RUSSIAN JOURNAL OF EARTH SCIENCES, VOL. 11, ES4002, doi:10.2205/2009ES000410, 2010

PROCEEDINGS OF THE INTERNATIONAL CONFERENCE
Electronic Geophysical Year: State of the Art and Results
3–6 June 2009 • Pereslavl-Zalessky, Russia

Comparison of formation conditions of secondary aerosol particles in boreal forests of Southern Finland and Siberia

Yu. S. Lyubovtseva,[1] V. A. Zagaynov,[2] T. V. Khodzher,[3] M. Kulmala,[4] M. Boy,[4] M. Dal Maso,[4] H. Junninen,[4] V. A. Obolkin,[3] V. L. Potyomkin,[3] Yu. G. Biryukov,[2] and A. A. Lushnikov[2]

Received 12 December 2009; accepted 2 March 2010; published 23 2010.

Complex measurements of size distributions and chemical content of aerosols, trace gases SO_2, NH_3, ozone, and meteorological parameters were performed in boreal forests of Central Siberia during 03.2005–05.2006 at the rural station of Listvyanka. The data from Listvyanka are then analyzed from the point of view of their similarities and differences with respective data at Hyytiälä background station (Southern Finland). The goal of this paper is to compare the conditions of formation of secondary aerosol particles in boreal forests in two different regions: in Southern Finland (Hyytiälä station) and Siberia (Listvyanka station). Our main conclusion is that the particle formation–growth processes in Listvyanka occur in presence of high (compared to Hyytiälä) concentrations of sulfur dioxide and ammonia. We show that their concentrations are enough for providing the particle growth up to 3.5 nm h^{-1} with much higher concentration of the nucleation mode particles in contrast to Hyytiälä, where the sulfuric compounds are much less concentrated and the nucleation mode particles grow by consuming low volatile organic vapors that result from photochemical processing of highly volatile organic plant emissions. On the other hand, the nucleation bursts in Siberia occur more seldom than in Hyytiälä. The difference in UVB irradiation regimes at Siberian stations and in Hyytiälä can lead to different seasonal patterns of the particle formation–growth rates and the concentrations of nucleation mode in Siberia. *KEYWORDS: secondary aerosol particles, boreal forests, Southern Finland, Siberia.*

Citation: Lyubovtseva, Yu. S., V. A. Zagaynov, T. V. Khodzher, M. Kulmala, M. Boy, M. Dal Maso, H. Junninen, V. A. Obolkin, V. L. Potyomkin, Yu. G. Biryukov, and A. A. Lushnikov (2010), Comparison of formation conditions of secondary aerosol particles in boreal forests of Southern Finland and Siberia, *Russ. J. Earth. Sci., 11,* ES4002, doi:10.2205/2009ES000410.

Introduction

Boreal forests covering the Northern parts of three continents (Europe, Asia and America) are commonly recognized as one of the most intense supplier of secondary aerosols to the Earth atmosphere [*Kulmala et al.,* 2004]. Although the seasonal variations of the atmospheric conditions typical for the boreal zone are similar, other physical-geographical characteristics are very diverse. This can tell on the productivity of particle sources and the properties of aerosols formed in different regions of the boreal zone.

In the Northern European boreal regions long term studies of aerosol formation and growth were performed at several measurement sites: in Hyytiälä, [*Dal Maso et al.,* 2005; *Kulmala et al.,* 2001a, 2001b, 2004; *Lyubovtseva et al.,* 2005; *Mäkelä et al.,* 2006a, 2006b], in Pallas and Varrio [*Vehkamäki et al.,* 2004], and in the Scandinavian rim of the boreal region in Aspvreten [*Tunved et al.,* 2003].

These studies clearly demonstrated that European boreal region is a substantial source of aerosol number and mass concentrations [*Kulmala et al.,* 2004; *Tunved et al.,* 2006]. Regional nucleation events occur here regularly (25–40% of days) and typically lead to the increase in particle number

[1]Geophysical Center of RAS, Moscow, Russia
[2]Karpov Institute of Physical Chemistry, Moscow, Russia
[3]Limnological Institute of RAS, Irkutsk, Russia
[4]Department of Physical Sciences, University of Helsinki, Helsinki, Finland

concentration by 2–10 times. The freshly formed particles grow up to 10–100 nm in diameter and thus reach the climatically active size range. Typically the particle formation processes are active in spring and early autumn.

Our knowledge on the aerosol activity of boreal forests in Siberia is much more poor, although the total forest coverage in Siberia exceeds that in Finland by ten times. Approximately a half of total forested area in Siberia is covered with larch forests in contrast to European boreal forests, where spruces and pines dominate.

Several expeditions studied formation of aerosols in the boreal forests of Siberia. First measurements of fine ($d >$ 20 nm) in Siberia were conducted by [*Zagaynov et al.,* 1989] over lake Baikal and on the territory of rural site Listvianka settlement on the East side of Baikal (70 km south-east from the city of Irkutsk) during summer periods of 1988 and 1989. The average number concentration of nanoparticles was shown to vary from 10^3 to 5×10^3 cm^{-3}. The number concentration of accumulation mode was low, 10–100 cm^{-3}. In the regions of Southern Baikal and the territories near the town of Baikalsk, where a highly productive source of anthropogenic pollution (the Baikalsk pulp and paper mill) is located, the number concentration of accumulation mode reached 10^4 cm^{-3}.

At the end of July and in the beginning of August of 1991 the measurements of atmospheric condensation nuclei size distribution within the size range 3–100 nm on the territory of Listvianka were performed by [*Bashirova et al.,* 1992] and [*Kutsenigii et al.,* 1994]. They found that the average particle number concentration stretched from 10^3 to 10^4 cm^{-3} and well correlated with the Sun radiation intensity. These measurements showed the presence of bimodal size distributions with well separated nucleation mode at $d_1 = 20$ nm, concentration $N_1 = 3200$ cm^{-3} and the Aitken mode at $d_2 = 100$ nm and $N_2 = 2900$ cm^{-3}. The authors paid a special attention on the occurrences of night maxima in the nucleation mode.

Complex measurements of aerosol size distributions at $d > 3$ nm, trace gases SO_2, NH_3, ozone and meteorological parameters were performed in the summer of 2003 and in the period 03.2005–05.2006 at the rural station of Listvyanka and at the background station of Mondy [*Zagaynov et al.*, 2006]. These measurement allowed for estimating the total concentration of aerosol particles, average size, the width of the size distribution of the nucleation mode, the formation rate (FR), and the growth rate (GR). The authors showed that for background and polluted atmosphere the formation rate differed by two orders of magnitude FR$= 1.4 \times 10^{-2}$ cm^{-3} s^{-1} at Listvyanka station, FR $= 2.5$ cm^{-3} s^{-1} at Mondy station), while the growth rate was higher by an order of magnitude at the background conditions of Mondy (GR$_{\max} = 20$ nm h^{-1}). The complexity of data processing in 2003 was related to intense forest fires in nearby regions. The fires distorted strongly the typical conditions for formation of secondary aerosols in this region. During the fire episodes the number concentration of submicron aerosols grew by several times leading to the growth of condensational sinks and thus suppressing formation of new particles. In addition, the concentrations of nitrogen oxides grew by 3–5 times changing substantially the diurnal dynamics of nanoparticle formation and growth.

Observations of particle formation events at two Siberian stations: Listvyanka and the Experimental Station Tomsk, April 2005–April 2006 were presented in [*Dal Maso et al.,* 2008]. They found that the fraction of event days (10–12%) in Siberia is lower than that observed in Nordic countries. The particle formation activity was centered around spring time in Tomsk as well as at the Nordic boreal sites. The characteristics of the particle formation events observed at the Tomsk station were of the same order as those reported in literature for other similar sites [*Kulmala et al.,* 2004]. The mean growth rate at Tomsk station was 5.5 nm h^{-1}, at Listvyanka station the growth rate was lower: only 1.8 nm h^{-1} on average. The mean formation rate was 0.4–1 cm^{-3} s^{-1} for both stations. The number of new particles formed during the nucleation events was on average 5.7×10^3 cm^{-3} at Tomsk station and 3.3×10^3 cm^{-3} at Listvyanka station.

The goal of this paper is to compare the conditions of formation of secondary aerosol particles in boreal forests in two different regions: in Southern Finland (Hyytiälä station) and Siberia (Listvyanka station). The data of observations from Listvyanka rural station during the period 03.2005–03.2006 were analyzed in order to investigate possible links between physical, chemical, and meteorological parameters in the process of formation of aerosol during different seasons. The seasonal patterns of trace gases O_3, SO_2, NH_3, HNO_3, the meteorological parameters (temperature and relative humidity), and UV–radiation were measured and used for evaluation of their influence on the seasonal patterns of particle number concentrations, size distributions within 3 nm–2 μm, and the particle formation and growth rates. The seasonal dependencies of all above mentioned parameters are compared for both stations.

Site Description

The station of Listvyanka (51.9°N, 104.7°E) is located on the south–western shore of lake Baikal 70 km toward SE from the city of Irkutsk (see the map in Figure 1). The samples were collected at the top of a hill (the elevation about 300 m over the level of the lake Baikal and 700 m above sea level) several km toward NE from the village of Listvyanka on the Baikal shore. Possible sources for the anthropogenic releases are the Listvyanka village itself, the town of Baikalsk on the opposite shore of the lake (approximately 80 km from Listvyanka), and the city of Irkutsk. The Listvyanka village is populated with about 2500 inhabitants and lays at the foot of the hill. No industrial enterprizes producing considerable atmospheric contaminations are located nearby. The main sources of the atmospheric contaminations are traffic and the stove heating of the houses in winter time. The city of Irkutsk (approximately 600 000 inhabitants) produces a large amount of gaseous and aerosol contaminations, but it is located rather far away from the observation point and is screened with mountains. The sampling site is surrounded with the forests of coniferous trees (larches, pines, cedars,

spruces) with an admixture of deciduous trees like birches and aspens.

The Listvyanka station works since 2000 within the International Monitoring Net of Acid Deposits in Eastern Asia (EANET). The monitoring includes weekly sampling of aerosols and four trace gases (SO_2, NH_3, HCl, HNO_3) The filter pack method is used for sampling the aerosols. The sample analysis performs in laboratories of Limnological institute of Siberian Branch of RAS. The chemical contents of the particle soluble fractions (NH_4^+, SO_4^{2-}, NO_3^-) are analyzed by the methods of highly effective gas chromatography, and atomic absorption spectrophotometry [*Golobokova et al.,* 2005; *Khodzher et al.,* 1994, 1997].

The aerosol size distribution data were obtained with the diffusion aerosol spectroscope DAS [*Julanov et al.,* 2002]. This instrument comprises the set of grid diffusion batteries, the particle magnifier enlarging the particle sizes up to 0.5 μm, and the laser aerosol counter that serves as a particle concentration counter and for determining the size spectra of submicron aerosols. The ratio of outlet to inlet concentrations determines the penetration of aerosols through the diffusion batteries with various number of grids. The data on penetrations are then converted to the particle size distributions within the size interval 3–50 nm. The laser aerosol spectroscope provides the information on the size spectra of the submicron particles of sizes $0.1 - 2$ μm.

Because the measurements of O_3 at Listvyanka station were not performed, the data on O_3 from the station of Bol'shie Koty (approximately 30 km to the North from Listvyanka) were used as the characteristics of the seasonal pattern of ozone in Listvyanka. The ozone concentration was measured using an UV absorption analyzer (Dylec Model 1007–AHJ) [*Pochanart et al.,* 2003; *Potyomkin et al.,* 2004].

As the data on Sun radiation at Listvyanka station were inaccessible we used the data on the seasonal pattern of ground level UV radiation $\lambda = 310$ nm in Irkutsk located 70 km far away from Listvyanka [*Mikhalev et al.,* 2001].

The station of Hyytiälä is located in Central Finland (61°51' N, 24°17' E, 181 m above sea level) with extended areas of Scots pine dominated forests. The conditions of this site are typical for remote locations. However, the measurements were occasionally affected by the pollution from the station building (0.5 km away) and the city of Tampere (60 km away). A more detailed description of this station – SMEAR (Station for Measurement Forest Ecosystem–Atmosphere Relation) and its instrumentation is available in *Kulmala et al.,* [2001a] and *Hari and Kulmala* [2005].

At the Hyytiälä forest station, the trace gas concentrations (SO_2, O_3, NO_x) were measured continuously at different heights using a 72-meter high mast. The NO and NO_x (NO + NO_2) concentrations were measured with one chemiluminescence analyzer (TEI 42C TL, Thermo Environmental, Franklin, MA, USA). The O_3 concentration was measured with an ultraviolet light absorption analyzer (TEI 49, Thermo Environmental, Franklin, MA, USA). SO_2 concentration was measured with a fluorescence analyzer (TEI 43 BS, Thermo Environmental, Franklin, MA, USA). Aerosols were measured using a twin Differential Mobility Particle Sizer System [see e.g., *Aalto et al.,* 2001], measuring particle number concentrations in the size range 3–600 nm by using

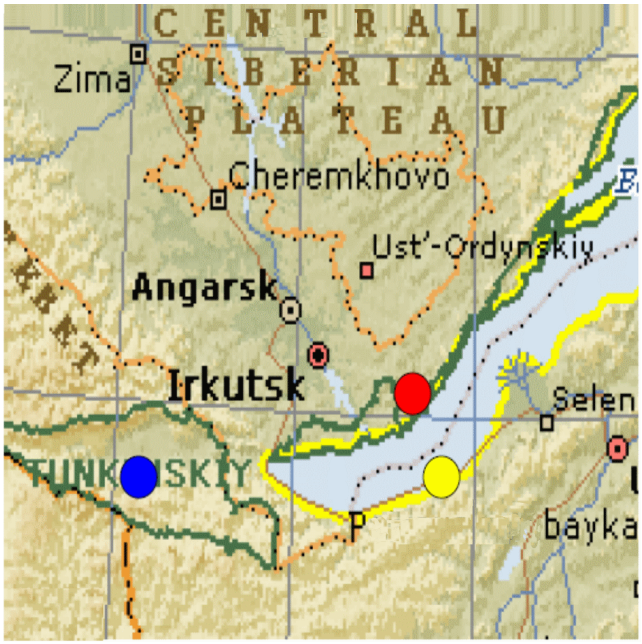

Figure 1. The map of the southern part of the lake of Baikal. Shown are: red circle – Listvyanka, blue circle – Mondy, yellow circle – Baikalsk.

electrical mobility sizing. The measurements were done at 2 m above ground level with time resolution of 10 min. A more complete description of the station and its instrumentation can be found in *Kulmala et al.* [2001a].

Results

The measurements of the aerosol characteristics at Listvyanka station were conducted during 03.2005–03.2006. A complete scan through the size range 3–2000 nm was completed every half hour. In addition, we measured the particle size distributions, particle chemical composition (sampling on filters, SO_4^{2-}, NH_4^+ NO_3^-), concentrations of (SO_2, HNO_3, NH_3) meteorological parameters, air pressure, temperature, RH, wind direction and speed.

Below we consider the seasonal trends of monthly mean values of meteorological parameters, UVB irradiation, and the concentrations of trace gases, as well as the seasonal patterns of the concentration of the nucleation mode, FR and GR, and compare their variations with those at Hyytiälä station for the whole period of measurements.

Meteorological Parameters

The influence of meteorological parameters on the processes of new particle formation was analyzed by *Boy et al.* [2002, 2003], *Weber et al.* [1997], *Birmili et al.* [2003], *Lyubovtseva et al.* [2005], *Hyvönen et al.* [2005]. Triggering role of solar radiation and the highest correlation of UV

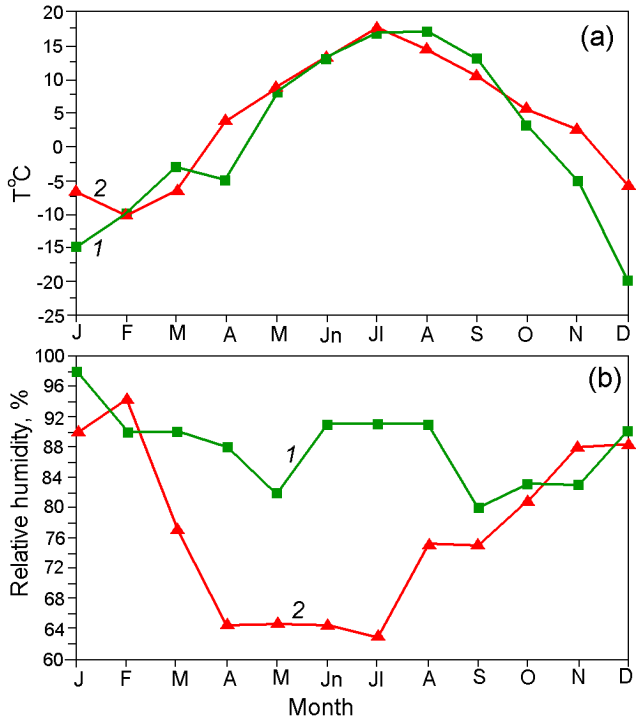

Figure 2. Seasonal behavior of monthly mean temperature (a) and relative humidity (b) in Listvyanka and in Hyytiälä. curves 1 – monthly mean temperature and humidity in Listvyanka, curves 2 – monthly mean temperature and humidity in Hyytiälä.

radiation with the particle formation rate were discussed in *Clement et al.* [2001], *Boy et al.* [2003], *Birmili et al.* [2003], *Lyubovtseva et al.* [2005]. In a number of works an anticorrelation of relative humidity and the formation rate of new particles was observed [*Hyvönen et al.,* 2005; *Lyubovtseva et al.,* 2005; *Weber et al.,* 1997]. This fact can be attributed to the increase of condensational sinks (growth of the surface of submicron particles due to condensation, [*Kulmala et al.,* 2001a] as well as to not well studied mechanisms of particle generation [*Boy et al.,* 2002; *Lyubovtseva et al.,* 2005; *Weber et al.,* 1997], The particle formation processes are also connected with the temperature regime of the lower atmospheric layer. The position of inversion layer defines the possibility of accumulation of trace gases in the atmosphere and thus the concentrations of gases participating in the processes of secondary aerosol formation. Emissions of organics and especially monoterpenes from plants also grow with temperature. In connection with these facts we focus on the detail characterization of meteorological parameters at Listvyanka station and compare them with those at the Hyytiälä station for the same period.

During the coldest winter months the observed mean temperature in Lisvyanka was about -15°C in December and the warmest summer months (July–August) it was about $+17^{\circ}$C. The seasonal patterns of the monthly mean temperature in Hyytiälä and Listvyanka are shown in Figure 2a. Only in wintertime the monthly mean temperature in Hyytiälä was higher by several degrees.

The values and seasonal variations of relative humidity (RH) were essentially different for both stations (Figure 2b). During the whole year the mean monthly RH in Listvyanka changed within a very narrow range, 80–95%. In summertime RH was 90%. In contrast, RH in Hyytiälä revealed a clearly expressed seasonal behavior. In wintertime monthly mean RH was higher than 90%. From April to July RH was about 60% (30% lower than in Listvyanka).

As the measurements of global and UV radiation are absent at Lisvyanka station, we used the spectral measurements of the ground level UV radiation at $\lambda = 310$ nm in Irkutsk (52°N, 104°E) [*Mikhalev et al.,* 2001] for the qualitative characterization of the Sun radiation level. The use of wavelength $\lambda = 310$ nm is justified because this part of UVB spectrum (280–320 nm) produces excited oxygen atoms $O(^1D)$ in the troposphere. *Boy et al.* [2002], and *Arshinov et al.* [2006] showed that the $O(^1D)$ production decreased by more than a decimal order at wavelengths below 295 nm and above 325 nm. Chemical cycles with $O(^1D)$ are the main source of hydroxyl radical OH, one of the most reactive species responsible for production of low volatile vapors.

The monthly mean UVB radiation intensity variations at $\lambda = 310$ nm in Irkutsk are shown in Figure 3. For a comparison analogous data on the ground level UV radiation at $\lambda = 310$ nm in Hyytiälä measured in 2005 year are presented. The mean level of UVB radiation in Irkutsk changed from $2-3$ mW m^{-2} nm^{-1} in December up to 50 mW m^{-2} nm^{-1} in June. The seasonal patterns of UVB radiation were principally different for both stations. In spring (March, April) the level of UVB radiation in Hyytiälä was higher by several times than in Irkutsk. In other months, especially in autumn, the picture was opposite: the level of radiation in Irkutsk was much higher than that in Hyytiälä. In other words, the non–symmetry of the seasonal patterns of UVB

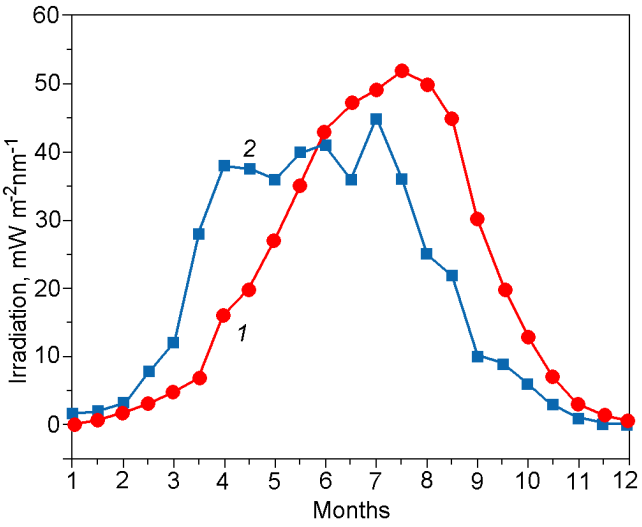

Figure 3. Seasonal behavior of monthly mean of UVB radiation intensity at λ =310 nm in the city of Irkutsk (1) and in Hyytiälä (2).

radiation at these both stations was principally different: the spring level of UVB radiation in Hyytiälä was much higher than that in August–September. A different picture was observed in Irkutsk: here the autumn UVB radiation level exceeded by 2–3 times its spring value. Asymmetry in the seasonal pattern of UVB in Eastern Siberia [*Mikhalev et al.,* 2001] was explained by a clearly expressed seasonal pattern of the total ozone content with a maximum in spring and the meteorological specifics of the region defining the cloudness and the aerosol background. Accounting for the high correlation between the formation rate of aerosol and UVB radiation one can expect the different seasonal patterns of the particle formation rates at these two stations.

The length of light day is also different for both stations. In winter the light-day in Lisvyanka is longer by three hours than in Hyytiälä. In summer the light day is longer in Hyytiälä by two hours.

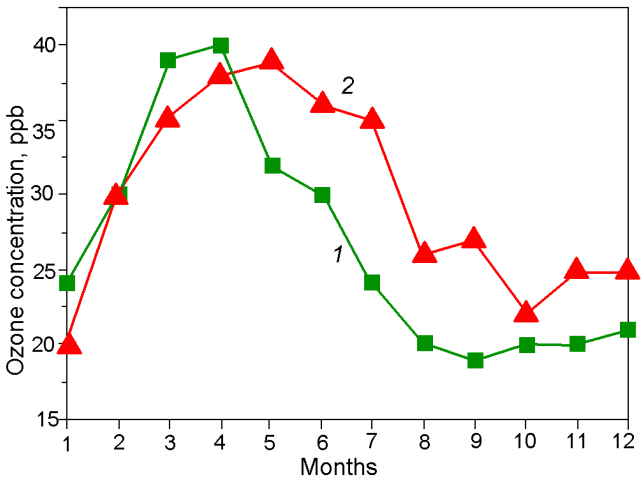

Figure 4. Seasonal behavior of monthly mean of ozone concentration at Bol'shie Koty (1) and at Hyytiälä (2).

Ozone

Tropospheric ozone plays an important role in the particle formation processes [e.g., *Janson et al.*, 2001; *Lyubovtseva et al.*, 2005 and extensive references therein]. First of all, ozone is a major source of the hydroxyl radical OH, which together with ozone are the primary oxidants of trace gases in the troposphere. As the oxidation is the main pathway for production of nucleating vapors and initiation of the chemical sinks of atmospheric trace gases, OH and ozone play an indirect but crucial role in determining the oxidation potential of the atmosphere and the lifetime of many trace gases. Because of absence of available data on ozone in Listvyanka in 2005 we used the data on ozone from the station Bol'shie Koty (30 km northward).

The seasonal cycle of O_3 at Bol'shie Koty station showed maxima in spring (up to 40 ppb, see Figure 4). After the spring maximum the concentration dropped down until August and then remained almost constant ($\approx$ 20 ppb).

At Hyytiälä station the maximal ozone concentration 40 ppb was observed in April and May. The minimal ozone (20 ppb) was observed in October and January. Beginning from April the ozone concentration in Bol'shie Koty was lower than that in Hyytiälä.

Concentration of Inorganic Trace Gases SO_2, NH_3, HNO_3

SO_2 The measurements of 2005 in Listvyanka showed that the weekly mean concentration of SO_2 reached 7–28 ppb in winter (Figure 5a) and 1–4 ppb in summer. In winter 2005 the highest concentrations of SO_2 in Listvyanka were observed at the coldest time when the temperature was below mean winter value by 5–7°. For example, the monthly mean concentration of SO_2 in 2003–2004 was 7 ppb in winter and 1.4 ppb in summer.

Seasonal pattern of SO_2 in Listvyanka practically coincided with that in Hyytiälä. The principal difference was the high level of SO_2 in Listvyanka. In winter the influence of local anthropogenic sources in Listvyanka becomes considerable, because of the stove heating and weak scattering of pollution in the inversely stratified atmosphere [*Khodzher et al.,* 1997]. In summertime the anthropogenic loading is related to the releases from Irkutsk and Baikalsk. For comparison, in the background region of Siberian boreal forests (Mondy) the monthly mean concentration of SO_2 was much lower: 0.9–1.7 ppb in winter and 0.3–0.6 ppb in summer. These concentrations are comparable to those in Hyytiälä.

The monthly mean concentrations of SO_2 at the Hyytiälä station according to the data of *Lyubovtseva et al.* [2005] were 0.5–1.3 ppb in winter and 0.1–0.4 ppb in summer and spring. The minimal concentration of SO_2 was observed in summer.

NH_3 During 2005 the NH_3 concentration in Listvianka changed from 0.1 ppb to 2.6 ppb (Figure 5b). Two maxima are clearly seen in the annual pattern, the summer one, where the concentration in August reached 1.3 ppb and the autumn maximum with the concentration 1.3–2.6 ppb. In the background conditions of Siberia the summer maximum in the NH_3 concentration in Mondy ($[NH_3] = 2$ ppb) are explained by the natural sources [*Khodzher et al.,* 1997]. In winter this concentration was much lower (0.2 ppb). Existing sporadic measurements of NH_3 in Hyytiälä in 1999 and 2003 [*Janson et al.,* 2001, 2005] showed that the concentration of NH_3 changed from 0.005 ppb to 0.4 ppb in spring with the average 0.049 ppb and from 0.01 ppb to 0.43 ppb with the average 0.05 ppb in summer. In general, the low concentration of NH_3 observed at low temperatures and in presence of the North air masses. Higher concentrations were observed when the air masses came from South. In Hyytiälä ammonia showed strong diurnal variations with concentration minima in the early morning and a maximum at the late afternoon.

The analysis of the aerosol samples from Listvyanka in the warm period of year showed the increase by factor of 2 in the molar concentration of NH_4^+ over SO_4^{2-} [*Golobokova et al.*, 2005]. Most likely that this is related to the fact that

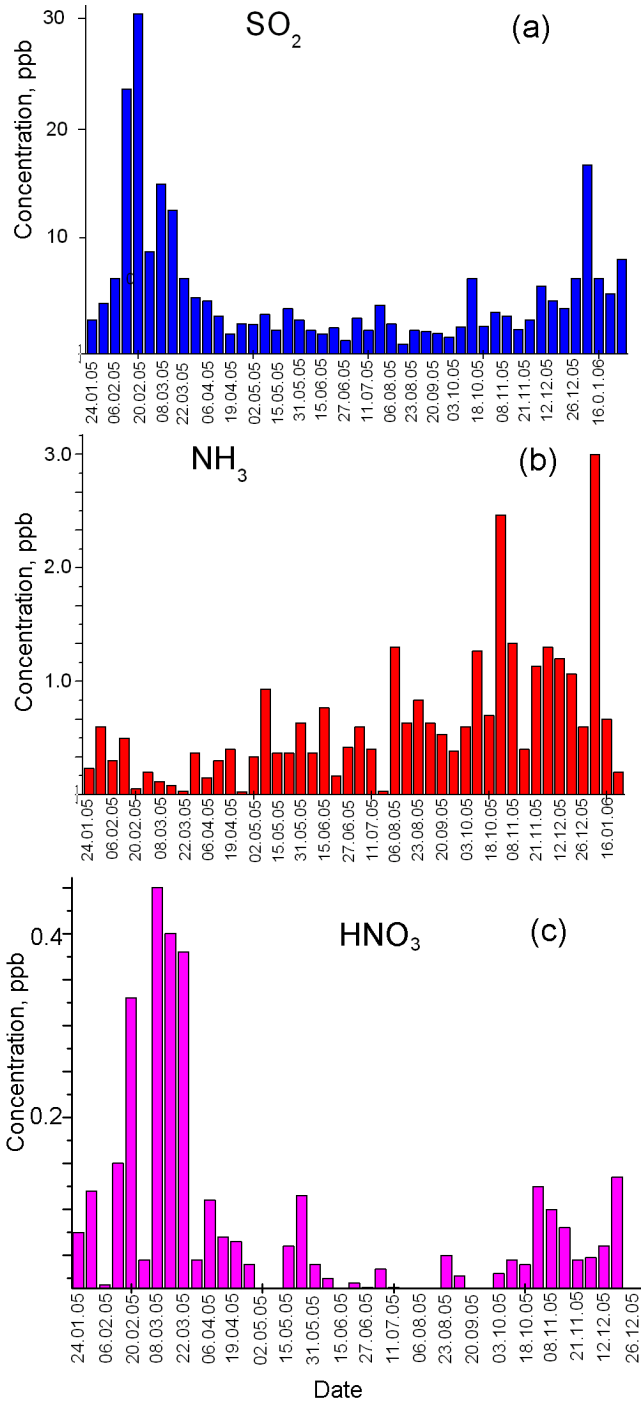

Figure 5. Weekly average variations of concentration of SO_2 (a), NH_3 (b), and HNO_3 (c) in Listvyanka in 2005 year.

the sulfates exist in entirely neutralized form $(NH_4)_2SO_4$. In cold periods these concentrations were approximately equal, i.e., the sulfates are not entirely neutralized and exist in various forms: e.g. H_2SO_4, $(NH_4)HSO_4$, $(NH_4)_2SO_4$.

HNO_3 The seasonal pattern of HNO_3 in Listvyanka practically repeated that of SO_2 (Figure 5c). The concentration of HNO_3 reached its maximum (0.5 ppb) in February and March. The minimal level of HNO_3 was observed in summertime. The data on HNO_3 for Hyytiälä are practically absent. We found the measurements of HNO_3 at Hyytiälä station in 1999 [*Janson et al.,* 2001]. According to this work HNO_3 varied from 0.01 ppb to 0.3 ppb with an average of 0.07 ppb in the summer period and from 0.01 to 1 ppb with an average 0.13 ppb during spring-99 campaign at Hyytiälä station. The lowest concentration was normally observed in early in the morning. The average concentration of HNO_3 in event and nonevent days showed no significant differences. Higher concentrations of HNO_3 were observed during springtime in nonevent periods.

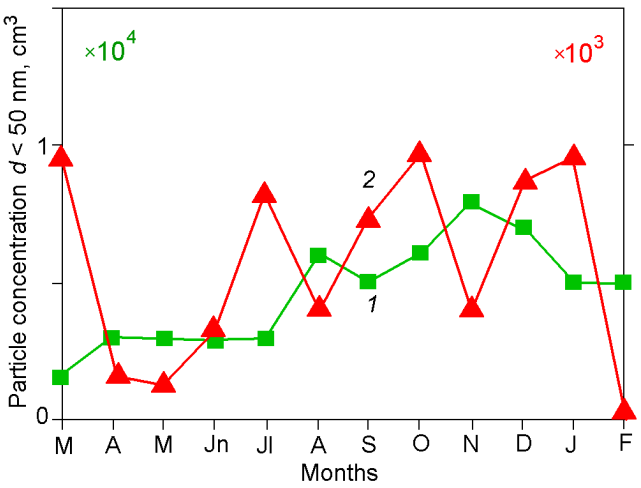

Figure 6. Time series of measured total concentration in Listvyanka and Hyytiälä (03.2005–03.2006). curve 1 shows the total number concentration of particle with $d < 50$ nm in Listvyanka, curve 2 displays the monthly mean number concentrations of particles $d < 50$ nm in Hyytiälä. (Listvianka – left axis, Hyytiälä – right axis).

Aerosol

Figure 6 shows the time series of the total particle concentrations ($d < 50$ nm) measured at the Listvyanka and the Hyytiälä sites during period March 2005–March 2006. The concentration of nucleation mode particles in Listvyanka grew beginning from July and reached the maximum in November 0.8×10^4 cm^{-3}. Similar maximum was also observed in the concentration of soluble ions NH_4^+ and SO_4^{2-} in November [*Golobokova et al.,* 2005].

We compared the seasonal patterns of total particle concentration ($d < 50$ nm) in Listvyanka for the whole period with the seasonal patterns of particle concentration of nucleation mode measured at the same period in Hyytiälä. In Listvyanka the mean particle concentration was higher by 3–10 times during the whole annual period. The minimal concentration of the nucleation mode was observed in April and May, in contrast to typical for Hyytiälä patterns with

the maximum concentration at springtime.

The seasonal dependencies of formation rate and growth rate for two stations, Listvyanka and Hyytiälä are presented in Figure 7 and Figure 8. The data of March 2005–March 2006 were used for the analysis of GR and FR in Listvyanka. The mean values of FR and GR are presented on the ground of the analysis of long term measurements performed in 1997–2005 by *Dal Maso et al.* [2005]. In spring the mean value of FR in Listvyanka was lower by 2–4 times than that in Hyytiälä. In contrast to Hyytiälä, where the maximal FR was observed in spring, the maximal FR in Lisvyanka was observed in summer. The GRs had well observed maxima at summertime at the both stations, the mean GR at Listvyanka being lower than at Hyytiälä during almost all year.

At present time it is commonly accepted that just formed particles have the size of order 1.5–1.8 nm *Kulmala et al.* [2006], whereas the particle size measurements overlap the size interval $d > 3$ nm. We thus never observe actual particle formation rate. Instead, the measurement give the values of formation rates at particle sizes 3 nm. *Kerminen and Kulmala* [2002] suggested an interpolation formula linking the formation rate FR(3 nm) with FR(1.5 nm). Recently *Lehtinen et al.* [2007] modified this formula for a more realistic form of the coagulation sinks. The modified Kerminen-Kulmala equation claims:

$$\mathrm{FR}_x = \mathrm{FR}_{1.5} \exp\left(-\gamma d_{1.5} \frac{\mathrm{CoagS}(d_{1.5})}{\mathrm{GR}}\right) \quad (1)$$

where FR_x=FR(x nm) is the "apparent" particle formation rate at $d = d_x$, GR is the particle growth rate,

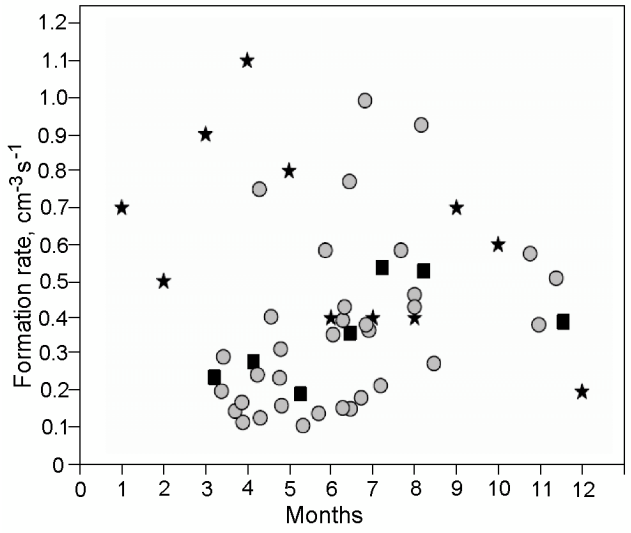

Figure 7. Seasonal behavior of particle formation rate (FR) in Listvyanka and Hyytiälä. Circles — FR in Listvyanka, squares — monthly mean FR in Listvyanka, stars — monthly mean FR in Hyytiälä [*Dal Maso et al.,* 2005].

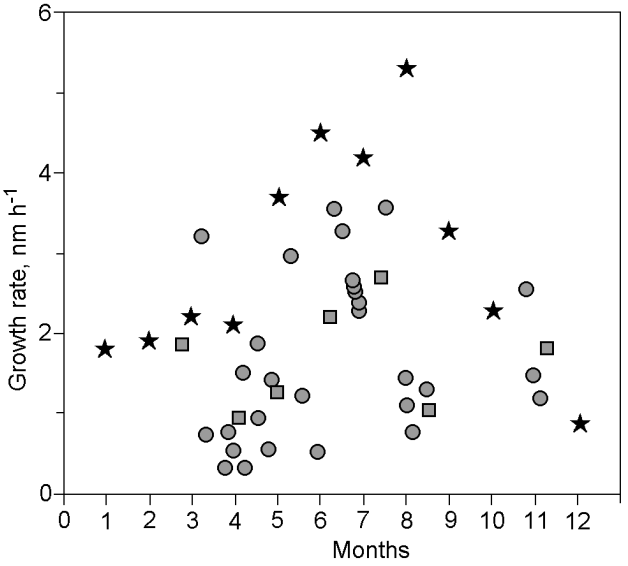

Figure 8. Seasonal behavior of particle growth rate (GR) in Listvyanka and in Hyytiälä. Circles — GR in Listvyanka, squares — monthly mean GR in Listvyanka, stars — monthly mean GR in Hyytiälä [*Dal Maso et al.,* 2005].

$$\gamma = \frac{1}{m+1}\left[\left(\frac{d_x}{d_{1.5}}\right)^{m+1} - 1\right] \quad (2)$$

In our calculation we used $m = -1.8$.

The coagulation sink is defined as the rate of the losses of freshly formed particles due to their intermode coagulation with preexisting aerosol particles of larger sizes,

$$\mathrm{CoagS}(d_p) = \int \beta(d_p, \bar{d}_p) n(\bar{d}_p) d\bar{d}_p \quad (3)$$

with $\beta(d_p, \bar{d}_p)$ being the collision frequency of particles of diameters d_p and $\bar{d}_p$, $n(\bar{d}_p)$ – the size distribution of preexisting particles, and

$$m = \frac{\ln[\mathrm{CoagS}(d_x)/\mathrm{CoagS}(d_{1.5})]}{\ln(d_x/d_{1.5})} \quad (4)$$

is the exponent in the size dependence of the coagulation sinks,

$$\mathrm{CoagS}(d_x) = \mathrm{CoagS}(d_{1.5}) \left(\frac{d_p}{d_{1.5}}\right)^m \quad (5)$$

The parameter m varies from -2 to -1, depending on the size distribution of preexisting particles. Coagulation sink is expressed through the condensation sink as follows:

$$\mathrm{CoagS}(1.5\,\mathrm{nm}) = 0.578\mathrm{CS} \quad (6)$$

[*Lehtinen et al.*, 2007].

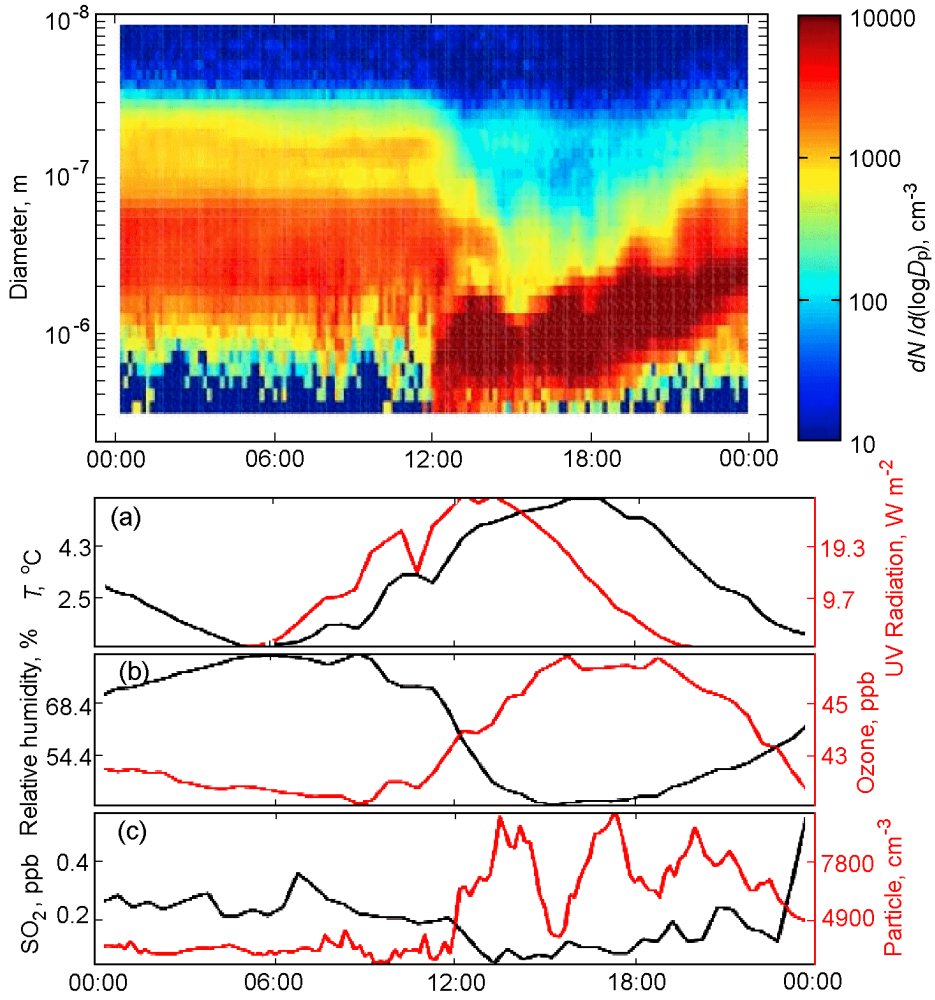

Figure 9. An event of new particle formation observed in Hyytiälä 06.04.2005. Top panel shows the diurnal evolution of the particle size distribution. Bottom panel: (a) diurnal evolution of temperature and UV radiation; (b) O_3 mixing ratio (ppb) and relative humidity; (c) total particle concentration ($3-50$ nm) and SO_2 mixing ratio (ppb).

We applied (1) for estimating the ratio of real formation rate to apparent formation rate, $FR_{1.5}/FR_3$ in Lisvyanka and Hyytiälä. For typical winter values of CS= 0.001 s^{-1}, GR= 1.8 nm h^{-1} (Hyytiälä) and CS= 0.002 s^{-1}, GR= 1 nm h^{-1} (Listvyanka) we found $FR_{1.5}/FR_3$ ≈2.5 for Hyytiälä and $FR_{1.5}/FR_3 \approx 28.5$ for Listvyanka. In summer we found respectively $FR_{1.5}/FR_3 \approx 2.5$ for Hyytiälä (CS= 0.0025 s^{-1}, GR= 4.5 nm h^{-1}), and $FR_{1.5}/FR_3 \approx 12.3$ for Listvyanka (CS= 0.0037 s^{-1}, GR= 2.5 nm h^{-1}). The values of CS and GR were taken from [*Dal Maso et al.,* 2005, 2008; *Zagaynov et al.,* 2007].

Nucleation Bursts

As an example let us consider two typical aerosol events in Hyytiälä and Listvyanka (Figure 9 and Figure 10). In Hyytiälä (Figure 9) 06.04.2005 the aerosol formation process began at about 12 p.m. The relative humidity during several hours (from 0 to 8 p.m.) remained almost constant $RH = 40-50\%$. The SO_2 concentration was low (0–0.2 ppb). The ozone concentration raised up to 47 ppb when the particle concentration reached its maximum. The total particle concentration grew by factor of 3. (from 3×10^3 to 10^4 cm^{-3}). The particle growth continued several hours and was accompanied with the conversion of the nucleation mode particles to the Aitken mode. According to *Ehn et al.* [2007] the particle solubility decreased in increasing the modal particle size for this event day. This fact indicates that the initial growth was due to the presence of more hygroscopic compounds, whereas the subsequent growth was caused mainly by less hygroscopic or even hydrophobic compounds. Hence, the species responsible for growth of freshly nucleated particles were different from those participating in nucleation. A number of works [e.g., *Boy et al.,* 2005;

Kulmala et al., 2004; *Sihto et al.,* 2006; *Weber et al.,* 1997] emphasized that although sulfuric acid, a very hygroscopic compound, is one of the most likely candidates responsible for formation of critical clusters, sulfur chemical cycle does not produce enough sulfuric acid in the atmosphere to explain the particle growth up to 20–100 nm in diameter. According to *Boy et al.* [2005], the contribution of H_2SO_4 to particle growth from 3 to 25 nm in Hyytiälä was about 10–12%. Respectively, the further particle growth was connected with other nonvolatile compounds, e.g., of organic origin [*Kerminen et al.,* 2000].

The estimates by *Lyubovtseva et al.* [2005] of the concentrations of monoterpenes oxidation products by OH and ozone showed that in summertime their monthly mean concentrations (10^8 cm^{-3}) exceeded [H_2SO_4] by 15–20 times for event days and were enough for explaining the observed GR in Hyytiälä (up to 10 nm h^{-1}). Besides, it was shown that the seasonal patterns of monoterpenes oxidation products correlated well with the seasonal pattern of GR in Hyytiälä.

In Listvyanka 07.07.2005 (Figure 10) the concentration of newly born aerosol particles during the nucleation burst grew by five times up to 6×10^3 cm^{-3}. The increase in the concentration of these new particles correlated well with the dynamics of SO_2 concentration whose level exceeded by 20 times that in Hyytiälä. We stress once more that the formation process and the particle growth in Listvyanka went at high level of SO_2 typical for urban places, and high relative humidity (RH= 70 − 100%) The measurements of NH_3 for this day are absent. Nevertheless, according to the data of weekly mean concentration of NH_3 shown in Figure 5b we can conclude that the NH_3 concentration was of order 0.3–0.5 ppb.

Let us estimate the ratio of sulfuric acid production rates in Listvyanka and Hyytiälä. The production rate of sulfuric acid is expressed as

$$Q(H_2SO_4) = k_1[SO_2][OH] \quad (7)$$

where k_1 is the rate constant for the reaction $SO_2 + OH \longrightarrow H_2SO_4$. Then the sulfuric acid concentration is

$$[H_2SO_4] = \frac{Q(H_2SO_4)}{CS} = \frac{k_1[SO_2][OH]}{CS} \quad (8)$$

Here CS stands for the condensation sink for the molecules of sulfuric acid [*Kulmala et al.,* 2006].

The contribution of sulfuric acid to the growth of newly born particles. is proportional to [H_2SO_4]. Then the ratio of sulfuric acid production rates in Listvyanka and Hyytiälä can be estimated as follows:

$$\frac{GR^L}{GR^H} = \frac{[SO_2]_L[OH]_L CS_H}{[SO_2]_H[OH]_H CS_L} \quad (9)$$

The subscripts L and H refer to Listvyanka and Hyytiälä respectively. It is important to emphasize that this ratio is independent of the reaction constants.

The concentration $[SO_2]_L$ exceeded by order of magnitude the respective value in Hyytiälä which gives us the grounds to assume that the particle formation and growth in Listvyanka was due to one and the same source (production of sulfuric acid) Indeed, the numerical estimate for $[OH]_L = 10^6 - 10^7$ cm^{-3} and $[SO_2]_L = 10^{11}$ cm^{-3} gives $[H_2SO_4]_L = 10^7 - 10^8$ cm^{-3} which is enough for the growth rate GR=0.6–3.5 nm h^{-1} observed in Listvyanka at RH= 80 − 100%.

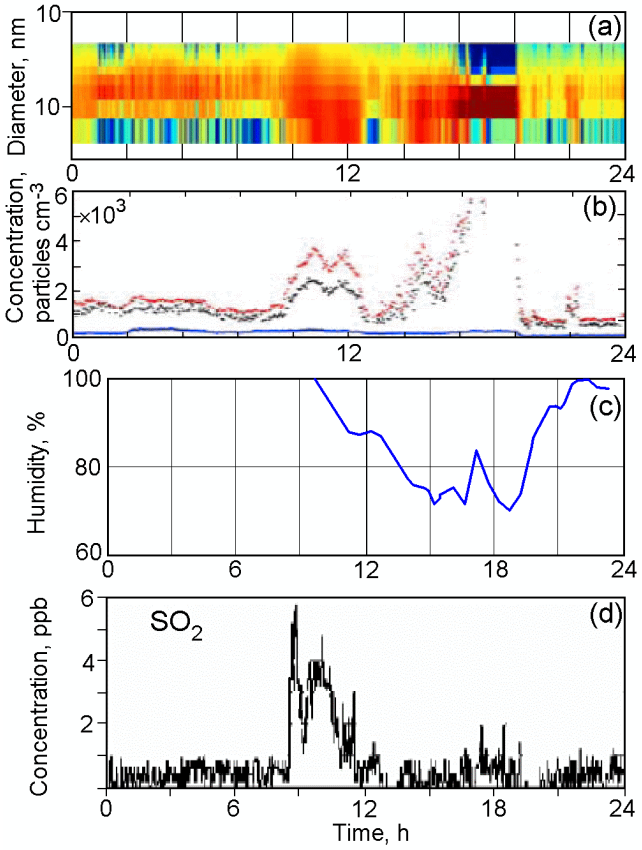

Figure 10. An event of new particle formation observed at Listvyanka 07.07.2005: (a) the diurnal evolution of the particle size distribution; (b) total particle concentration with $d < 50$ nm (red line), the concentration of particles with $d < 20$ nm (dotted blue line), and the concentration of submicron particles (solid blue line); (c) the diurnal evolution of relative humidity; (d) the SO_2 mixing ratio (ppb).

Our analysis thus supports the hypothesis that the particle formation–growth process in Listvyanka could be driven by sulfuric acid.

Summary and Conclusions

The observations by [*Dal Maso et al.*, 2008], demonstrated that the nucleation events in Listvyanka are more seldom phenomena than those in Central Finland. Moreover, the nucleation bursts over there were not so clearly expressed as it was observed at Hyytiälä station and other similar sites in

Northern and Central Europe. The explanation can be found in the fact that local mountains are mainly covered with larches which emit organic substances different from those emitted by Scott pines [*Khodzher et al.*, 1997; *Ruuskanen et al.*, 2007]. Next, in summertime the period of rains and mists in Siberia is much longer than in Finland which essentially influences the formation of aerosol particles.

Seasonal patterns of UVB radiation at $\lambda = 310$ nm are principally different for boreal forests around Hyytiälä and in Siberia. The non-symmetry of the seasonal patterns of UVB radiation in these both regions are different. The spring level of UVB radiation in Hyytiälä is much higher than the UVB radiation in August–September. A different picture is observed in Siberia, where the autumn UVB radiation level exceeds by 2–3 times its spring value. The high correlation between the aerosol formation rate and UVB radiation accounts for the different seasonal pattern of the formation rates for these two regions of boreal forest.

During all period of observation the concentration of nucleation mode in Listvyanka was higher than in Hyytiälä. Two main maxima in Listvyanka were observed in late summer and late autumn. The analysis of the Siberian data revealed seasonal correlations between the concentrations of nucleation mode and NH_3. The maximal formation rate in Listvyanka was also observed in summer and autumn and correlated well with UVB radiation. In spring the formation rate was a half or a third of that in summer. A comparison of the seasonal patterns of formation rates in Listvyanka and Hyytiälä displayed their considerable difference. In Hyytiälä the particle formation rate was maximal in springtime and minimal in midsummer, whereas the growth rate was maximal in summer and minimal in spring. The seasonal patterns of FR and GR in Listvyanka were similar and have simultaneous maxima in summer. This fact can be explained by common origin of the substances responsible for nucleation and the particle condensation growth.

The concentration of SO_2 in Listvyanka exceeded that in Hyytiälä by several times. High SO_2 concentrations presumably could lead to high concentrations of sulfuric acid vapor produced photochemically in the atmosphere. The growth rate of freshly nucleated particles is expected in this case to be related to the condensation of sulfuric acid. Since the concentration level of NH_3 was also high (0.5–2 ppb), a considerable contribution of ternary nucleation to the particle formation process could be expected [*Korhonen et al.,* 1999; *Napari et al.,* 2002]. In some extent this is confirmed by the correlations between the seasonal pattern of the nucleation mode and NH_3. High relative humidity (RH$>$ 80%) typical for most event days in Listvyanka allows us to assume that the growth process occurred on aqueous ammonium sulfate solution rather than on dry ammonium particle. According to *Stolzenberg et al.* [2005] and *Kulmala et al.* [2001b] in this case the particle growth rate enhanced at the same concentration of H_2SO_4. The data of chemical analysis of the aerosol samples [*Golobokova et al.,* 2005] confirmed that in summertime a stable twofold excess of NH_4^+ over SO_4^{2-} in aerosol particles was observed which means that the particles consisted of $(NH_4)_2SO_4$. In contrast to Listvyanka the formation process in Hyytiälä occurred at small relative humidity (40–50%) and low concentration of SO_2 during most event days [*Lyubovtseva et al.,* 2005].

The difference in the kinetics of nucleation bursts could be assumed to be related to different chemical pathways of the particle formation. As we mentioned the considerable amount of sulfur dioxide in the atmosphere of Listvyanka suggests that the sulfuric compounds contribute mainly to the particle formation and growth in contrast to Hyytiälä, where they give only 10–12% to the particle growth. In both these places the sulfuric compounds and, in particular, sulfuric acid are most likely responsible for the formation of supercritical embryos [*Kulmala et al.,* 2004, 2006]. The reaction of hydroxyl radical with sulfur dioxide leading to formation of sulfuric acid is more efficient in the conditions of Listvyanka, where this reactions go in presence of much higher content of sulfur dioxide.

Nucleation and subsequent growth in Hyytiälä follow a different pathway: sulfuric acid drives nucleation whereas growth is provided by nonvolatile organics, a product of photochemical processing of highly volatile plant emissions. Respectively, one expects that the particles of the Aitken mode produced by the nucleation bursts of different nature will have principally different chemical composition. The particles from boreal forest near Hyytiälä should comprise organic substances whereas the particles from Listvyanka are inorganic and comprise mainly ammonium sulfate. The climatic activity of above mentioned two types of aerosols is also very different. The sulfate particles are hygroscopic and thus are more active as CCN as compared to hydrophobic organic particles.

References

Aalto, P., et al. (2001), Physical characterization of aerosol particles during nucleation events, *Tellus, 53B,* 344–358.

Arshinov, M. Yu., B. D. Belan, D. V. Simonenkov (2006), Photochemical formation of microdisperse aerosol in the atmosphere of a continental region, *Opt. Atm. Ocean, 19,* 328–339.

Bashirova, V. S., et al. (1992), Measurements of atmospheric condensation nuclei size distribution in Siberia, *J. Aerosol Sci., 23,* 191–199.

Birmili, W., H. Beresheim, C. Plass-Dulmer, T. Elste, S. Gilge, A. Weidensohler, U. Uhrner (2003), The Hohenpeisenberg aerosol formation experiment (HAFEX): A long term study including size resolved aerosol, H_2SO_4, OH, and monoterpene measurements, *Atmos. Chem. Phys., 3,* 361–376.

Boy, M., M. Kulmala (2002), Nucleation events on the continental boundary layer: infuence of physical and meteorological parameters, *Atmos. Chem. Phys., 2,* 1–16.

Boy, M., U. Rannik, K. E. Lehtinen, V. Tarvainen, H. Hakola, M. Kulmala (2003), Nucleation events in the continental boundary layer: Long-term statistical analysis of aerosol relevant characteristics, *J. Geophys. Res., 108*(D21), 4667–4675. doi:10.1029/2003JD003838

Boy, M., et al. (2005), Sulfuric acid closure and contribution to nucleation mode particle growth, *Chem. Phys., 5,* 863–878.

Clement, C. F., L. Pirjola, M. Dal Maso, J. M. Mäkelä, M. Kulmala (2001), Analysis of particle formation bursts observed in Finland, *J. Aerosol Sci., 32,* 217–236. doi:10.1016/S0021-8502(00)00059-8

Dal Maso, M., M. Kulmala, I. Riipinen, R. Wagner, T. Hussein, P. P. Aalto, K. Lehtinen (2005), Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribu-

tion data from SMEAR II, Hyytiälä, *Finland Boreal Env. Res., 10,* 323–336.

Dal Maso, M., et al. (2008), Aerosol particle formation events at two Siberian stations inside the boreal forest, *Boreal Env. Res., 13,* 81–92.

Ehn, M., T. Petäjä, H. Aufmhoff, P. Aalto, K. Hämeri, E. Arnold, A. Laaksonen, M. Kulmala (2007), Hygroscopic properties of ultrafine aerosol particles in the boreal forest: diurnal variations, solubility and the influence of sulfuric acid, *Atmos. Chem. Phys., 7,* 211–222.

Golobokova, L. P., I. V. Latysheva, V. I. Mordvinov, T. V. Khodzher, V. A. Obolkin, V. L. Potemkin (2005), Peculiarities of the chemical content of the atmospheric aerosol at the extreme weather conditions of the Southern Siberia, *Optics Atmosph. Ocean, 18,* 688–693.

Hari, P., M. Kulmala (2005), Station for measuring ecosystem – atmosphere relations (SMEAR II), *Boreal Env. Res., 10,* 315–322.

Hyvönen, S., et al. (2005), A look at aerosol formation using data mining techniques, *Atmos. Chem. Phys., 5,* 3345–3356.

Janson, R., K. Rozman, K. Karlsson, H.C. Hansson (2003), Biogenic emission and gaseous precursor to forest aerosols, *Tellus, B53,* 423–440.

Janson, R., H. Wideqvist, H. Hakola, H. Lihavainen (2005), VOC, ammonia and organic carbon measurement during Hyytiälä campaign, March–April 2003, *Report series in aerosol science, 76 Quantification of aerosol nucleation in the European boundary layer,* 247–251, QUEST, Helsinki.

Julanov, Yu. V., A. A. Lushnikov, V. A. Zagaynov (2002), Diffusion aerosol spectrometer, *Atmospheric research, 62,* 295–302. doi:10.1016/S0169-8095(02)00015-7

Kerminen, V-M., A. Virkkula, R. Hillamo, A. S. Wexler, M. Kulmala (2000), Secondary organics and atmospheric cloud condensation nuclei production, *J. Geophys. Res., 105,* 9255–9264. doi:10.1029/1999JD901203

Kerminen, V-M., M. Kulmala (2002), Analytical formulae connecting the "real" and apparent nucleation rate in the nuclear number concentration for atmospheric nucleation events, *J. Aerosol Sci., 33,* 609–622. doi:10.1016/S0021-8502(01)00194-X

Khodzher, T. V., V. L. Potemkin, V. A. Obolkin (1994), Chemical composition of aerosol and trace gases in the atmosphere over Lake Baikal, *Optics Atmos. Ocean, 7,* 566–569.

Khodzher, T. V., L. P. Golobokova, V. A. Obolkin, V. L. Potemkin, O. G. Netsvetaeva (1997), Diurnal and seasonal variability of the ion composition of atmospheric aerosol at the Southern part of Eastern Siberia, *Optics Atmos. Ocean, 10,* 650–655.

Korhonen, P., M. Kulmala, A. Laaksonen, Y. Viisanen, R. McGraw, J. H. Seinfeld (1999), Ternary nucleation of H_2SO_4, NH_3 and H_2O in the atmosphere, *J. Geophys. Res., 104,* 26,349–26,353. doi:10.1029/1999JD900784

Kulmala, M., et al. (2001a), Overview of the International project on biogenic aerosol formation in the boreal forest (BIOFOR), *Tellus, 53B,* 324–343.

Kulmala, M., M. Dal Maso, J. M. Mäkelä, L. Pirjola, M. Väkevä, P. Aalto, P. Miikulainen, K. Hämeri, C. D. O'Dowd (2001b), On the formation, growth and composition of nucleation mode particles, *Tellus, 53B,* 479–490.

Kulmala, M., H. Vehkämäki, T. Petaja, M. Dal Maso, A. Lauri, V. M. Kerminen, W. Birmilli, P. H. McMurry (2004), Formation and growth rates of ultrafine atmospheric particles: review of observations, *J. Aerosol. Sci., 35,* 143–176. doi:10.1016/j.jaerosci.2003.10.003

Kulmala, M., K. E. J. Lehtinen, L. Laaksonen (2006), Cluster activation theory as an explanation of the linear dependence between formation rate of 3 nm particles and sulphuric acid concentration, *Atmos. Chem. Phys., 6,* 787–793.

Koutsenogii, P., R. Jaenicke (1994), Number concentration and size distribution of atmospheric aerosol in Siberia, *J. Aerosol Sci., 25,* 377–383. doi:10.1016/0021-8502(94)90088-4

Lehtinen, K. E. J., M. Dal Maso, M. Kulmala, V–M. Kerminen (2007), Estimating nucleation rates from apparent particle formation rates and vice versa: Revised formulation of the Kerminen–Kulmala equation, *J. Aerosol Sci., 38,* 988–994. doi:10.1016/j.jaerosci.2007.06.009

Lyubovtseva, Yu. S., L. Sogacheva, M. Dal Maso, B. Bonn, P. Keronen, M. Kulmala (2005), Seasonal variations of trace gases, meteorological parameters and formation of aerosols in boreal forests, *Boreal Env. Res., 10,* 493–510.

Mikhalev, A. V., M. A. Chernigovskaya, A. Y. Shalin, E. S. Kazimirovskii (2001), Variations of the ground level ultraviolet radiation in East Siberia, *Adv. Space Res., 27,* 1109–1114. doi:10.1016/S0273-1177(01)00181-8

Mäkelä, J., I. Koponen, P. Aalto, M. Kulmala (2000a), One year data of submicron size modes of tropospheric background aerosol in Southern Finland, *J. Aerosol Sci., 31,* 595–611.

Mäkelä, J., M. Dal Maso, L. Pirjola, P. Keronen, L. Laakso, M. Kulmala, A. Laaksonen (2000b), Characteristics of the atmospheric particle formation events observed at a boreal forest site in Southern Finland, *Boreal Environ. Res., 5,* 299–313.

Napari, I., M. Noppel, H. Vekhamäki, M. Kulmala (2002), An improved model for ternary nucleation of sulfuric acid–ammonia–water, *J. Chem Phys., 117,* 8418–8425.

Pochanard, P., H. Akimoto, Y. Kajii, V. M. Potyomkin, T. V. Khodzher (2003), Regional background ozone and carbon monoxide variations in remote in Siberia/East Asia, *J. Geophys. Res., 108*(D1), 4028. doi:10.1029/2001JD001412

Poteyomkin, V. L., E. V. Shul'gin (2004), Seasonal dynamics of ozone concentration over Eastern Sayan mountain ridge, *Opt. Atmos. Ocean, 17,* 317–320.

Ruuskanen, T., H. Hakola, M. K. Kajos, H. Hellen, V. Tarvainen, J. Rinne (2007), Volatile organic compound emissions from Siberian larch, *Atmos. Env., 41,* 5807–5812. doi:10.1016/j.atmosenv.2007.05.036

Sihto, S.-L., et al. (2006), Atmospheric sulphuric acid and aerosol formation: implications from atmospheric measurements for nucleation and early growth mechanisms, *Atmos. Chem. Phys., 6,* 4079–4091.

Stolzenburg, M., P. McMurry, H. Sakurai (2005), Growth rates of freshly nucleated atmospheric particles in Atlanta, *J. Geophys. Res., 110*(D22S05), doi:10.1029/2005JD005935

Tunved, P., et al. (2003), One year boundary layer aerosol size distribution from five Nordic background stations, *Atm. Chem. Phys., 3,* 2183–2205.

Tunved, P., et al. (2006), High natural aerosol loading over boreal forests, *Science, 312,* 261–263. doi:10.1126/science.1123052

Vehkamki, H., et al. (2004), Atmospheric particle formation events at Varrio measurement station in Finnish Lapland. 1998–2002, *Atmos. Chem. Phys., 4,* 2015–2023.

Weber, R. J., J. J. Marti, P. H. McMurry, F. L. Eisele, D. J. Tanner, A. Jeferson (1997), Measurement of new particle formation and ultrafine particle growth rate at a clean continental site, *J. Geophys. Res., 102,* 4375–4385. doi:10.1029/96JD03656

Zagaynov, V. A., A. A. Lushnikov, O. N. Nikitin, P. E. Kravchenko, T. V. Khodzher, I. V. Petryanov–Sokolov (1989), Background aerosol over lake of Baikal, *Dokl. Akad Nauk SSSR, 308,* 1087–1090.

Zagaynov, V. A., M. Kulmala, Yu. S. Lyubovtseva, A. A. Lushnikov, L. Sogacheva, T. V. Khodzher (2006), Nucleation bursts in the atmosphere of Central Siberia, *Report Series in Aerosol Science, 83,* 419–423.

M. Boy, M. Dal Maso, H. Junninen, and M. Kulmala, Department of Physical Sciences, University of Helsinki, P.O. Box 64. FIN-00014, Helsinki, Finland

Yu. S. Lyubovtseva, Geophysical Center of RAS, Molodezhnaya, 3, Moscow, Russia. (u.lubovtseva@gcras.ru)

Yu. G. Biryukov, A. A. Lushnikov, and V. A. Zagaynov, Karpov Institute of Physical Chemistry, 10, Vorontsovo Pole, 105064 Moscow, Russia

T. V. Khodzher, V. A. Obolkin, and V. L. Potyomkin, Limnological Institute of RAS, 3, Ulan-Batorskaya ul., Irkutsk, Russia