BOREAL ENVIRONMENT RESEARCH 13: 81–92
ISSN 1239-6095 (print) ISSN 1797-2469 (online)
Helsinki 28 April 2008

Aerosol particle formation events at two Siberian stations inside the boreal forest

Miikka Dal Maso[1)8)]*, Larisa Sogacheva[1)], Mikhail P. Anisimov[2)], Mikhail Arshinov[3)], Anatoli Baklanov[2)], Boris Belan[3)], Tamara V. Khodzher[4)], Vladimir A. Obolkin[4)], Anna Staroverova[5)], Alexey Vlasov[5)], Valery A. Zagaynov[6)], Alex Lushnikov[1)], Yulia S. Lyubovtseva[1)], Ilona Riipinen[1)], Veli-Matti Kerminen[7)] and Markku Kulmala[1)]

[1)] *Division of Atmospheric Sciences, Deptartment of Physical Sciences, P.O. Box 64, FI-00014 University of Helsinki, Finland*
[2)] *Institute of Chemical Kinetics and Combustion, SB RAS, 630090, Novosibirsk, Russia*
[3)] *Institute of Atmospheric Optics, SB RAS, 634055, Tomsk, Russia*
[4)] *Limnological institute, Siberian branch of RAS, 3 Ulanbatroskaja, 664043 Irkutsk, Russia*
[5)] *Russian State Hydrometeorological University, 195196 St. Petersburg, Russia*
[6)] *Karpov institute of physical Chemistry, Vurontsovo Pole, 105064 Moscow, Russia*
[7)] *Finnish Meteorological Institute, P.O. Box 503, FI-00101 Helsinki, Finland*
[8)] *FZ Jülich, ICG-II, D-52428 Jülich, Germany*

Received 10 May 2007, accepted 9 Nov. 2007 (Editor in charge of this article: Hanna Vehkamäki)

Dal Maso, M., Sogacheva, L., Anisimov, M. P., Arshinov, M., Baklanov, A., Belan, B., Khodzher, T. V., Obolkin, V. A., Staroverova, A., Vlasov, A., Zagaynov, V. A., Lushnikov, A., Lyubovtseva, Y. S., Riipinen, I., Kerminen, V.-M. & Kulmala, M. 2008: Aerosol particle formation events at two Siberian stations inside the boreal forest. *Boreal Env. Res.* 13: 81–92.

We present one-year data on atmospheric aerosol particle size distributions covering the ultrafine size range from two measurement stations situated in central Siberia: Tomsk and Listvyanka. The size distributions were measured using Diffusion Aerosol Spectrometers (DAS), which are able to detect particles as small as 3 nm in diameter. The analysis of the size distribution time series revealed about 30 new-particle formation and growth events at both stations. The events occurred predominantly during the springtime. The average particle formation rates were 0.4 cm^{-3} s^{-1} at both stations, whereas the particle growth rates were on average 5.5 nm h^{-1} at Tomsk and 1.8 nm h^{-1} at Listvyanka. The formation and growth rates were comparable with those observed in the western part of the Eurasian boreal forest.

Introduction

Formation of atmospheric aerosol particles by nucleation events is a significant source of new atmospheric particles. Particle formation events have been observed in widely-variable environments and conditions all around the world, from extremely remote Antarctic regions to densely-populated megacities (*see* e.g. Kulmala *et al*. 2004 and references therein). The particles formed by these events have, after growing by condensation, the potential to act as cloud condensation nuclei (CCN) (Kerminen *et al*. 2005). Changes in CCN concentrations and composi-

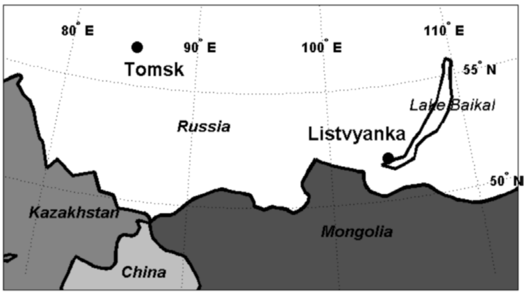

Fig. 1. Locations of the Tomsk and Listvyanka stations.

tion may lead to changes in cloud albedo, water content and lifetimes and thus affect the Earth's radiative balance as well as the hydrological cycle (Albrecht 1989, Twomey 1991, Hu and Stamnes 2000).

The boreal forest, characterized by coniferous trees, is a biome covering about 14% of the Earth's land area. By covering the northern parts of Eurasia and North America, it is the world's largest terrestrial biome. In the western part of the Eurasian boreal forest, continuous size distribution measurements have revealed frequent particle formation events (Mäkelä *et al.* 1997, Tunved *et al.* 2003, Vehkamäki *et al.* 2004, Dal Maso *et al.* 2005, 2007), and that the forest acts as a source of aerosol particles (Tunved *et al.* 2006). However, literature data on ultrafine aerosol particle concentrations and size distributions in the vast forested areas of the Siberian forests are still scarce.

Bashurova *et al.* (1992) performed measurements with screen diffusion batteries and condensation particle counters in Listvyanka for a few weeks in the late summer 1990. Their results suggested that photochemical production of particles was occurring. Koutsenogii and Jaenicke (1994) described a two-week measurement campaign performed at Listvyanka in July 1991 and one-month campaign near Novosibirsk in July 1992. They found sharp midday increases in aerosol concentrations at both sites and concluded that gas-to-particle conversion was the main particle formation mechanism. Zagaynov *et al.* (1990) reported non-continuous measurements over Lake Baikal. Koutsenogii (1997) reported three years of measurement data on Siberian aerosol mass and number concentrations. He found that submicron aerosol concentrations were higher in winter than in summer. The average number concentration of the smallest mode (diameter 24 nm) was 5200 cm^{-3}. No information on the diurnal variation was given. Matthias-Maser *et al.* (2000) studied Siberian particles of natural origin, but they concentrated on large particles larger than 200 nm in diameter. Recently, Vartiainen *et al.* (2007) reported size distributions along the route of the trans-Siberian railway and they found two occurrences of new-particle formation events in Siberia.

In this work we will present two one-year datasets from stations located in central Siberia. At both sites, ultrafine aerosol size distributions were measured and particle formation events with subsequent particle growth were observed. We will present general statistics on the measured particle number concentrations and observed particle formation events and compare the observations to those made at Finnish sites in Hyytiälä, Värriö and Pallas and the Swedish site in Aspvreten (Dal Maso *et al.* 2007).

Materials and methods

Measurement stations

The Tomsk measurement station (56.5°N, 85.1°E) is situated about 20 km from the city of Tomsk (Fig. 1). The station itself is in a grass-covered meadow, surrounded by mixed forest consisting mainly of birch, pine, spruce and larch. There are no roads or industrial activities in the immediate vicinity. During summer it is possible that forest fires influence the aerosol concentrations. Zuev *et al.* (1998) stated that being "in a vast forest zone far from oceans and mountains" makes the station suitable for observations on continental aerosol particles.

The Listvyanka measurement station (51.9°N, 104.9°E) is located on the south-western shore of Lake Baikal, a great freshwater basin surrounded by mountains (Fig. 1). The station is situated about 70 km southeast from the city of Irkutsk (population ca. 600 000), and a few km northeast of Listvyanka, a small lakeside village (population ca. 2500). The station itself is located on the top of a hill about 300 m over the surface of Lake Baikal. The sampling site is surrounded by coniferous forest consisting mainly of pine, cedar and spruce, with an admixture of deciduous trees. The main sources of possible anthropogenic pollution are the Listvyanka village and the cities Baikalsk (ca. 80 km to the east across the lake) and Irkutsk. As mentioned earlier, Listvyanka is a site where aerosol size distribution measurements were performed at the beginning of the 1990s (Bashurova *et al*. 1992, Koutsenogii and Jaenecke 1994).

Measurements

At both stations, the aerosol size distribution data were obtained by using Diffusion Aerosol Spectroscopes (DAS, Mavliev *et al*.1984, Julanov *et al*. 2002). The DAS consists of a set of grid diffusion batteries and condensation particle counter. The particle concentrations at both the inlet and outlet of the diffusion battery are measured to determine the penetration through a set of specially designed grids. By measuring the penetration with varying numbers of grids, and by knowing the size-dependent particle diffusivity, one can retrieve information about the size distribution of atmospheric aerosol particles. The DAS inversion method by Bashurova *et al*. (1991) was used for the Listvyanka data and that by Eremenko and Ankilov (1995) for the Tomsk data. The obtained size distribution is restored from integral characteristics and should be treated with caution, especially if multiple aerosol modes are present. In this analysis we were, however, interested in mainly two characteristics: the particle number concentration and the mean diameter of the particle population. We considered our methods robust enough to obtain these characteristics for our analysis.

The measurements we report here covered approximately one year, from March 2005 to March 2006 (Table 1). The coverage of the Tomsk data set was 78%, which includes a 23-day break in June 2005. If this break is omitted, the coverage of the rest of the data rises to 85%. The coverage of the Listvyanka measurements was lower: at least some measurements were recorded on 70% of all days, and the total data coverage was around 50% for the one-year period. The low coverage resulted from data outages of the length of hours rather than days.

The diameter size range covered by the DAS measurements was from 3 to about 250 nm for the Tomsk setup and from 3 to 50 nm at Listvyanka. The time resolutions of measured size distributions were seven and three minutes for the Tomsk and Listvyanka stations, respectively. The size and time resolutions were such that they enabled the analysis of the size distribution time series for the occurrence of particle formation events.

Identification of particle formation

A preliminary overview of the size distribution time series revealed that the new-particle formation occurred during a number of days. In order to quantify the occurrences of these events, the size distributions resulting from the data inversion were analysed visually. Since the new-particle formation events occurred mostly near the noontime, and since more than one event per day was rarely observed, the data analysis was performed on a day-to-day basis.

In the Tomsk dataset, each day was classified into one of three classes: event, non-event or undefined. This classification is similar to the one used in analyzing size distribution data measured in the Nordic countries (Dal Maso *et al*. 2005, 2007). In order to be classified as an event day, a day had to fulfil the following criteria:

Table 1. Overview of the measurement stations.

Station	Tomsk	Listvyanka
Coordinates	56.5°N, 85.1°E	51.9°N, 104.9°E
Measurement period	7 Mar. 2005–15 Mar. 2006	22 Mar. 2005–30 Mar. 2006
DAS size range	3–250 nm	3–50 nm

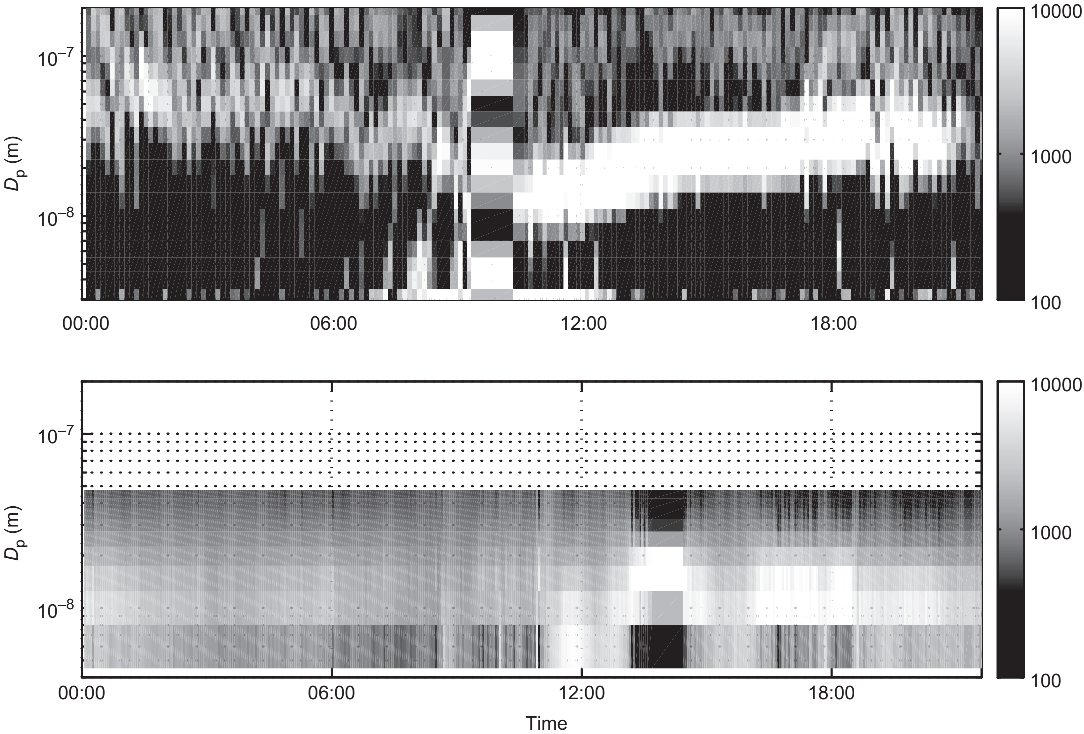

Fig. 2. Examples of particle formation events at Tomsk (top panel) and Listvyanka (bottom panel). The size distributions were measured with DAS on 22 Mar. 2005. Note that the size distributions were restored from integral characteristics and are thus approximations of the real size distributions.

— a new mode of particles had to appear in the measured size distribution,
— the mean diameter of the particles in the new mode must initially be less than 25 nm,
— the mode should prevail for several hours,
— the (geometric) mean diameter of the particles must grow.

Nonevent days were days when no clear regional new-particle formation could be observed. The days when small particles were present but either (a) they were present for only a short time (less than one hour), (b) they did not form a clearly expressed new mode or (c) their diameter did not increase with time, were classified as undefined days. The event days were classified into two classes termed class I and class II. Class I days were the days for which we could determine the particle formation and growth rates, while for class II days such a determination was not reliably possible.

For the Listvyianka dataset a similar classification was not appropriate because the large number of data outages, often around noon, would have lead to an excessive number of undefined days (as no knowledge of whether a particle formation event occurred could be obtained). Additionally, we had SO_2 concentration data available for about 100 days. We interpreted sharp peaks in the SO_2 concentration as markers of local pollution. These peaks often led to high number concentrations of ultrafine particles, which made the detection of non-event days very difficult. Therefore, we only concentrated on identifying the days where clear indications of regional particle formation events were present: the appearance of small particles and their subsequent growth over several hours.

Examples of days with particle formation events at both stations are shown in Fig. 2. It should be noted that although the size resolution of the DAS is not as good as for some other instruments currently in use, we are confident that the resolution is good enough to for the signal of a new-particle formation event to be identified, especially when we use the criterium of growth for several hours.

Event properties

In order to compare the observed particle formation events with observations at other sites, especially those in the other parts of the boreal forest, we attempted to obtain the basic characteristics of particle formation during these events. The properties of interest were the magnitude of the particle source, characterized by the rate of formation of particles, and the growth rate which in turn can give information on the amount of vapour condensing onto the particles.

The formation rate was determined as the change of the number concentration of particles with diameter < 25 nm. A first-order polynomial fitting was applied to approximate the concentration of particles smaller than 25 nm as a function of time. The formation rate is the coefficient at the linear time term. We determined the growth rate in a similar way. The geometric mean diameter was again approximated by a first-order polynomial in time, with the coefficient in time giving the growth rate. As we were using the geometric mean diameter, this method does not require a high size resolution. We were, however, assuming a unimodal distribution. This is usually the case during particle formation, when the new mode dominates the distribution. In the case more modes were present, the results obtained from our analysis would underestimate the growth rate.

The amount of pre-existing aerosol, or more specifically the condensation sink (CS), describing the sink for both the newborn small particles and the vapours condensing on the particles (*see* e.g. Kulmala *et al*. 2001), is considered to be an important factor in the particle formation process (Hyvönen *et al*. 2005). The relevant particle size range for the condensation sink is 50–200 nm (e.g. Dal Maso *et al*. 2002). Larger particles do not contribute significantly to CS because of their low concentration. We calculated the CS for all size distributions measured at the Tomsk station. Unfortunately, for the Listvyanka station the size range covered was too narrow to obtain a plausible value for CS.

Results and discussion

Number concentration

The maximum concentrations observed were of the order of tens of thousands particles per cm^3 (Fig. 3), which is typical for rural or semi-rural areas (Seinfeld and Pandis 1998). In Tomsk, the 20-day average concentration varied between about 1000 and 5000 cm^{-3}, being the highest in May 2005 and lowest in September 2005 (Fig. 3).

The mean particle number concentration during the measurements at the Tomsk station was 2480 cm^{-3} (median = 1950 cm^{-3}, SD = 2000 cm^{-3}) (Fig. 3). The total particle number concentrations in Tomsk were comparable with those in Hyytiälä, central Finland, where the average total concentration was 2300 cm^{-3} (median = 1850 cm^{-3}). The concentrations in Tomsk and Hyytiälä are not directly comparable, since the size distribution measurements at the Finnish site extend to larger sizes. However, these larger sizes are generally less populated, so we considered the agreement quite good. According to the measured particle number concentrations, the location of the Tomsk station can be termed rural or at least semi-rural, as it does not seem to be heavily influenced by the nearby city.

In Listvyanka, particle number concentrations were clearly higher than those at the Tomsk station (mean = 4690 cm^{-3}, median = 4090 cm^{-3}, SD = 3150 cm^{-3}). This feature is somewhat surprising, since the Tomsk station is situated closer to an urban centre than the Listvyanka station. The measured particle count in the 3–50 nm diameter range in Listvyanka was on average twice as high as that in Tomsk and Hyytiälä. In Hyytiälä, for example, the average particle number concentration in the size range 3–50 nm was 1240 cm^{-3} (median 700 cm^{-3}), less than a third of that observed in Listvyanka.

Number and seasonal distribution of particle formation events

By using the classification criteria described earlier, we found 32 days that fulfilled the criteria of a regional-scale particle formation event for the Tomsk station. This is approximately 10% of

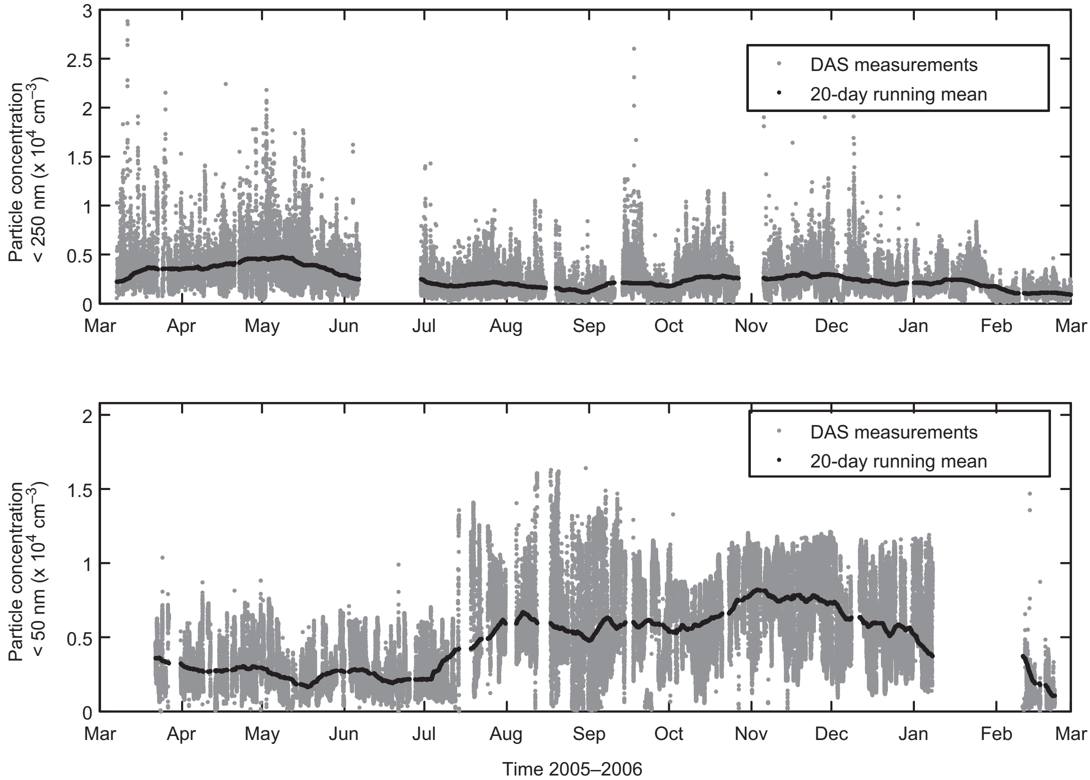

Fig. 3. Time series of the measured total particle number concentrations at the Tomsk (top panel) and Listvyanka (bottom panel) stations.

all days that could be classified. 147 days (44% of the total) were classified as undefined and 34 (10% of the total and 23% of the undefined days) of those were so because measurements were not available from some part of the day. 153 days (46%) were classified as non-event days. If undefined days are left out of the classification, the event fraction was 20%.

Table 2. Number of event, nonevent and undefined days each month at the Tomsk station.

Month	Events	Nonevents	Undefined	No data
January	0	19	12	0
Feruary	1	18	8	1
March	5	22	11	2
April	7	4	18	1
May	11	7	13	0
June	1	1	6	22
July	0	20	11	0
August	2	15	11	3
September	1	12	15	2
October	2	11	13	5
Novermber	0	9	17	4
December	2	15	12	2
Total	32	153	147	42

The majority of the classified formation events in Tomsk occurred in spring, in April and May (Table 2 and Fig. 4, top panel). Of the 32 events, 18 (56%) occurred during these two months. Autumn (September–October) was a time with few particle formation events, but also less non-event days. The statistics for June was poor, as data were available for only eight days.

In Listvyanka, we observed 31 event days of the 253 days (12%) that could be analysed. Most of these events happened in spring–summer from April to July. Only four events were seen in October–February (Table 4 and Fig. 4, bottom panel).

By comparing the occurrence times of particle formation events with the time series of the total particle concentration, some differences between the stations could be seen. In Tomsk, the period of most particle formation events coincided with the period of the highest particle concentrations in March–May. In Listvyanka, events were not frequent during the period of the highest particle concentrations from August to December. This could be due to two different reasons: either the high particle concentrations prevented new-particle formation or local pollution "masked" regional particle formation in the measured size distributions.

Event properties

Of the 32 event days observed in Tomsk, 18 were classified as class I events and analyzed for the properties of the particle formation burst (Table 4). The growth rate of the geometric mean diameter of newly-formed particles varied between 2.6 and 23 nm h^{-1}, being on average 5.5 nm h^{-1} (median = 3.9 nm h^{-1}). The average value was influenced quite much by the very high growth rate (23 nm h^{-1}) observed on 4 April 2005. Without this day the average growth rate was 4.5 nm h^{-1} with the median = 3.5 nm h^{-1}. The formation rates of new particles varied between 0.04 and 1.1 cm^{-3} s^{-1}, with both mean and median being equal to 0.4 cm^{-3} s^{-1}. The number concentration of new particles formed by the formation events was on average 5700 cm^{-3} (median = 4700 cm^{-3}). Typically, the period of increasing particle number concentrations lasted for three hours, while the duration from the start of the appearance of new particles to the end of observable growth was on average six hours.

Table 3. Number of event days, days with measurements and days with no data at the Listvyanka station.

Month	Events	Data	No data
January	2	7	24
Feruary	1	10	18
March	3	24	17
April	7	23	7
May	4	28	3
June	7	26	4
July	4	21	10
August	2	20	11
September	0	15	15
October	1	24	6
Novermber	0	30	0
December	0	25	6
Total	31	253	121

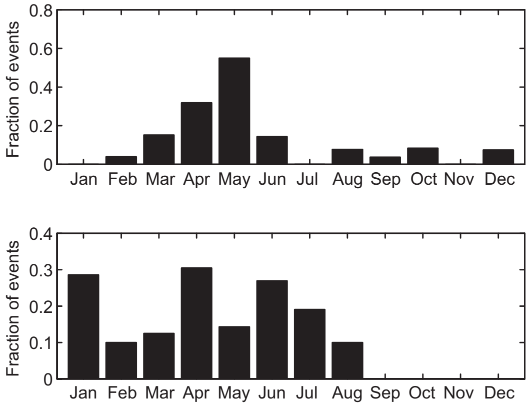

Fig. 4. Seasonal variation of events at the Tomsk (top panel) and Listvyanka (bottom panel) stations.

Of the Listvyanka dataset, 18 days were analysed (Table 4). The growth rates were found to be lower than those in Tomsk, varying between 0.1 and 3.5 nm h^{-1} with the mean and median values of 1.8 and 1.7 nm h^{-1}, respectively. These growth rates should be treated as estimates of the lower limit of the range, since in at least some cases the pre-existing particles influenced the geometric mean diameter. The particle formation rates were on average similar to those in Tomsk, varying between 0.13 and 0.6 cm^{-3} s^{-1} with both mean and median being equal to 0.4 cm^{-3} s^{-1}. The average number concentrations of particles produced during a particle formation event was 3340 cm^{-3}, ranging from 930 to 7360 cm^{-3}. The lower values, compared to Tomsk, were mainly due to the shorter durations of particle-formation bursts.

We did not find a clear seasonal variation in the formation and growth rates, mainly due to the poor statistics of the events. In Tomsk, the

Table 4. Results obtained by analysis of the size distribution time series.

Station	Tomsk	Listvyanka
Mean concentration	2480 cm^{-3}	4690 cm^{-3}
Median concentration	1950 cm^{-3}	4090 cm^{-3}
Number of NPF events	32	31
NPF events analysed	18	18
Mean growth rate	5.5 nm h^{-1}	1.8 nm h^{-1}
Average particles formed	5700 cm^{-3}	3340 cm^{-3}
Mean formation rate	0.4 cm^{-3} s^{-1}	0.4 cm^{-3} s^{-1}

highest formation rates occurred in April 2005, and the lowest in February and October. There were a few events with very high growth rates in April–May, but the rest of the days showed only a slight rising trend towards summer. The Listvyanka data displayed even a weaker seasonal variability.

Diurnal behaviour of the aerosol size distributions

In order to get a better overview of the general behaviour of the particle size distribution, the diurnal-mean size distributions for event and non-event days were calculated to produce surface plots, along with plots of calculated total and nucleation mode particles number concentrations, of the "average days" (Figs. 5 and 6). Note that for Listvyanka, no figure for non-event days could be drawn because non-events were not classified. Instead, we averaged over all the days that were not events. Note also that, as already discussed, the inversion of a size distribution is made from its integral properties, so the inverted size distribution represents only an approximation to the real size distribution.

In Tomsk, the total particle number concentration was 2000–3000 cm^{-3} during the night and morning, with a minor contribution (fewer than 500 cm^{-3}) from the nucleation mode (< 25 nm). After 07:00 am, the particle number concentration started to increase, resulting from the increase in the nucleation mode particle number concentration. At the same time, the number concentration of larger Aitken-mode particles (25–100 nm) decreased. The total particle number concentration rose, reaching its maximum of about 5500 cm^{-3} around noontime, after which the concentration dropped again. The growth of the nucleation mode particles is clearly visible, and the growth rate of this "average event" was 4 nm h^{-1}.

On non-event days, the particle concentration in the early morning was lower than that on event days, between 1500 and 2000 cm^{-3}. The concentration rose again after 07:00 am, but most of this increase was in the size range > 25 nm. The number concentration of nucleation mode particles stayed below 1000 cm^{-3}, and the average total particle number concentration did not exceed 2500 cm^{-3}. No growth was observable in the size distribution.

The diurnal cycle of the particle number concentration observed on non-event days can be explained either by particles produced by anthropogenic activities (traffic, industry, burning) or by atmospheric new-particle formation at a point or line source away from the measurement station.

The daytime condensation sink (CS) at the Tomsk station was on average 1.6×10^{-3} s^{-1}, with both median and geometric mean being equal to 1.1×10^{-3} s^{-1}. The average condensation sink on event days was 1.6×10^{-3} s^{-1} (median = 1.1×10^{-3} s^{-1}, geometric mean = 1.2×10^{-3} s^{-1}). On non-event days, CS was on average 1.5×10^{-3} s^{-1} (median = 1.0×10^{-3} s^{-1}, geometric mean = 0.9×10^{-3} s^{-1}), slightly lower than on event days. This was surprising, since we expected that event days would have a lower condensation sink because a high value of CS is considered to inhibit new-particle formation (*see* e.g. Hyvönen *et al.* 2005). We compared these values with the analysed CS at the Nordic stations (Dal Maso *et al.* 2007). The overall mean CS was lower than those in southern Finland (2.4×10^{-3} s^{-1}) and Sweden (3.1×10^{-3} s^{-1}) but higher than that in the Finnish Lapland ($0.6–1.0 \times 10^{-3}$ s^{-1}). The event day values of CS in Tomsk were lower than those in southern Finland and Sweden, but higher those in Lapland. In light of these observations, and keeping in mind that the size range covered was narrower in Tomsk than at the other sites, CS does not seem to be a limiting factor in whether a particle formation event occurs at the Tomsk site.

At the Listvyanka station the event-average diurnal evolution showed total particle number concentrations of 2500–3000 cm^{-3} in the morning, of which about 2000 cm^{-3} were nucleation mode (< 25 nm) particles. Again, there was a rise in the total particle number concentrations to around 4000 cm^{-3} after 07:00, caused by an increase in the nucleation mode particle number concentration. The growth of the new particles is visible in the averaged size distributions. On all the other days (Fig. 6), the diurnal pattern of the number concentration was very similar, except

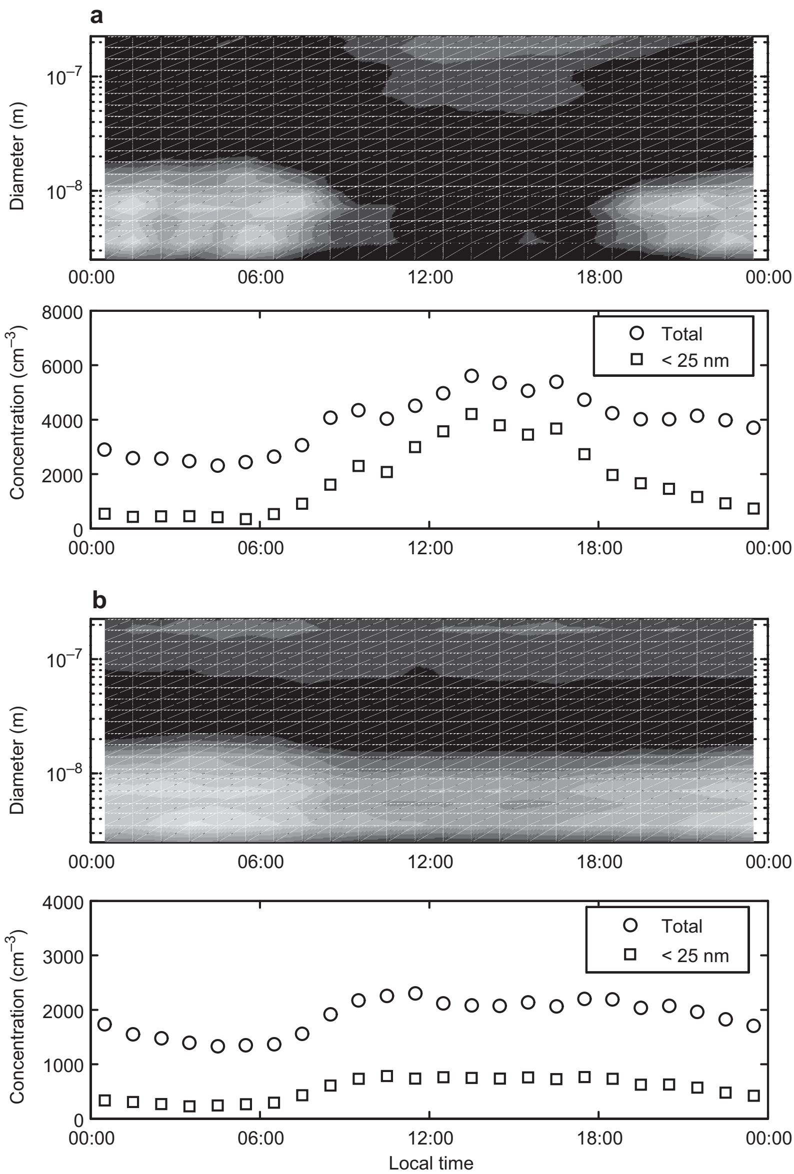

Fig. 5. The average diurnal evolution of the measured size distributions and total number concentrations at the Tomsk measuring station on (**a**) event and (**b**) non-event days. Note that the size distributions were restored from integral characteristics and are thus approximations of the real size distributions.

that the total particle number concentrations were higher in the morning and no evident particle growth was observed. We speculate that this variation was at least partly due to anthropogenic sources, even though it is also possible that some natural, photochemical new-particle formation occurred on these days as well. If present, it is possible the new-particle formation was not strong or well-defined enough to be captured by our event classification.

Air mass influence

At the Nordic stations, particle formation is strongly influenced by the air mass history, with clean air advection from the North Atlantic being most favourable to new-particle formation (Nilsson *et al*. 2001, Sogacheva *et al*. 2005). Arctic air is observed in our area of interest only rarely, and the original properties are transformed while the air travels over the Eurasian continent. To

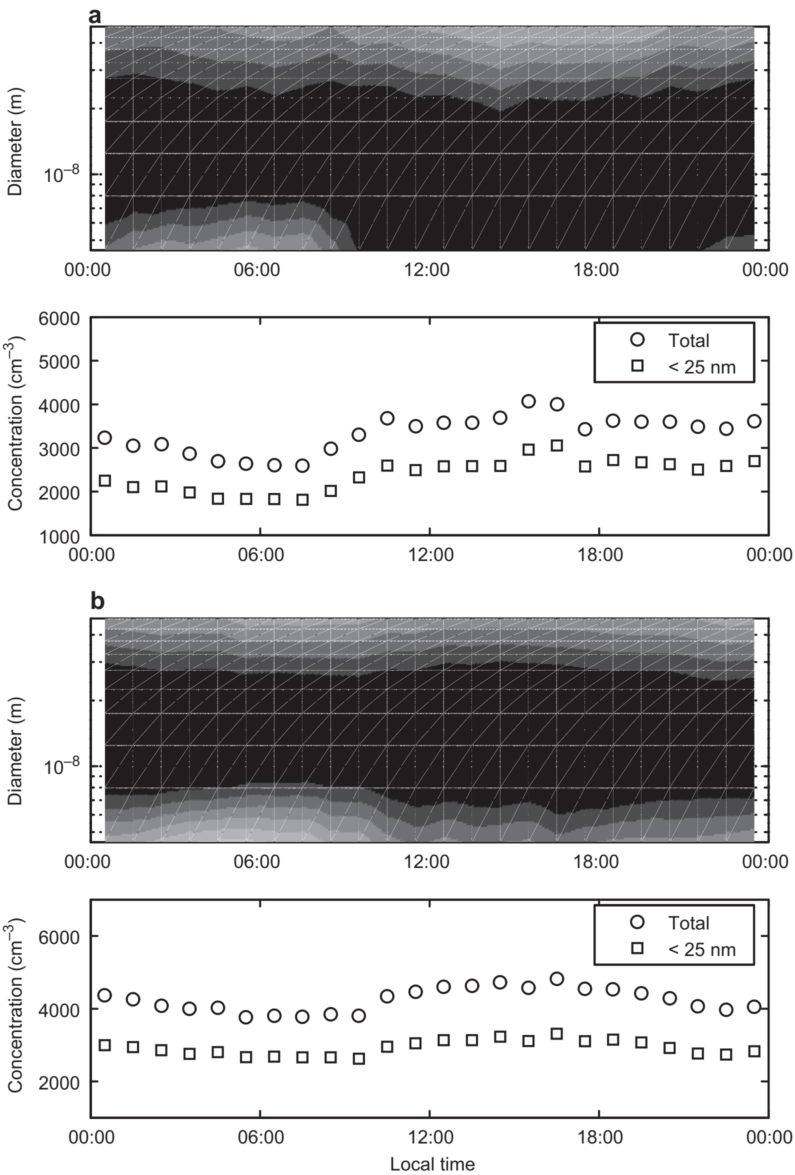

Fig. 6. The average diurnal evolution of the measured size distributions and total number concentrations at the Listvyanka measuring station on (**a**) event days and (**b**) all other days. Note that the size distributions were restored from integral characteristics and are thus approximations of the real size distributions.

investigate the air mass history, we performed a basic trajectory analysis using the NOAA HYSPLIT4 model (Draxler and Hess 1998). We calculated 96-hour (four days) backward trajectories of both stations for all observation days using the FNL meteorological archives (NCEP model output, http://www.arl.noaa.gov/ss/transport/archives.html).

At the Listvyanka station, the great majority of air masses arrived from western directions, and no difference between event and non-event days was found. For the Tomsk station, the air mass history was more variable, with transport occurring from all directions except southeast (Mongolia). On event days, air masses arrived predominantly from northern and north-western directions, with some days with western transport. Non-event and undefined days were more mixed with respect to air masses. The analysis showed that at least for the Tomsk station, new-particle formation was occurring in air masses advected over the boreal forest area.

Concluding remarks

In this paper, we have presented two datasets of atmospheric aerosol size distributions with a size detection limit down to about 3 nm in diameter, measured at the central Siberian stations Tomsk and Listvyanka. One of the stations, Tomsk, samples air representative of a rural or semi-rural background, while Listvyanka is probably influenced by anthropogenic emissions.

By analysing measured particle number size distributions, we found a number of days with clear new-particle formation characterized by an increasing number concentration of very small particles and their subsequent growth to larger sizes during the daytime. The fraction of event days, 10%–12%, (20% if undefined days were disregarded) was lower than that observed in the Nordic countries (Dal Maso *et al*. 2007). The number of non-event days in Tomsk was about 45% of all days, which is lower than that observed in the Finnish Lapland. The new-particle formation was most active around the springtime as is also the case also at Nordic boreal sites. The characteristics of the particle formation events observed at the Tomsk station were similar to those reported in the literature for other sites of the same kind (Kulmala *et al*. 2004). The mean particle growth and formation rates in Tomsk were 5.5 nm h^{-1} and 0.4 cm^{-3} s^{-1}, respectively. At Listvyanka, the average particle growth rate was lower (1.8 nm h^{-1}) than that in Tomsk, whereas the average particle formation rate was the same (0.4 cm^{-3} s^{-1}).

In Tomsk, the condensation sink caused by pre-existing particles had no significant effect on new-particle formation events, but the events were favoured by the transport of air masses from northern to western directions. In Listvyanka, no air mass preference was found but the particle number concentrations shortly before the new-particle formation events were lower than on other days.

In the absence of comprehensive measurements of trace gas concentrations, it is difficult to say anything definite about the mechanism of new-particle formation or about vapours making these particles to grow in size. It is tempting, however, in light of the observed particle formation and growth rates that were similar to those observed at the Nordic stations, to speculate that also the mechanism would be the same, at least in the Tomsk station. The obvious influence of local pollution in the Listvyanka means that new-particle formation observed there could also be driven by anthropogenic emissions. In any case, our findings show that regional new-particle formation is occurring regularly also in the Siberian boreal forest, as was suggested by the findings of Vartiainen *et al*. (2007).

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