Observations of new particle formation and size distributions at two different heights and surroundings in subarctic area in northern Finland

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[1] Two years of continuous aerosol particle number size distribution measurements have been carried out in a subarctic area in northern Finland. Measurements have been made at two different heights (340 m and 560 m above sea level) and surroundings (inside a forest and at the top of a fjeld) to find out the effects of local-scale conditions on new particle formation and growth as well as on the size distribution of aerosol particles. Measured aerosol particle size range was 7–500 nm. Average total number concentration in the year 2001 was 700 cm^{-3} at the higher and 870 cm^{-3} at the lower site. One-day averages varied between 40 and 3500 cm^{-3}. Seasonal variation of total concentration was observed to be similar at both stations, high values in spring and summer and low values in winter. Also, modal concentrations were compared. Diurnal variation of total particle concentration was similar at both sites. In total, 65 new particle formation events were recorded during the measuring period. The largest number of events occurred in April and May. Particle formation events started between 0825 and 1550 (UTC plus 2 hours), and the calculated starting times of 1 nm particle formation (nucleation) varied between 0450 and 1420. The particle growth rate was found to be 1.4–8.2 nm h^{-1}, and the formation rate of 7 nm particles varied from 0.06 to 0.40 particles cm^{-3} s^{-1}. Starting times of the formation events at the two stations had maximum difference of 30 min, and it could not be explained by wind direction and speed. One possible explanation for the time difference is vertical movement of air masses caused by turbulence. Solar radiation was observed to be one key factor needed for particle formation. Wind direction was mostly between west and north on formation days, indicating polar or arctic air masses. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0325 Atmospheric Composition and Structure: Evolution of the atmosphere; 0399 Atmospheric Composition and Structure: General or miscellaneous; *KEYWORDS:* particle formation and growth, nucleation, atmospheric aerosol

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1. Introduction

[2] Atmospheric aerosols have large impact on climate forcing by effecting global cloud albedo, radiative forcing, ozone layer, acid rain and visibility [*Charlson et al.*, 1992; *Seinfeld and Pandis*, 1998; *Jacobson*, 2001]. Aerosol particles influence the climate directly by reflecting solar radiation back to space. Indirectly enhanced particle concentration increases the number of cloud droplets by acting as cloud condensation nuclei, which increases cloud reflectivity. *Houghton* [2001] has reported that uncertainties in the estimation of direct and indirect aerosol effects on global climate are large. Increasing number of aerosol particles may also cause significant health effects [*Dockery and Pope*, 1994; *Donaldson et al.*, 1998].

[3] New particle formation is one of the current issues in atmospheric science. Particle formation increases the total number concentration of ambient submicron size particles and thereby effecting human health and climate forcing. New particle formation has been observed in several studies in different surroundings. So far, studies have been made at least in the marine boundary layer [*Covert et al.*, 1992], in

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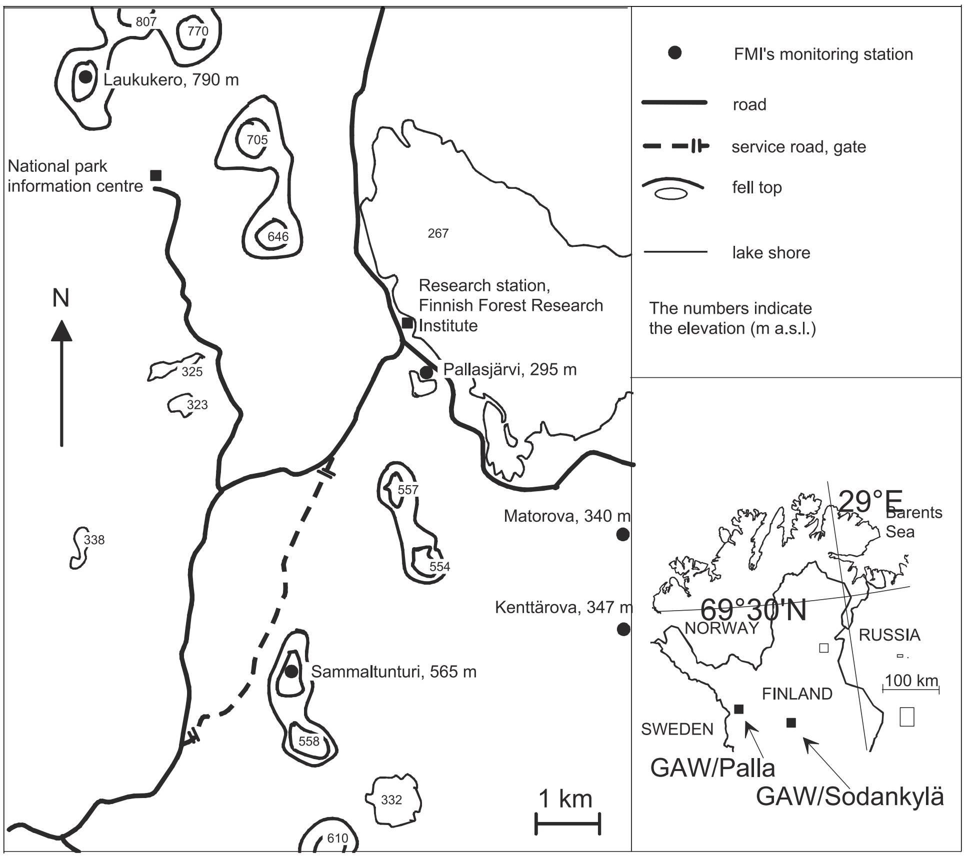

Figure 1. Location of the measurement site Pallas, Finland.

the free troposphere [*Weber et al.*, 1999], at coastal sites [*O'Dowd et al.*, 1999], in the arctic boundary layer [*Aalto et al.*, 1995], in the Arctic [*Pirjola et al.*, 1998; *Wiedensohler et al.*, 1996], in the Antarctic [*O'Dowd et al.*, 1997], and over 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 a boreal forest site in southern Finland [*Mäkelä et al.*, 1997; *Kulmala et al.*, 1998; *Mäkelä et al.*, 2000b]. Bursts of charged nanometer particles have also been observed during measurements of air ions [*Hõrrak et al.*, 1998].

[4] Formation and growth mechanisms of aerosol particles are still not finally resolved. Vertical profiles of particle concentrations and size distributions during new particle formation event are important factors to find out. By measuring these, more information is obtained about species and parameters influencing formation events. The geographical extent of particle formation event and influence of different surroundings are not well known. Therefore the Finnish Meteorological Institute (FMI) has started continuous particle size distribution measurements in northern Finland at two different heights and surroundings. Measurements were started within subarctic region at Pallas area (Figure 1) in April 2000.

2. Instrumentation and Methods

[5] The Finnish Meteorological Institute has installed two differential mobility particle sizer (DMPS) systems in two sites, Sammaltunturi and Matorova, in Pallas Global Atmosphere Watch (GAW) station area. The station is located in Pallas-Ounastunturi National Park in northern Finland. Continuous measurements were started on 13 April 2000. DMPS of Matorova station was under repair from June 2000 to August 2000. Analysed data presented in this study are from the start to 16 February 2002.

2.1. Site Description

[6] One of the main criteria for selecting the Pallas area as the site for these measurements was the absence of large local and regional pollution sources. The main station, Sammaltunturi (67°58′N, 24°07′E, World Meteorological Organization index number 05821), is one of the 22 GAW stations around the world. Sammaltunturi station lies on a top of a fjeld at the height of 560 m above sea level (asl) and ~300 m above the surrounding area. The tree line lies some 100 m below the station. Because of its elevation the Sammaltunturi station is from time to time within the cloud cover. The station is within the cloud cover for at least part of the day in about 10% of all days. The other station, Matorova (68°00′N, 24°14′E), is ~6 km northeast of Sammaltunturi at an elevation of 340 m asl. The station lies in forested area. Both stations have an automatic weather station, and in addition Sammaltunturi station has a present weather sensor (Vaisala FD12P). This provides a good opportunity to investigate the effects that various meteorological parameters have on aerosols. Stations are within the Pallas-Ounastunturi National Park (total area 501 km^2) near the northern limit of boreal

forest zone. A more detailed area description is presented by *Aalto et al.* [2002].

2.2. Instrumentation

[7] Similar DMPS systems are used at both measuring sites for nanometer particle sizing. DMPSs are built up with 28-cm-long Hauke-type differential mobility analyser (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. Before sizing aerosol is neutralized with a 370-MBq Nickel-63 beta source. Used sheath airflow rate is 11.0 L min^{-1}, and sample aerosol flow rate is 1.0 L min^{-1}. Aerosol sample at Sammaltunturi station is taken from a sampling line (1500 L min^{-1}), with inner diameter of 56 mm. The sample is taken from the line to the instruments with short 4-mm inner diameter tubing. At the Matorova station the sample is taken straight from the outside with short 4-mm inner diameter tubing. All tubes are made of acid resistant stainless steel.

[8] The measured particle size range is from 7 to 500 nm, which is divided into 30 discrete bins. DMPS systems are equipped with sheath air temperature, pressure, and relative humidity sensors. The sheath air is kept at relatively low and constant humidity (<20%) with silicagel dryer. One particle size spectrum takes around 5 1/2 min to measure (30 bins times 10 s plus decreasing the voltage from highest value to zero). The daily temperature and pressure means are used to improve the accuracy of result calculations. At Sammaltunturi station, FMI also has a condensation particle counter (TSI 3010) measuring total aerosol number concentration. It gives a good comparison and monitoring value for DMPSs.

[9] Before the measurements were started, the instruments were calibrated with silver particles at the University of Helsinki with a similar method presented by *Aalto et al.* [2001]. During measurements, DMPS data were monitored on a weekly basis and compared with each other and CPC values. At the end of the measuring period both DMPS systems were operated in parallel for 18 days at Sammaltunturi to check the instrument differences. The difference between the two DMPSs was reasonable, within ±10%. Data analysis was based on routines and programs developed at the University of Helsinki [*Aalto et al.*, 2001]. Particle losses in sampling lines and losses inside the DMAs were also taken into account.

2.3. Particle Concentrations and Size Distributions

[10] Total and modal particle number concentrations were calculated from the measured data. Fixed size limits used for different modes are used to get more comparable results for both measuring sites. Modal limits are set for the nucleation mode between 7 and 25 nm, for the Aitken mode between 25 and 95 nm, and for the accumulation mode between 95 and 500 nm. This three-modal structure and these size limits are close to the ones typically assumed for continental particle size distribution [see, e.g., *Mäkelä et al.*, 2000a]. Average daily total concentrations and monthly averages of modal concentrations were compared between the two sites. Also, diurnal variations of total and modal concentrations were compared.

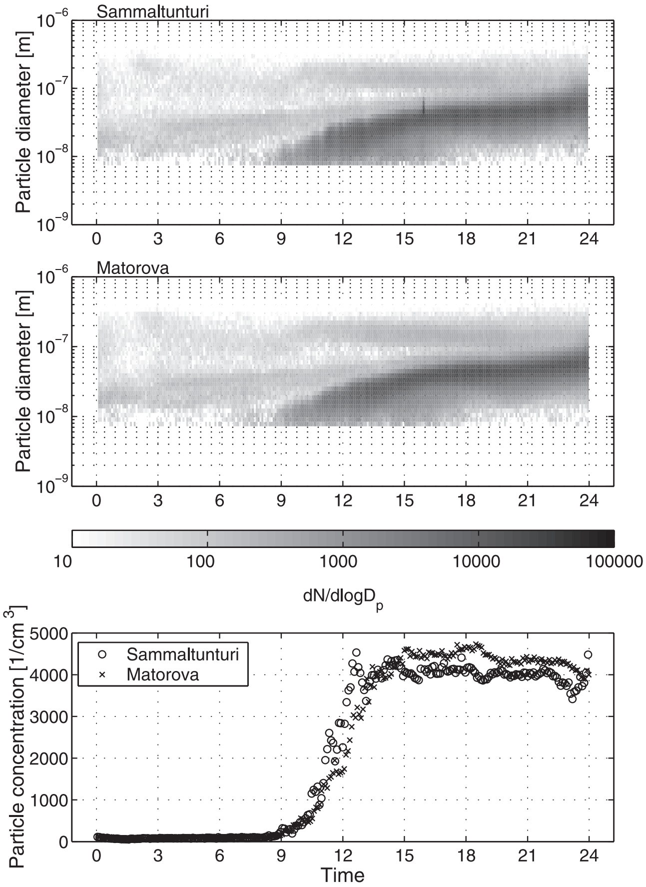

Figure 2. Particle formation event at Matorova and Sammaltunturi occurring at 0830 on 3 August 2001. A surface plot of 1-day size distributions at (top) Sammaltunturi and (middle) Matorova, and (bottom) the total particle concentrations during the day at both stations.

Aerosol particle number size distributions were compared on daily basis.

2.4. Definition and Classification Methods of a Particle Formation Event

[11] A new particle formation event can be seen as increasing particle concentrations occurring in the smallest size channels of the DMPS system. An event can be observed from a surface plot (Figure 2), in which the submicron particle size distribution is presented as a function of time [*Mäkelä et al.*, 1997; *Kulmala et al.*, 1998]. In Figure 2 the total particle concentration of the example day is shown. On a particle formation day the newly formed particles enter the measurement range at around midday with initial sizes of 7–15 nm. During rest of the day, subsequent growth of these particles can be seen with a growth rate of few nanometers per hour. Classification between a particle formation event and a nonevent is sometimes difficult to make and is always somewhat subjective. The classification method is explained in more detail by *Mäkelä et al.* [2000b]. Particle formation events were observed from daily surface plots by visually searching for sudden increases in number of smallest particle sizes and subsequent growth. After the event days were found, events were classified into three classes from 1 to 3 according to their intensity and the distinctness of growth.

The best ones, which showed clear formation of small particles and clear subsequent growth, were classified as class 1 events (as the event in Figure 2). Class 2 events do not have such intense formation of new particles or particle growth as class 1 events. In class 3 events some new particles are formed, but no growth can be seen, or the growth is weak. However, class 3 events still have some indications of particle formation. Defining starting and ending times of an event is sometimes difficult because of fluctuation in the smallest size classes due to measurement uncertainties. Because of the subjectivity of the classification some overlapping within the classes may occur. In the resulting analysis, only class 1 and 2 events are considered.

2.5. Particle Formation Rate and Growth Rate

[12] 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 the purpose at the University of Helsinki [*Mäkelä et al.*, 2000b]. This program calculates the apparent formation rate of 7 nm particles and the growth rate. Starting and ending times of the events and upper diameter limit for the event mode (maximum new particle diameter) for every hour from the start of the event were manually determined from the 1-day spectrum. The program calculates the 7-nm 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 1–8 hours of growth, and the total concentration at the start of the event are calculated by the program.

[13] It has been suggested that 1 nm is the size of a thermodynamically stable clusters, which under certain conditions grow to detectable sizes [*Kulmala et al.*, 2000]. For this reason, the approximate starting time of formation of 1-nm particles was calculated. This was made to estimate the starting time of the nucleation process and, for example, how close to the sunrise nucleation starts. Calculations were made backward in time with the estimated growth rate. This estimate should be reasonable because of the stable growth rates observed during the events.

2.6. Meteorological Measurements

[14] Meteorological data in particle formation days were closely investigated to find out dependencies between event days and meteorological parameters. Data were obtained from meteorological measurements from both measurement sites. Hourly averages in SO_2, NO, and NO_2 concentrations at Sammaltunturi station were also obtained. Starting and ending times of the events were compared to the time of sunrise and sunset. Earlier studies show that new particle formation days are closely related to higher solar radiation and temperature rise and are mainly occurring under polar or arctic air masses a few hours after the sunrise [*Mäkelä et al.*, 1997; *Kulmala et al.*, 1998; *Mäkelä et al.* 2000b]. Large downward particle fluxes, higher turbulence intensities, and strong vertical mixing have been observed during the nucleation days [*Buzorius et al.*, 2001]. Also, temperature and water content of the air was observed to be very low on average during the formation days. Turbulent sensible heat fluxes in the surface layer and turbulent kinetic energy in the lower mixed layer have been observed to be twice as high on particle formation days as on other days [*Nilsson et al.*, 2001]. The smallest detectable particles (3 nm) have been observed to occur within 10 min to 2 hours from the onset of strong turbulent energy in measurements in southern Finland [*Nilsson et al.*, 2001].

3. Results and Discussion

3.1. Total Particle Number Concentrations

[15] In Figure 3, daily concentration averages are shown from April 2000 to February 2002 at Sammaltunturi and at Matorova. Average particle concentrations measured with DMPSs in 2001 were 870 cm^{-3} at Matorova and 700 cm^{-3} at Sammaltunturi. A great deal of the difference is caused by the higher altitude of the Sammaltunturi station. It is from time to time within the cloud cover. On low cloud cover days, particles larger than ~80 nm are activated as cloud condensation nuclei, and they grow rapidly outside the detection limit, which lowers the number concentration at measured range. Pallas area has low particle concentration values compared to southern Finland because of the dominance of pollutant-free air. In southern Finland, at Hyytiälä station, yearly average particle concentrations (2000 cm^{-3}) are about twice as high compared to Pallas.

[16] Seasonal variation of particle concentration is clearly seen in Figure 3. High values are found in spring and summer, and lower values are found in winter. Large variation of daily average concentration is also seen. This variation is mainly due to particle formation events that increase number concentration considerably on event days. Day to day variation of Aitken mode concentration is not as significant. Seasonal variation in concentration is similar in both stations, as expected. The maximum 1-day average was 3480 cm^{-3} at Matorova and 3210 cm^{-3} at Sammaltunturi. The minimum 1-day averages were 90 cm^{-3} and 40 cm^{-3}, respectively. The maximum 1-hour average concentration 9290 cm^{-3} was found on 17 May 2000 at Matorova, and a minimum of 10 cm^{-3} was found in late October 2000 at Sammaltunturi. Generally speaking, during spring and summer the daily averages may rise up to 3500 cm^{-3}, and in winter the daily averages may drop below 100 cm^{-3}.

[17] The daily ratio of total particle concentration between Sammaltunturi and Matorova stations was around 0.9 on days when Sammaltunturi station was not inside the cloud cover. On days when Sammaltunturi station was inside the cloud cover, the ratio dropped under 0.5. Average concentration ratio was 0.8. The lowest ratio was 0.2, and the highest was 1.4. Sammaltunturi concentration exceeded Matorova only on few occasions.

[18] When comparing the results of the CPC at Sammaltunturi against the total number concentration obtained from DMPS, the average CPC values are a little higher (around +15%). Correlation between CPC and DMPS values over the measuring period is good, 0.986. The difference in average value is partly due to the particles >500 nm, which DMPS does not count. However, the accuracy in total concentration is reasonable for DMPS measurements.

3.2. Modal Particle Concentrations

[19] Modal concentrations were compared with fixed mode size limits as presented earlier. Monthly averaged

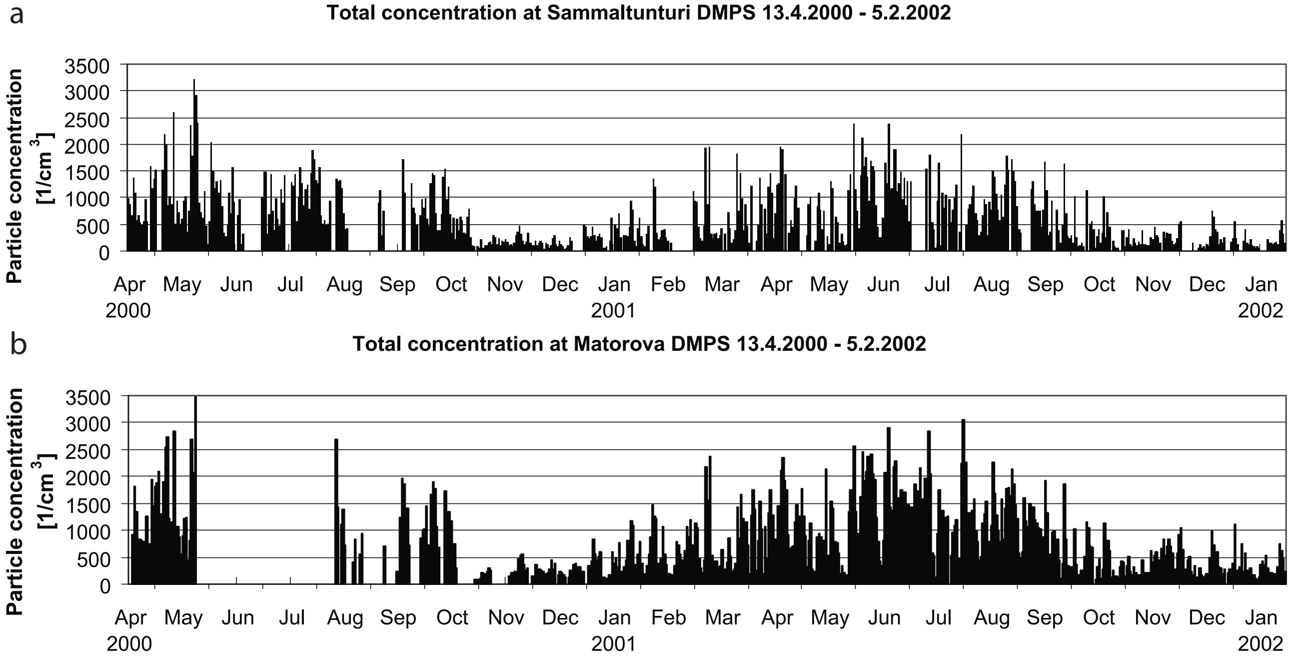

Figure 3. Daily particle number concentration averages from 13 April 2000 to 5 February 2002 at (a) Sammaltunturi and (b) Matorova.

modal values at both stations are presented in Figure 4. All modes have their minimum concentration during wintertime when total concentrations are low and no nucleation events occur. Nucleation mode has its highest concentrations during April and May when most of the new particle formation events occur. Another slight peak is observed in August and September. Nucleation mode average concentrations in 2001 are very similar at both stations, 130 cm^{-3} at Matorova and 150 cm^{-3} at Sammaltunturi. In Aitken mode the monthly concentrations are little higher at Matorova almost through the whole measuring period (Figure 4b). The average concentration in 2001 was 420 cm^{-3} at Matorova and 350 cm^{-3} at Sammaltunturi. In accumulation mode, the concentration difference between the stations is largest (Figure 4c). The average accumulation mode concentrations in year 2001 were 320 cm^{-3} at Matorova and 200 cm^{-3} at Sammaltunturi. The occasional low cloud cover at Sammaltunturi mostly decreases the accumulation mode and partially decreases Aitken mode concentrations. This reduction is seen through the whole year.

3.3. Diurnal Variation of Particle Number Concentrations

[20] Diurnal variation of particle concentration was calculated from half-hour averages over a 1-year period from May 2000 to April 2001. No difference between the stations was found. The diurnal variation of accumulation mode is the lowest of all modes. It has a minimum at around 0700 (all times are given in UTC plus 2 hours) and maximum around 1500 from spring to autumn. Aitken mode does not have very large diurnal variation either. It has a slight minimum around 1000 and a maximum around midnight in summer and autumn. During winter, no diurnal variation is seen.

[21] Most of the diurnal variation and its shape are caused by the particle formation events in spring and early summer. In nucleation mode the diurnal variation is the most obvious. This is due to particle formation always occurring within a few hours around noon. The variation is largest in April and May. Diurnal variation of nucleation mode in four different months is presented in Figure 5. Diurnal variation of nucleation mode concentration has its minimum around 0900 and maximum around 1800, as has the total concentration. High variation in spring (April and May) and low concentration and variation in winter (November) is clearly seen. The maximum particle concentration peak is getting earlier, when moving toward summer. This is probably due to earlier sunrise, meaning a larger amount of solar radiation. However, the time of the minimum value stays at around 0900. This might be an indication of higher nucleation rate and shorter duration of formation events or higher growth rate of newly formed particles. However, this cannot be verified with the small number (13) of class 1 and 2 events.

3.4. Particle Formation Events

[22] In total, 65 particle formation events were recorded during the period of 22 months. Formation events were not as frequent and not as intense as seen in the measurements in southern Finland. In Hyytiälä, over 50 events per year have been observed [*Mäkelä et al.*, 1997; *Kulmala et al.*, 1998; *Mäkelä et al.*, 2000b]. The low number and low intensity of events made the event classification somewhat difficult. Event days were divided into three classes. Nine event days were classified as class 1 events, 13 were classified as class 2 events, and 43 were classified as class 3 events. Furthermore, around 30 possible events were left out for being indistinct. In further analysis, only class 1 and 2 events are used because they are the most obvious ones. Because of missing data from Matorova (as seen in Figures 3a and 3b), there are 13 days of class 1 and 2 events to compare from both stations.

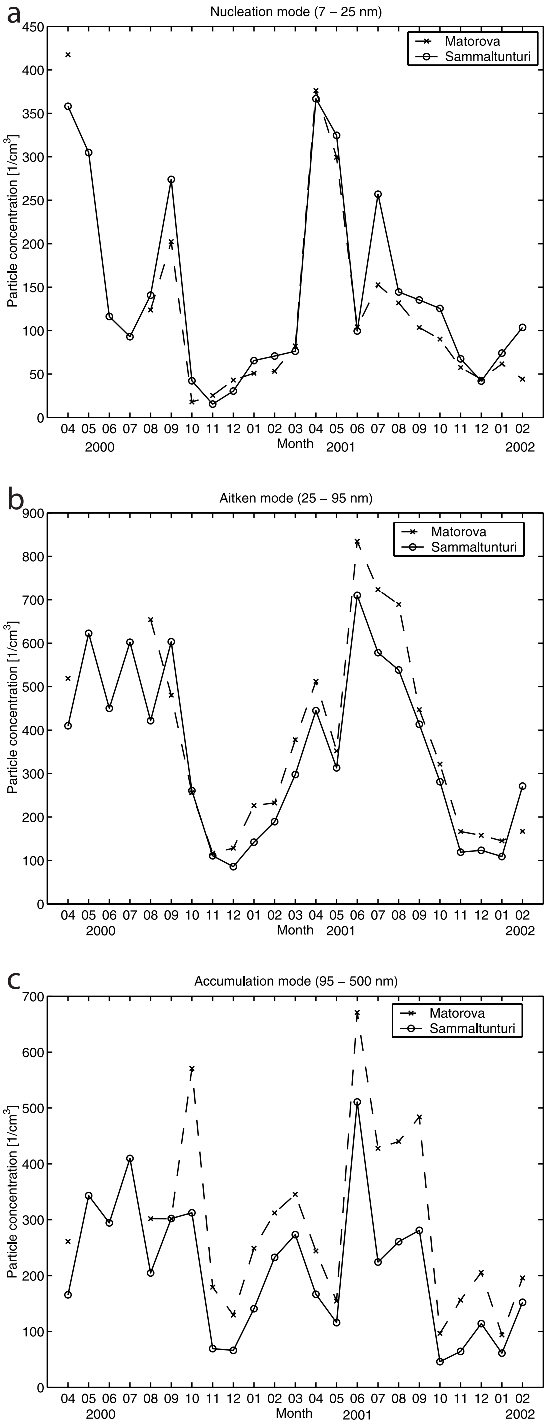

Figure 4. Monthly modal average particle concentrations at Sammaltunturi and Matorova from April 2000 to February 2002: (a) nucleation mode (7–25 nm), (b) Aitken mode (25–95 nm), and (c) accumulation mode (95–500 nm).

[23] In Figure 6 the fraction of days on which particle formation events of class 1, 2, or 3 occurred is presented. The fraction is calculated by dividing the monthly number of formation days by the number of days with data available in that particular month. The largest number of particle formation events occurs in April and May. Another slight peak is observed in August. In winter, practically no particle formation events were detected. As an exception, two class 3 events were observed in January 2001. The particle formation season from April to September is shorter than that observed in southern Finland, where events are already occurring in early March.

[24] All the events occurred nearly at the same time in both measuring sites. No event was found that would occur only at one of the sites. Various parameters related to particle formation events observed are presented in Table 1. Starting times for formation of 7-nm particles varied between 0825 and 1550, and calculated starting times of 1-nm particle formation varied between 0450 and 1420. Figure 7 shows starting times and the time differences of 7-nm particle formation events between the stations. With few exceptions, starting times seem to get earlier toward autumn. All events occur at both sites within 30 min of each other. The length of the events varied from ~3 hours to 9.5 hours. The average duration was ~5 hours.

[25] In calculations, particle growth was taken into consideration for maximum of 8 hours from the start of the formation event. On days when growth stopped before 8 hours, the growth rates were calculated until the point when the growth stopped. Growth times of individual days are presented in Table 1. Particle growth rate values varied between 1.4 and 8.2 nm h^{-1}. The average value for size that the particles reached during the event day was 50 nm. Formation rate of 7-nm particles varied between 0.06 and 0.40 particles cm^{-3} s^{-1}. The rate at Matorova was slightly higher compared to Sammaltunturi. The average number of new particles produced during the particle formation events was higher at Matorova (2700 cm^{-3}) than at Sammaltunturi (2360 cm^{-3}), though more particles were produced at Sammaltunturi in almost half of the events.

[26] The presence of existing particles, their surface area, particle volume, and aerosol condensation sink at the start of the formation event were also a point of interest (Table 1). The aerosol condensation sink determines how rapidly molecules condense onto preexisting aerosols and depends strongly on the shape of the size distribution [*Pirjola et al.*, 1999]. Detailed calculation procedure of condensation sink is presented by *Kulmala et al.* [2001]. Overall, these parameters were similar at both stations. Condensation sinks varied between 0.8 and 17.0 $\times$ 10^{-4} s^{-1}. The existing particle

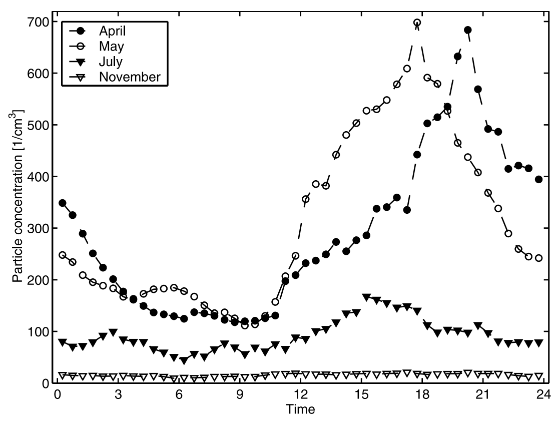

Figure 5. Diurnal variation of nucleation mode (7–25 nm) particle concentration at Sammaltunturi in April, May, July, and November, 2000.

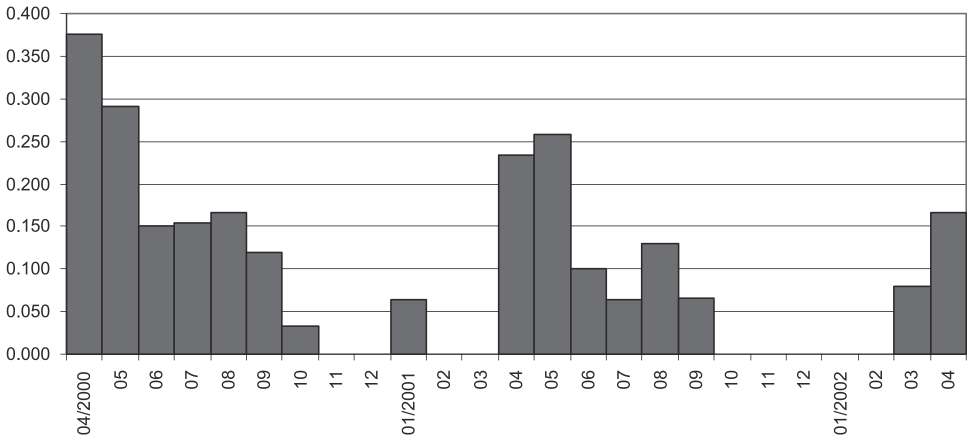

Figure 6. Monthly fraction of days on which particle formation occurred from April 2000 to April 2002.

concentration at the start of the events showed large variation (Table 1). Observed preexisting particle concentrations in event days were considerably below the monthly average of the month in question. Average surface area at the start of the event was 3.4 μm^2 cm^{-3} at Sammaltunturi and 3.8 μm^2 cm^{-3} at Matorova, and the average particle volumes were 0.19 μm^3 cm^{-3} and 0.21 μm^3 cm^{-3}, respectively.

3.5. Effect of Meteorological Parameters on New Particle Formation

[27] Formation events at Pallas occurred with a maximum of 8.5 hours after the sunrise. The start time of formation events is not dependent on the time of the sunrise, although solar radiation is clearly necessary. The minimum global solar radiation intensity on a formation day was 220 W m^{-2}, and the average was 450 W m^{-2} (Table 2). For comparison, the average solar radiation for the 2-year period was 80 W m^{-2}, and for the main particle formation season (1 April to 15 September) it was 160 W m^{-2}. In southern Finland, particle production was not observed when global solar radiation was below 400 W m^{-2} [*Aalto et al.*, 2001]. All solar radiation values on formation days were well over the average values. At Pallas area the stability of the air was investigated by calculating potential temperatures for both stations and the difference between them. On all particle formation days the potential temperature difference between Sammaltunturi and Matorova stations had large variations (maximum ±2°C). The difference was on average around 2°C. This indicates that the surrounding atmosphere was in an unstable condition, i.e., undergoing turbulent mixing and experiencing fluxes.

[28] Temperature on formation days was lower compared to other days. The 1-hour average temperature before formation events varied between −13.0°C and 13.5°C depending on the season. The mean temperature on the particle formation days was 2.3°C (Table 2). This was significantly lower than the average temperature for the whole formation season (1 April to 15 September), 5.8°C. The lower temperature on formation days indicates colder polar or arctic air masses from north. The wind directions on formation days support this. On formation days the wind was dominantly blowing between west and north. Wind direction distribution in percent at Sammaltunturi for the whole year 2001 and particle formation season and formation days are presented in Figure 8. Yearly wind direction is almost evenly distributed. Only winds between northwest and northeast are not so common. In particle formation, season winds are usually from northeast to south, and also some western winds are observed. All wind directions on particle formation days are between west and north, i.e., from the North Atlantic or from the Arctic Sea. This is the usual wind pattern on particle formation days observed also in southern Finland [*Mäkelä et al.* 2000b; *Kulmala et al.*, 2001]. These northern and arctic air masses consist of cold and clean air.

[29] Wind speed did not show any difference between formation days and the other days. Relative humidity was observed to be a little lower on average on formation days, although some high relative humidity values were also found. Average relative humidity on formation days was 74.9% and for the whole formation season was a little below 80% at Sammaltunturi. Atmospheric pressure showed no difference between the formation days and the other days. Wind, relative humidity, and pressure values for each event are also presented in Table 2.

[30] When studying the time differences in the starting times of formation events at the stations, wind speed and direction were used to calculate the time for an air parcel to travel from one station to the other. This was carried out to find out if newly formed particles could have been transported in a horizontal direction from one station to another. Only four of the 13 events could be reasonably explained by this. For some of the other event days, wind was blowing from the wrong direction, or wind speed was too slow to explain the time difference between the observed start times of the events. This leaves the vertical movement of the air as the most likely explanation. Unfortunately, at the moment, there are not enough data to verify this assumption.

[31] Daily weather data for the formation days were studied closely to find any repeating patterns. A temperature rise of 2°–6°C was found to be a normal pattern before or during the start of the formation event. Also, increase of solar radiation before the event was found. Some variation of solar radiation, i.e., variable cloudiness, was observed during most event days. The visibility on formation was days usually high (over 30 km), but on some occasions it

Table 1. Main Parameters of Each Particle Formation Event at Sammaltunturi and Matorova[a]

Date	Start Time[b]	End Time[b]	Length, hours:min	Start Time,[b] 1 nm	Formation Rate, $cm^{-3}\ s^{-1}$	Growth Rate, $nm\ h^{-1}$	Maximum Diameter, nm	Growth Time, hours	Particles Produced, cm^{-3}	Existing Concentration, cm^{-3}	Surface Area ($\mu m^2\ cm^{-3}$)	Volume ($\mu m^3\ cm^{-3}$)	Condensation sink ($10^{-4}\ s^{-1}$)
							Sammaltunturi						
2000													
24 April	1200	1815	6:14	1023	0.09	3.7	65.5	8	2093	400	2.06	0.09	3.5
26 April	1412	2109	6:56	1040	0.07	1.7	31.7	8	1706	779	9.79	0.63	14.0
5 May	1107	1741	6:34	0647	0.06	1.4	23.7	6	1526	83	1.51	0.09	2.2
11 August	1026	1430	4:03	0905	0.08	4.4	87.8	8	1129	424	2.28	0.12	3.6
12 August	1013	1307	2:54	0912	0.18	5.9	75.6	5	1904	403	2.32	0.12	3.7
5 September	1541	2053	5:12	1404	0.13	3.7	42.2	8	2419	143	1.48	0.07	2.4
2001													
4 April	1359	1642	2:42	1255	0.37	5.6	56.9	3	3565	630	6.13	0.39	9.0
21 April	0916	1815	8:58	0450	0.09	1.4	23.9	8	3068	511	2.80	0.12	4.7
13 May	1200	1711	5:10	0734	0.11	1.4	27.5	8	2059	155	1.75	0.10	2.7
25 Jun	1131	1658	5:26	1040	0.15	7.1	75.6	5	2923	588	7.85	0.42	12.0
30 June	1039	1444	4:05	0918	0.11	4.4	65.8	8	1676	355	4.78	0.23	7.8
3 Aug	0838	1301	4:22	0727	0.23	5.0	56.7	7	3645	124	0.48	0.02	0.8
Minimum	0838	1301	2:42	0450	0.06	1.4	23.7	3	1129	83	0.48	0.02	0.8
Mean	1138	1644	5:06	0924	0.15	3.7	50.8	6.6	2356	407	3.42	0.19	5.3
Maximum	1541	2110	8:58	1404	0.37	7.1	87.8	8	3645	779	9.79	0.63	14.0
							Matorova						
2000													
24 April	1134	1812	6:37	1011	0.14	4.3	64.3	8	3318	511	2.44	0.11	4.1
26 April	1421	2142	7:21	1043	0.09	1.7	31.1	8	2388	1022	11.82	0.81	17.0
5 May	1111	1513	4:02	0855	0.22	2.6	27.0	5	3217	717	1.32	0.04	2.4
12 May	1055	1744	6:49	0628	0.10	1.4	23.4	6	2505	69	1.64	0.10	2.5
11 August	1048	1452	4:03	0935	0.08	4.9	86.2	7	1146	368	2.16	0.11	3.4
12 August	0958	1316	3:17	0856	0.16	5.8	74.6	5	1843	372	1.96	0.09	3.2
5 September	1547	2007	4:19	1419	0.13	4.1	35.9	7	1991	203	1.44	0.07	2.4
2001													
4 April	1352	1637	2:45	1258	0.40	6.7	56.0	3	3966	493	6.97	0.45	10.0
21 April	0918	1842	9:24	0451	0.12	1.4	23.5	6	4047	495	2.95	0.13	5.0
13 May	1205	1738	5:33	0805	0.09	1.5	27.0	7	1861	151	1.97	0.10	3.1
25 June	1133	1640	5:07	1049	0.20	8.2	74.2	5	3747	674	8.63	0.44	14.0
30 June	1104	1513	4:09	0941	0.10	4.3	64.3	8	1524	444	5.73	0.27	9.2
3 Aug	0825	1326	5:00	0713	0.20	4.9	55.5	7	3570	105	0.51	0.02	0.8
Minimum	0825	1316	2:45	0451	0.08	1.4	23.4	3	1146	69	0.51	0.02	0.8
Mean	1136	1653	5:16	0926	0.16	4.0	49.5	6.3	2702	432	3.81	0.21	5.9
Maximum	1547	2143	9:24	1419	0.40	8.2	86.2	8	4047	1022	11.82	0.81	17.0

[a]Minimum, mean and maximum values for each parameter are also presented.
[b]Times are given in UTC plus 2 hours.

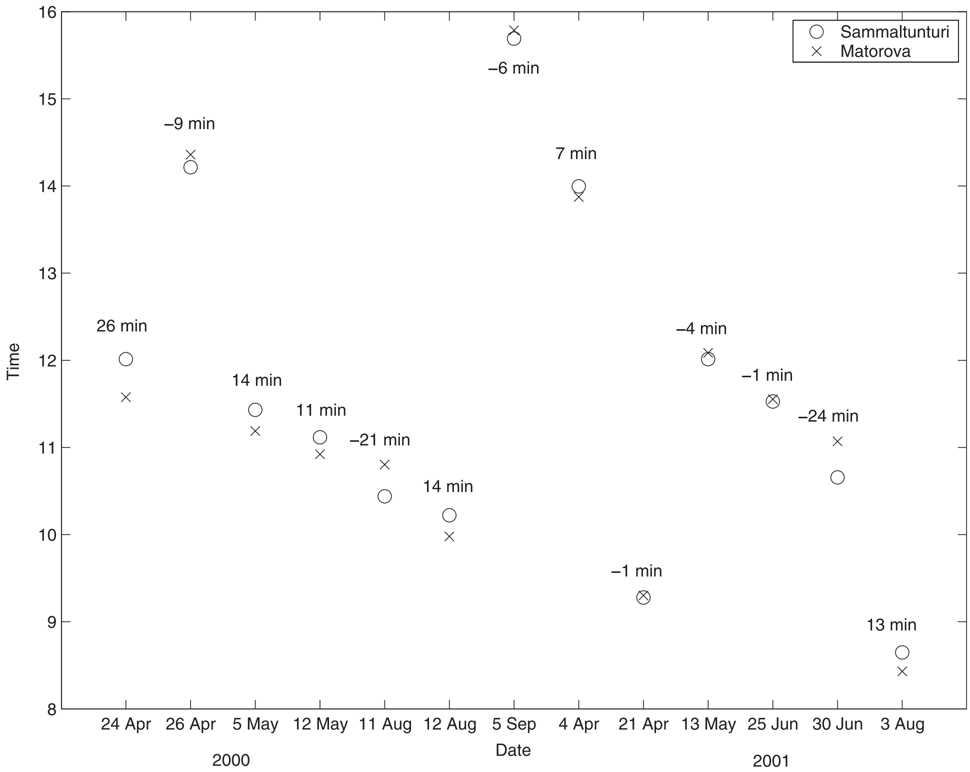

Figure 7. Starting times of observed 7–nm particle formation events at Sammaltunturi and Matorova. Also presented are the time differences of starting times (Sammaltunturi minus Matorova) of the events.

was very low (below 1 km) in the morning and rose fast before the start of the formation event. On one day, 4 April 2001, the visibility was lower (below 5 km) throughout the whole day, and slight snowfall was observed just before the event started. On all other event days the relative humidity was observed to take a large dive of 10–40%. On most occasions, there was no precipitation observed during the whole day. On some days, slight rainfall was observed in the morning well before the event.

[32] Low levels of SO_2, NO, and NO_2 concentrations during particle formation days also indicate clean polar or arctic air masses. Most of the time these concentrations were below the detection limit of the instruments (SO_2, 0.2 $\mu g\ m^{-3}$; NO_x, 0.4 $\mu g\ m^{-3}$), especially on formation days. Only during one of the 13 event days, 4 April 2001, was a significant increase of SO_2 and NO_2 concentrations seen at the time of the event, which was the only formation day having wind from the east. In one case

Table 2. Weather Parameters at Sammaltunturi During Particle Formation Events[a]

Date	Start Time[b]	Wind Direction, deg	Wind Speed, $m\ s^{-1}$	Sun Radiation, $W\ m^{-2}$	Temperature, °C	Relative Humidity, %	Pressure, hPa
2000							
24 April	1200	339	5.1	512.6	−1.5	98.8	939.0
26 April	1412	355	8.2	346.0	−3.2	85.3	946.6
5 May	1125	298	15.9	658.0	2.4	70.6	931.7
12 May	1107	336	8.2	521.4	−3.9	54.6	949.9
11 August	1026	275	5.6	393.5	7.0	83.2	941.3
12 August	1013	44	3.5	520.0	9.7	89.2	943.8
5 September	1541	16	2.5	394.9	3.1	59.9	948.4
2001							
4 April	1359	97	16.1	217.6	−13.1	91.9	932.5
21 April	0916	266	9.5	270.0	−2.6	82.6	937.1
13 May	1200	313	5.4	265.8	3.3	65.7	938.2
25 Jun	1131	41	4.1	747.8	13.5	48.3	953.6
30 June	1039	357[c]	2.1[c]	648.0	10.4	70.3	944.7
3 August	0838	306	5.9	387.9	4.6	73.2	945.9
Minimum	0838		2.1	217.6	−13.1	48.3	931.7
Mean	1138		7.1	452.6	2.3	74.9	942.5
Maximum	1541		16.1	747.8	13.5	98.8	953.6

[a]Values are 1–hour averages before the start of the event.
[b]Times are given in UTC plus 2 hours.
[c]Wind parameters on this day are from Matorova because of missing data.

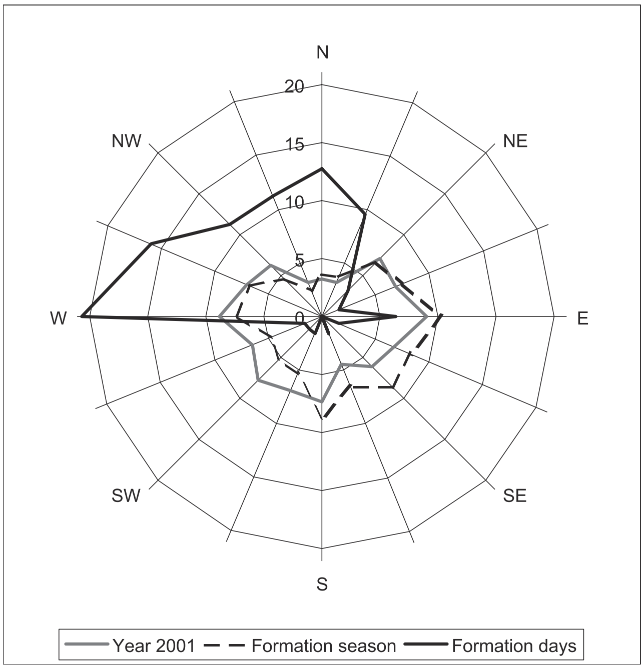

Figure 8. Wind direction at Sammaltunturi for the year 2001 for the formation season (1 April to 15 September) and for all particle formation days. Values are presented in percents of all winds.

an increase in SO_2 concentration was observed 5 hours before the event.

[33] Differences in the starting times of the events between Sammaltunturi and Matorova station could not be explained by wind direction and wind speed or by different growth rates between the stations. In southern Finland it has been observed that during formation events particle concentration is not increasing equally at different heights, and it has been suggested that particles are not born in the canopy level but higher [*Aalto et al.*, 2001]. Earlier studies in southern Finland also exclude free troposphere as a possible origin of the new particles and assume that the new particles are originating somewhere between the canopy layer and free troposphere [*Nilsson et al.*, 2001]. One explanation for the starting time differences could be vertical-scale variations in particle formation. Particle formation could be occurring at different height levels on different days and then spreading by turbulent mixing toward other vertical layers. It has to be noted that starting times of the events are manually picked from the daily surface plot and that one measurement cycle takes as long as 5 1/2 min. These two factors are causing some error in the starting times, but they still do not explain the observed differences. To determine the time difference more accurately, smaller particles at different heights should be measured with a shorter measuring cycle.

4. Conclusions

[34] Two years of continuous aerosol particle size distribution measurements have been carried out at Pallas area in northern Finland. Measurements have been made at two different heights (340 m and 560 m above sea level) and surroundings to find out the effects of local-scale conditions on new particle formation and growth, as well as the effects on the size distribution of aerosol particles. Seasonal variation was observed to be similar at both stations, high values in spring and summer and low values in winter. Aitken and accumulation modes have some difference in seasonal variation between the stations. Differences are mainly due to cloud droplet activation at Sammaltunturi.

[35] A total of 65 particle formation events were recorded during 22 months. Most of the events occurred in April and May. Elevation or surroundings of the measuring site did not have noticeable effect on new particle formation and growth. Formation events were always observed at both stations with a maximum time difference of 30 min between starting times, which could not be explained directly with wind direction and speed. Calculated potential temperatures indicated that surrounding atmosphere was undergoing turbulent mixing and experiencing fluxes during all formation days. This mixing can explain detected time differences.

[36] Solar radiation was observed to be one of the key factors needed for particle formation, even though some variable cloudiness during the events was observed. Average temperature was observed to be lower on formation days compared to the other days. Wind direction on formation days was between west and north, indicating cold and clean polar or arctic air masses. Low levels of SO_2, NO, and NO_2 concentrations on formation days also indicate clean air masses.

[37] The surrounding forest at Matorova did not have a significant effect on the variation of physical properties of aerosol particles. The slightly larger overall number concentration at Matorova could be caused by the influence of the forest, but the difference is rather small. In general, the results between the stations are surprisingly similar taking into account the different elevations and surroundings. Results indicate that the detected particle formation events are caused by larger than local-scale conditions.

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