an indication that in plumes originating from the Kola industrial area, new particles are formed by nucleation involving H_2SO_4 from SO_2 oxidation. The distance from Kola Peninsula to Pallas is long enough to reduce the SO_2 concentration level lower than needed to launch the nucleation process at Pallas. Also on some occasions the air masses arriving at Pallas had been reasonably higher up in the atmosphere as compared with air masses at the site closer to Kola Peninsula. Air masses at higher altitudes are not influenced so much by the Kola industry. Based on this work about one third of the nucleation events observed at Värriö are effected or caused by the Kola Peninsula industry.

Fig. 1. Location of the measurement sites, Pallas and Värriö.

Introduction

Atmospheric aerosol particles have large impacts on the global cloud albedo, radiative forcing, ozone layer, acid rain and visibility (Charlson *et al*. 1992, Seinfeld and Pandis 1998, Jacobson 2001). Aerosol particles influence climate both directly and indirectly. Directly, aerosol particles reflect solar radiation back to space. Indirectly, enhanced particle concentrations increase the number of cloud droplets by acting as cloud condensation nuclei, which increases cloud reflectivity and lifetimes. Houghton (2001) has reported that uncertainties in the estimation of direct and indirect aerosol effects on global climate are large. These uncertainties are greatly due to information on spatial and temporal distribution as well as optical properties of aerosol particles and clouds.

New particle formation is one of the main research issues in atmospheric sciences. Particle formation increases the total number concentration of ambient submicron particles and contributes thereby climate forcing. New particle formation events have been observed in several different environments, including in the marine boundary layer (Covert *et al*. 1992), free troposphere (Weber *et al*. 1999), coastal sites (O'Dowd *et al*. 1999), arctic boundary layer (Aalto *et al*. 1995), Arctic areas (Wiedensohler *et al*. 1996, Pirjola *et al*. 1998), Antarctica (O'Dowd *et al*. 1997) and boreal forests (Mäkelä *et al*. 1997). Long-term measurements have been made on a high alpine site (Weingartner *et al*. 1999), on a flat field close to Leipzig, Germany (Birmili and Wiedensohler 1997) and at boreal forest sites in southern (Mäkelä *et al*. 1997, Kulmala *et al*. 1998, Mäkelä *et al*. 2000) and northern Finland (Komppula *et al*. 2003). Bursts of charged nanometre particles have also been observed during measurements of air ions (Hõrrak *et al*. 1998).

Formation and growth mechanisms of aerosol particles are still not quantified. The geographical extent of particle formation events and the conditions affecting these events are not well known. In a subarctic area in northern Finland, there are two stations separated by 250 kilometres with similar kinds of instrumentation for aerosol particle measurements. This gives a good opportunity to investigate the geographical extent of particle formation events and the influence of atmospheric conditions on this phenomenon.

Experimental description

Site description

Both stations, Pallas and Värriö, are located in a subarctic region in northern Finland (Fig. 1). There are practically no local sources of pollutants close to the stations. The closest major pollutant sources are the smelters Nikel and Montshegorsk in Russia, located 200 kilometres to the north and 140 kilometres to the east of the Värriö station, respectively. Nikel is located directly to the north of Värriö and Monchegorsk to the east.

The main measuring site at Pallas GAW station, Sammaltunturi (67°58´N, 24°07´E, WMO index number 05821), lies on the top of a fjeld at the height of 560 m above the sea level and ca. 300 m above the surrounding area. The tree line lies some 100 m below the station. The station is within the Pallas–Ounastunturi National Park (total area 501 km^2) near the northern limit of the boreal forest zone. A more detailed description

of the area can be found in Aalto *et al.* (2002).

The Värriö environmental measurement station is located 250 kilometres away and almost directly to the east of Pallas (Hari *et al.* 1994, Ahonen *et al.* 1997). The station is near the Russian border (67°46´N, 29°35´E), 390 m above sea level. The station is located below the alpine timber line, which is about 400 m above sea level. The main tree species in the area is Scots pine. The station area belongs to the Värriö National Park.

Instrumentation

Similar DMPS (Differential Mobility Particle Sizer) systems are used at both measuring sites for nanometer particle sizing. DMPSs are built up with 28-cm-long Hauke-type differential mobility analyzer (DMA) (Winklmayr *et al.* 1991) with a closed loop sheath flow arrangement (Jokinen and Mäkelä 1997) and a condensation particle counter (CPC), TSI model 3010. The closed loop sheath air is dried with silica gel dryer and the relative humidity is kept below 20%. This sheath air setup keeps also the temperature more constant. Before sizing, the aerosol is neutralized with a radioactive source. The used sheath air flow rate is about 11 liters min^{-1} and sample aerosol flow rate is 1 liter min^{-1}. The measured particle size range is from 7 to 500 nm, which is divided into 30 discrete bins at Pallas and 20 bins at Värriö.

A number of trace gases are measured at both stations. In this study SO_2 and NO_X concentrations were found useful. At both stations measurements are continuous and one hour averages are calculated for further use.

Methods

DMPS data covered a period of 21 months, from April 2000 until the end of the year 2001. Various parameters describing the formation events, including trace gas concentrations and weather-related parameters were compared. Trajectories were calculated for every formation day for both sites (Hysplit 4 model 1997). This was done to get more information on the geographical extent of formation events and on parameters launching the events. In this study, we were focusing on those event days during which new particle formation was observed at one of the sites only, with less emphasis put on the days during which new particles were formed at both sites.

Definition and classification methods of a particle formation event

A new particle formation event can be seen as increasing particle concentrations in the smallest size channels of the DMPS system. An event can be observed from a surface plot (Fig. 2), in which the submicron particle size distribution is presented as a function of time (Mäkelä *et al.* 1997, Kulmala *et al.* 1998). On a typical particle-formation day, newly-formed particles enter the measurement range at around midday at initial sizes of 7–15 nm. During the rest of the day subsequent growth of these particles can be seen at a rate of a few nanometres per hour. Classifying between a particle formation event and non-event is sometimes difficult and always subjective. The classification method applied here has been explained in more detail by Mäkelä *et al.* (2000).

Particle formation events were observed from daily surface plots by visually searching for sudden increases in the number of smallest particles and their subsequent growth. After the event days were found, they were classified into three classes (1–3) according to the intensity of the particle formation and the distinctness of the particle growth. The best ones, showing a clear formation of small particles and their subsequent growth, were classified as class 1 events (as the event in Fig. 2). Class 2 events did not have such an intense new particle formation and/or growth as class 1 events. In class 3 events some new particles were formed, but no growth could be seen or the growth was weak. However, class 3 events still have some indications of particle formation. Also class 0 was introduced for unclear events. Defining the starting and ending times for an event is sometimes difficult due to fluctuations in the smallest size classes due to measurement uncertainties. Because of the subjectivity of the classification, some overlapping within the

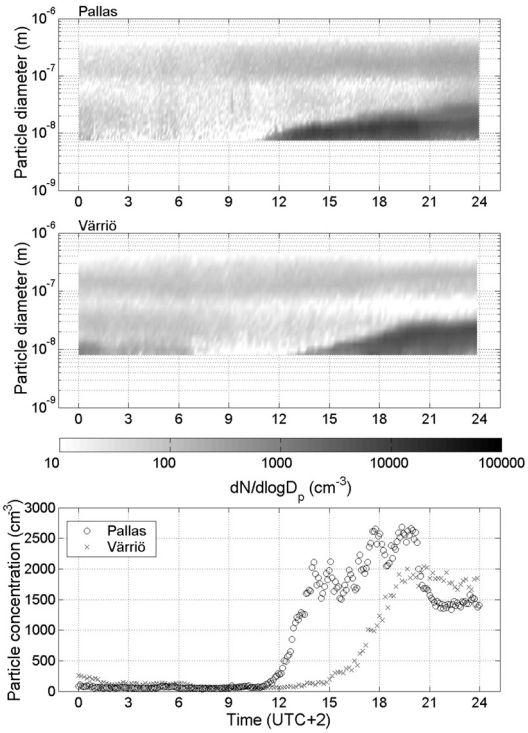

Fig. 2. A particle formation event observed at Pallas and at Värriö with a time difference of 2 hours, on 12 May 2000.

classes may occur. In the following analysis only class 1 and 2 events are considered to ensure the data quality.

Parameters describing the event

Some features associated with the events were estimated from the size distributions, such as the particle formation rate (particles cm^{-3} s^{-1}) and particle growth rate (nm h^{-1}). Particle formation days were mainly examined with a program made for this purpose at the University of Helsinki (Mäkelä *et al*. 2000). The program calculates the apparent formation rate of 7 nm particles and their growth rate. The starting and ending times of the events, as well as the upper diameter limit for the event mode (maximum new particle diameter) for every hour from the start of the event were manually determined from one-day spectra. The program calculates the particle formation rate by dividing the event mode concentration difference at the end and at the start with the length of the event. Furthermore, the number of new particles formed during the event, maximum new particle diameter after one to eight hours of growth and the total concentration at the start of the event were calculated.

The presence of existing particles affecting the condensation sink just before the formation event was also a point of interest. The condensation sink determines how rapidly molecules condense onto pre-existing aerosols and depends strongly on the shape of the particle size distribution (Pirjola *et al*. 1999). A detailed calculation procedure for the condensation sink has been presented by Kulmala *et al*. (2001). The condensation sink was calculated as a daily average for all days of the measuring period. On particle formation days a two-hour average just before the event was calculated. The condensable vapour concentration required for the observed growth during each event day was estimated. Also the source rate of condensable vapour required to sustain the concentration needed for the observed growth was estimated as presented by Kulmala *et al*. (2001).

Meteorological parameters and trace gases

Basic weather parameters were used to explain the differences found in particle formation. The temperature, pressure, relative humidity, wind speed and global radiation were observed at both stations. At Pallas there was a rain intensity sensor and at Värriö an on/off-type rain sensor. Visibility measurements at Pallas revealed the occasional presence of fog or low clouds (Komppula *et al*. 2003). NO_X and SO_2 concentrations were measured at both stations. One-hour averages of these gas concentrations were used in the analyses. 96-hour backward trajectories were calculated by Hysplit 4 at NOAA website (Hysplit 4 model 1997) to get the similarities of origin of air masses present at the particle formation time.

Results and discussion

In total, 87 classified event days were observed at Värriö and 71 event days at Pallas during the

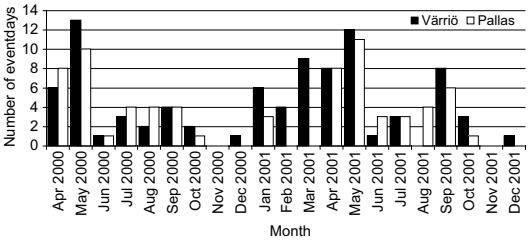

Fig. 3. Monthly distribution of particle formation events (classes 1–3) observed at Pallas and at Värriö from April 2000 to December 2001.

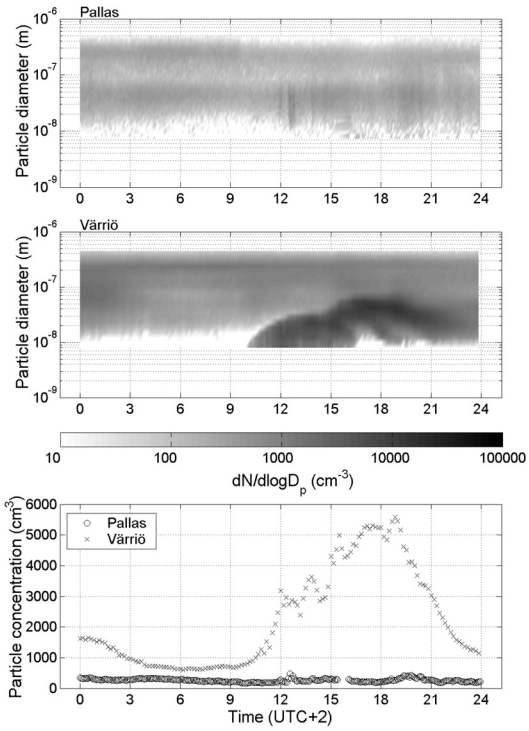

Fig. 4. A typical polluted new particle formation event observed only at Värriö, on 19 March 2001.

21-month measuring period (Fig. 3). The largest number of events was observed in April and May. Another peak was observed in late summer, in August and September. Among these, only the days that had class 1 or 2 event occurring at one of the stations and a clear event (classes 1–3) or non-event on the other, were taken into further inspection (41 days). By this selection method unclear class 0 events were left out of the analysis, to ensure the data quality. At Värriö 35 and at Pallas 29 event days filled these selection criteria. On 23 of these days new particle formation

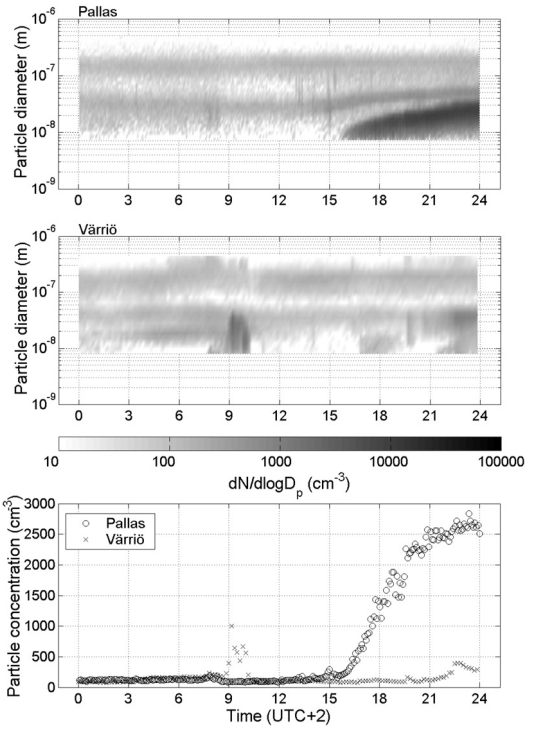

Fig. 5. A typical new particle formation event observed only at Pallas, on 5 September 2000.

was observed at both stations. On 12 days the event was observed only at Värriö (Fig. 4) and on six days only at Pallas (Fig. 5).

New particle formation at both sites on the same day

Table 1 provides a statistical summary on the values of some parameters related to nucleation events during those 23 days at which particle formation events were observed at both sites. The largest differences and variation are seen in the starting times of the events. The starting times varied between 9:20 and 18:00 (Fig. 6), while their difference between the two sites varied from 0 to 5.2 hours with an average of 1.6 hours. The average starting time was 12:10 (local time UTC+2) at Värriö and 12:50 at Pallas. This 40-minute difference compares somehow to the fact that at Värriö the sun rises 18 to 23 minutes earlier than at Pallas. The average length of the events was 6.1 hours at Värriö

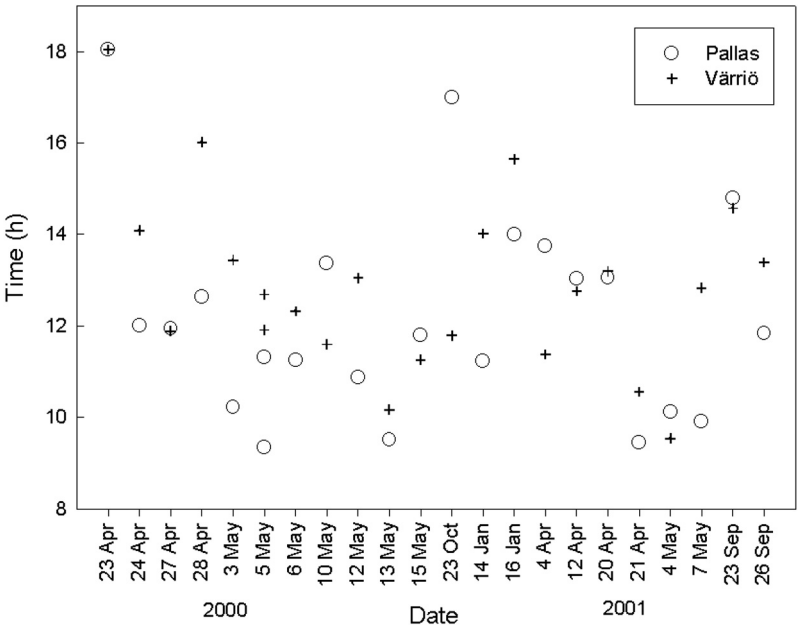

Fig. 6. Starting times of particle formation events observed at Pallas and Värriö during measuring period.

and 5.5 hours at Pallas. This may also be related to the earlier sunrise at Värriö. On average the number of pre-existing particles before the event was almost double at Värriö as compared with that at Pallas. This leads to higher condensation sinks (averages 8.2×10^{-4} s^{-1} at Värriö and 4.8×10^{-4} s^{-1} at Pallas) and higher vapour sources needed for a similar particle growth. The average formation rate of 7 nm particles (0.08 cm^{-3} s^{-1} at Värriö and 0.09 cm^{-3} s^{-1} at Pallas) and the number of new particles produced during the event (1750 cm^{-3} at Värriö and 1700 cm^{-3} at Pallas) were also very similar. The average growth rates were equal (2.9 nm h^{-1}) at both sites. Based on trajectories, the air masses on all event days arrived from the Arctic Ocean or

Table 1. Statistical parameters of events observed on the same day at Pallas and at Värriö.

	Starting time[a] (hh:mm)	Existing conc.[b] (cm^{-3})	Particles produced[c] (cm^{-3})	Formation rate[d] (cm^{-3} s^{-1})	Growth rate[e] (nm h^{-1})	Duration[f] (h)	Condensation sink[g] (10^{-4} s^{-1})	Vapour conc.[h] (10^{7} cm^{-3})	Vapour source rate[i] (10^{4} cm^{-3} s^{-1})
Pallas									
Min	9:20	90	130	0.01	1.1	2:30	1.6	1.5	0.3
Mean	12:10	440	1700	0.09	2.9	5:30	4.8	4.0	1.9
Max	18:00	1590	3840	0.29	8.2	8:30	19.8	11.5	7.7
Median	11:50	390	1300	0.06	2.4	5:20	4.0	3.4	1.4
S.D.	2:20	400	1180	0.07	1.7	1:40	3.8	2.3	1.7
Värriö									
Min	9:30	80	150	0.01	0.4	2:10	1.3	0.5	0.2
Mean	12:50	760	1750	0.08	2.9	6:10	8.2	4.0	3.4
Max	18:00	3350	4110	0.23	6.1	10:10	38.6	8.6	20.1
Median	12:50	490	1580	0.06	2.3	6:30	4.9	3.2	1.4
S.D.	2:00	800	1140	0.06	1.5	2:10	8.5	2.2	4.2

[a]Starting time of 7 nm particle formation; [b]Existing particle concentration at the time of start; [c]New particles produced during the event; [d]Average formation rate of 7 nm particles during the event; [e]Average growth rate of newly formed particles; [f]Duration of the formation event; [g]The condensation sink for vapour by existing particles; [h]Vapour concentration needed for the observed growth; [i]Vapour source rate needed to sustain the growth.

the northern Atlantic. One common pattern was the decrease of relative humidity and increase of temperature before the event. Also sunlight was needed to cause an event. Due to the low levels of SO_2, the observed particle growth rate could not be explained by sulphuric acid from SO_2 oxidation. Other vapours are clearly needed.

New particle formation observed only at Värriö

Reasons for the differences

On 12 days new particle formation was observed only at the Värriö site (*see* Fig. 4). Factors used to explain these events are presented in Table 2. Based on back trajectories, on three of these days the stations were under air masses of totally different origin. On two days the air masses were of the same origin but made a large loop to central Finland before reaching Pallas. On four days Pallas experienced a fog or low cloud cover, and on one day Pallas had hard rain just before noon. On one day the air masses were coming from the Atlantic Ocean, whereas the air masses at Pallas originated further away, closer to southern Greenland, and had also travelled at higher levels in the atmosphere. The air masses at Värriö had travelled closer to the sea level. On the same day Pallas experienced a smaller temperature rise and smaller relative humidity decrease than Värriö. On the last unexplained day, Värriö had higher concentrations of SO_2 and NO_X, indicating polluted air masses from the Kola Peninsula industrial area. High SO_2 concentrations were present on nine of the 12 days when particle formation was observed only at Värriö.

General observations

Overall, on 11 of the 12 days when a particle formation event was observed only at Värriö, the air masses were coming from the Arctic Ocean. On most of the days the air masses at Värriö had been travelling through the Kola Peninsula industrial area. On only one day the air masses were coming from the northern Atlantic. On 10 days Värriö experienced a larger relative humidity decrease than Pallas. Pallas was also affected by a fog or low-cloud cover on six of these days. Cloud droplets are an effective sink for small particles and gases. Pre-existing particle concentrations in Värriö were higher than or equal to those in Pallas in all cases. Also the condensation sink was higher at Värriö on all these days except on two low-cloud days at Pallas. On these days condensation sink at Värriö was only half of that at Pallas.

Table 2. The main explanations used for the events observed only at Värriö site. (X means that this explanation is used on that particular day.)

Date	Trajectories[a]	RH decrease[b]	T increase[c]	Rain/fog/clouds[d]	SO_2[e]	Condensation sink[f]
1 May 2000	X	X			X	X
8 May 2000		X	X			
2 Aug 2000		X		X		X
29 Oct 2000				X	X	
27 Feb 2001		X	X	X	X	
14 Mar 2001		X	X	X	X	
15 Mar 2001		X	X	X	X	
17 Mar 2001	X	X	X		X	
19 Mar 2001	X	X	X	X	X	
20 Mar 2001	X	X	X		X	
5 Jun 2001	X			X		
5 Oct 2001		X	X		X	

[a]The air masses have different origin; [b]RH decrease only at this station; [c]Temperature increase only at this station; [d]Rain/fog/clouds are preventing the formation on the other station; [e]Particle formation launched by polluted (SO_2) air masses; [f]Low condensation sink on this station, high on the other.

High SO_2 concentrations

On the 11 days when the air masses arriving at Värriö had travelled over the Kola Peninsula industrial area, the most obvious reason for new particle formation events was the high concentration of pollutants. On nine of these 11 days Värriö experienced high concentrations of SO_2, up to 70 $\mu g\ m^{-3}$, while Pallas had normally very low (< 2 $\mu g\ m^{-3}$) SO_2 concentration. Only on two days did Pallas have SO_2 concentrations close to 10 $\mu g\ m^{-3}$, which is still substantially lower than those at Värriö. High SO_2 concentrations observed at Värriö are an indication of a plume from the Kola Peninsula industrial area. On six of these nine days also Pallas was affected by air masses coming from the Kola area, but it seems that the distance from Kola to Pallas decreases the gas concentration lower than what is needed to launch a nucleation process at Pallas. Trajectories also showed that on four of the these nine days the air masses arriving at Pallas had been somewhat higher up in the atmosphere as compared with air masses arriving at the Värriö site. Air masses at higher levels might not be influenced so much by the Kola industry. This is one possible reason for lower SO_2 concentrations at Pallas on these days.

The events at Värriö are an indication of new sulphate particles formed in plumes originating from the Kola industry. Previous studies indicate that nucleation involving H_2SO_4 could be a viable mechanism in plumes containing high concentrations of SO_2. H_2SO_4 is also expected to participate in particle growth by condensation on existing particles (Mueller and Imhoff 1994, Kerminen and Wexler 1996, Pirjola *et al.* 1998).

New particle formation observed only at Pallas

Reasons for the differences

On six days new particle formation was observed only at the Pallas site. Factors used to explain these events are presented in Table 3. On three of the six days the stations were exposed to air masses of different origin, and on one day the air masses arriving at Pallas came straight from the north and made a large loop to central Finland before reaching Värriö. On the two cases left, the air masses came from the north, but travelled over the Kola Peninsula industrial area prior to arriving at Värriö. This polluted area may act as a sink for condensable gases formed above the sea. On the same two days the air masses arriving at Pallas had been somewhat higher up in the atmosphere as compared with the air masses arriving at the Värriö site. On one of the two days Pallas had a higher global radiation and larger relative humidity decrease before the event than Värriö. On the other day Pallas had a larger temperature increase and higher relative humidity decrease before the event than Värriö.

General observations

Some other observations were made out of the additional data on the six days that new particle formation was observed only at the Pallas site. One interesting thing to notice is that on four occasions of the six days the air masses arriving at Pallas had been reasonably higher up in

Table 3. The main explanations used for the events observed only at Pallas site. (X means that this explanation is used on that particular day).

Date	Trajectories[a]	RH decrease[b]	T increase[c]	Rain/fog/clouds[d]	SO_2[e]	Condensation sink[f]
26 Apr 2000	X	X		X		
25 Jul 2000		X		X		
11 Aug 2000	X	X				
12 Aug 2000	X	X	X	X		X
5 Sep 2000		X	X			
30 Jun 2001	X	X		X		X

[a]The air masses have different origin; [b]RH decrease only at this station; [c]Temperature increase only at this station; [d]Rain/fog/clouds are preventing the formation on the other station; [e]Particle formation launched by polluted (SO_2) air masses; [f]Low condensation sink on this station, high on the other.

the atmosphere as compared with air masses arriving at Värriö site. This could indicate less polluted air although the air masses had travelled nearby the Kola Peninsula industrial area. On three of the six days Värriö was experiencing rain or more cloud cover. On three of the six days Värriö did not experience a decrease of relative humidity and increase in temperature, which are the normal behaviours for an event day. Only on one day a slight increase in SO_2 concentration was seen and the concentration was higher at Pallas. This indicates that the pollutants from the Kola industry are not the factors launching these events at Pallas. All events occurred under clean polar air masses. Pre-existing particle concentrations at Pallas were only half of those at Värriö on two days. This can be also seen in the values of the condensation sink, which was 60% and 80% lower at Pallas on these two days. Otherwise pre-existing particle concentrations and condensation sinks were similar at the two sites.

Conclusions

During a 21-month period at two northern sites 250 kilometres apart from each other, a total of more than 90 nucleation events were observed. The most obvious events (41) were taken into a more detailed analysis. According to trajectory analysis, all the events were associated with marine/polar air masses originating from the northern Atlantic or the Arctic Ocean. On 18 days particle formation was observed only at one of the two stations. Most of these cases could be explained by air masses of different origin, or by the presence of rain/fog at the other station. During the events observed only at Värriö, high SO_2 concentrations (up to 70 μg m^{-3}) were measured in nine of the 11 cases when the air masses had travelled over the Kola Peninsula industrial area. At the same time, SO_2 concentrations at Pallas were much lower. It appears likely that these events were caused by nucleation involving H_2SO_4 from SO_2 oxidation in plumes originating from the Kola industry. On six of these nine days Pallas experienced also air masses from the Kola area, but it seems that the much longer distance from Kola to Pallas decreases the SO_2 concentrations lower than what is needed to launch the nucleation process at Pallas. On some days the air masses arriving at Pallas had been higher up in the atmosphere as compared with air masses arriving at the Värriö site. Air masses at higher altitudes might not be influenced as much by the Kola industry. This is one possible reason for the lower SO_2 concentrations at Pallas on those days. Based on this work about one third of the particle formation events observed at Värriö seem to be affected or caused by the Kola Peninsula industry.

The 23 particle formation events observed at both sites on the same day were all under the same air masses originating from the northern Atlantic or from the Arctic Ocean. The means and medians of the particle formation and growth rates were very similar at both sites. Overall these simultaneous events indicate that in clean and pollutant free air masses the geographical extent of a nucleation event is at least 250 kilometres which was the distance between Pallas and Värriö stations.

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