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The atmospheric aerosol over Siberia, as seen from the 300 m ZOTTO tower

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ABSTRACT

This report describes a unique setup for aerosol measurements at the new long-term Tall Tower monitoring facility near Zotino, Siberia (ZOTTO). Through two inlets at 50 and 300 m aerosol particle number size distributions are measured since September 2006 in the size range 15–835 nanometer dry diameter. Until the end of May 2007 total number (N_{300}) concentrations at 300 m height ranged between 400 cm^{-3} (5%) and 4000 cm^{-3} (95%) with a median of 1200 cm^{-3}, which is rather high for a nearly uninhabited boreal forest region during the low productivity period of the year.

Fitting 1-h average distributions with a maximum of four lognormal functions yielded frequent ultrafine modes below 20 nm at 50 m height than at 300 m, whereas the latter height more frequently showed an aged nucleation mode near 30 nm. The positions of Aitken (≈80 nm) and accumulation modes (≈210 nm) were very similar at both inlet heights, the very sharp latter one being the most frequent of all modes. The encouraging first results let us expect exciting new findings during the summer period with frequent forest fires and secondary particle sources from vegetation emissions.

1. Introduction

Since the seminal papers by Charlson et al. (1990, 1991) research on the global distribution of atmospheric aerosol particles and on their effects on the budget of incoming solar radiation has intensified. Indications of direct and indirect aerosol effects on atmospheric chemistry (Lelieveld and Crutzen, 1990; Ravishankara, 1997), and human health (WHO, 2002) also increased atmospheric aerosol research. Over the past 10 yr the reports by the Intergovernmental Panel on Climate Change (IPCC) have stressed the need to understand the past and possible future influence of aerosols on climate because of anthropogenic activities during the period of industrialization.

Aerosol-driven atmospheric effects are evaluated using global circulation models (e.g. Kinne et al., 2003) satellite-borne observation systems (Ichoku et al., 2002; Kaufman et al., 2002), ground-based observation networks using integrated column (Dubovik et al., 2002) or ground-based LIDAR measurements (Bösenberg et al., 2003), and in situ observation networks (WMO/GAW, 2003).

In situ aerosol observations are especially useful for studying physical, chemical and optical effects of aerosol properties within a confined volume of air and as necessary complement to remote sensing approaches. In situ aerosol studies may address the following general issues. First, they allow the quantification of aerosol-optical and cloud-forming properties as a function of the physical and chemical particles properties, such as number concentration, number size distribution, particle shape, chemical composition as well as their ability to absorb water (e.g. Fiebig et al., 2003). Second, in situ studies allow to establish connections between observed aerosol characteristics and potential source regions through the analysis of meteorological transport, notably back trajectories (e.g. Jaffe et al., 1999; Dal Maso et al., 2007) and retroplumes (Stohl et al., 2003). Third, in situ observations are an essential basis to validate the predictions of prognostic aerosol models based on chemistry transport models (Charlson, 2000).

Siberia, as large parts of Asia in general, has only received limited attention with respect to the verification of aerosol-driven climate effects. The resulting gap of knowledge is in sharp contrast to the obvious global relevance of Siberia with respect to constituent exchange between the biosphere and the atmosphere (IGBP, 2007). Notably, the Siberian biosphere is a seasonally dependent source and sink of biogenic carbon species (Schulze

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et al., 1999; Lloyd et al., 2002), and has been acknowledged to be one of the regions representing the largest uncertainties within the context of global carbon balance (Schimel et al., 2001).

Importantly, Siberia has widely been documented to be a source region of biomass burning aerosols. Aerosol particles released by Siberian forest fires could be tracked down many thousand kilometres downwind within the entire Northern hemisphere (e.g. Cahoon et al., 1994; Stocks et al., 1998; Tanimoto et al., 2000; Damoah et al., 2004; Müller et al., 2005). Furthermore, boreal forests have been shown to represent a large-scale source of organic secondary aerosols (e.g. Tunved et al., 2006). The lack of knowledge regarding secondary aerosol source strengths, however, propagates into the poorly represented organic aerosol species in global climate models. Since the ecological system in the northern boreal forests is sensitive to global temperature chance, it will also be a challenge in examine the role of a changing Siberia within the global context of primary and secondary aerosol production as part of the task: "to develop [. . .] predictive capability of terrestrial ecosystems dynamics over Northern Eurasia for the 21st century to support global projections as well as informed decision making and numerous practical applications in the region" (Groisman et al., 2007).

The present report introduces the study climatically relevant atmospheric AEROsols over SIBeria (AEROSIB), the objective of which is to examine the occurrence and the source regions of the main aerosol types over Siberia and adjacent parts of Asia, and their effects upon the balance of atmospheric aerosol particles including number and mass. The related experiment is based on the new 300-m tower of the Zotino Tall Tower facility (ZOTTO) established by the Max-Planck-Institute for Biogeochemical Cycles, Jena. After a description of the unique instrumental set up and its potential for monitoring large parts of Asia, data on number size distributions taken during the time period September 2006–May 2007 will be discussed.

2. Instrumental

2.1. The Zotino Tall Tower Facility (ZOTTO)

Since 2006, a unique research platform is available in Central Siberia as the ZOTTO at 60.8°N; 89.35°E, about 20 km west of the Yenisey river. The site lies in a vast region of forests and bogs, still relatively undisturbed by anthropogenic influences and relatively inhospitable because of its continental climate. The population density is low but a moderate human impact on vegetation, for example, by logging activities, is already visible. The climate is dominated by a large seasonal temperature cycle reaching from minima below −55 °C in winter to maxima above 30 °C in summer. The tower is 300 m high and was designed for long-term atmospheric observations in Siberia that are representative for a very large spatial area. A significant benefit of tall tower sampling is also that measurements aloft are less prone to contamination by unavoidable local sources such as the power generator of the facility.

Tower and the adjacent underground laboratory were constructed between 2004 and 2006 by the Max-Planck-Society (http://www.bgc-jena.mpg.de/bgc-systems/projects/zotto/index.shtml). The scientific leader of the ZOTTO facility is the Max-Planck-Institute for Biogeochemistry in Jena, Germany, together with the Max-Planck-Institute for Chemistry, Mainz, Germany and the VN Sukachev Institute of Forest, Krasnoyarsk, Russia as additional consortium members. The VN Sukachev Institute of Forest continually operates and maintains the ZOTTO facility.

Trace gas measurements obtained by aircraft in summer in the Zotino area (Lloyd et al., 2002) indicated a well-mixed planetary boundary layer by the depletion of carbon dioxide concentration as a result of daytime photosynthesis. This measurement shows that a 300 m sampling point on the tower should be largely representative for boundary layer air, thereby avoiding effects that are related to the shallow surface layer, indicated by an excess in biogenic oxygen isotopes.

2.2. The aerosol inlet at ZOTTO

To collect ambient aerosol from a height of 300 m, a sampling pipe of the corresponding length was erected at ZOTTO in August and September 2006. There was no alternative to a solid inlet tube at ZOTTO because sensitive aerosol particle instrumentation, such as particle size spectrometers cannot be operated on the 300 m tower under the harsh Siberian climatic conditions, nor could such exposed devices be regularly maintained. By using a long inlet ambient aerosols can be conducted into an underground laboratory container where they can be analysed in a temperature-controlled climate.

The details of the current set-up at ZOTTO as well as the inlet calibration for particle losses are described in a specialized paper (Birmili et al., 2007). Briefly, the aerosol inlet pipe constructed for ZOTTO is a 300 m stainless steel pipe with an internal diameter of about 2.8 cm and was designed for a laminar nominal sampling flow of 40 l min^{-1}. The main features of the inlet pipe are: Resistance against the harsh climatic conditions, smooth inner surface, laminar flow and division into 100 sections (3 m each) to facilitate transport and assembly, smoothly joined by modified Swagelok-type fittings. For aerosol sampling at the two levels of 50 and 300 m two corresponding independent inlets of the same construction were mounted.

Before deployment at ZOTTO, the 300 m inlet pipe was assembled horizontally outdoors at IfT, Leipzig and calibrated with ambient aerosol particles for size-dependent particle transport losses (Birmili et al., 2007). The calibration showed that particles with diameter (D_p) > 50 nm are nearly perfectly transmitted through the pipe, and that 10 nm particles are still transmitted at a reproducible efficiency of 20%. The calibration and the related numerical calculations were generally used in this paper to

correct all particle size distributions between 15 and 900 nm for the transmission losses occurring in the sampling pipes.

For aerosol sampling at height levels of 50 and 300 m two aerosol independent inlets of the same construction were mounted in August/September 2006 at ZOTTO. A computer-controlled valve system switches the aerosol sampling between the two levels every five minutes while maintaining the nominal 40 l min^{-1} flow in both inlets.

2.3. Size distribution measurements

Aerosol particle number size distributions are counted in 18 logarithmically equal size bins between 15 and 835 nm dry particle diameter by a Differential Mobility Particle Sizer (DMPS). The basic principles of this instrument are described in Knutson and Whitby (1975), Winklmayr et al. (1991) and Birmili et al. (1999). After multiple charge inversion, size distributions with 18 logarithmically equal size bins between 15 and 835 nm dry particle diameter were obtained. This particle size range encompasses the accumulation mode (100–835 nm), representative for aged atmospheric aerosols, the Aitken mode (50–100 nm)—representative for young aerosol particles only a few days old, and the nucleation mode (15–50 nm), representing the most recently formed aerosol particles.

To meet the special requirements of long-term operation at a remote site like ZOTTO, the following specifics of the DMPS system had to be optimized before installation: automatic operation, low level of maintenance, long-term stability, avoidance of radioactive sources as a charging device. The ZOTTO-DMPS uses, as a Differential Mobility Analyser (DMA), a Hauke type DMA with a centre rod length of 28 cm (Winklmayr et al., 1991). Particles downstream of the DMA are counted using a laminar flow condensation particle counter (model 3762, TSI Inc., St. Paul, USA). The sample flow through the particle counter is limited by a critical orifice to 0.78 l min^{-1}. The sheath airflow of 5.0 l min^{-1} is circulated in a closed loop, regulated by a computer-controlled air blower with adjustable rotational frequency. The sheath air is conducted through an adsorption dryer with silica gel as an adsorbing agent. Due to the relative dryness of the air sampled in Siberia, we observed no critical enrichment of the circulated sheath air with moisture to date; in fact the relative humidity in the sheath air circuit stayed in the range between 10 and 20% at all times during the measurement period, which makes technical measures to reduce the sheath air humidity redundant.

Since legal restrictions prevented the deployment of a radioactive source as an aerosol neutralizer, a corona-discharge based aerosol neutralizer was used upstream of the DMPS system. This aerosol neutralizer consists of an aerosol chamber with an AC electrical discharge and was developed by Stommel and Riebel (2004). Several tests with this aerosol neutralizer have shown that its performance is equivalent to that of a commonly used radioactive Kr^{85} neutralizer for a wide range of aerosol concentrations and sampling flows (Stommel and Riebel, 2004, 2005). The particle number concentrations found at ZOTTO lie within the lower concentration range of the device so that the aerosol exiting the aerosol neutralizer can be safely regarded in thermodynamic charge equilibrium at room temperature.

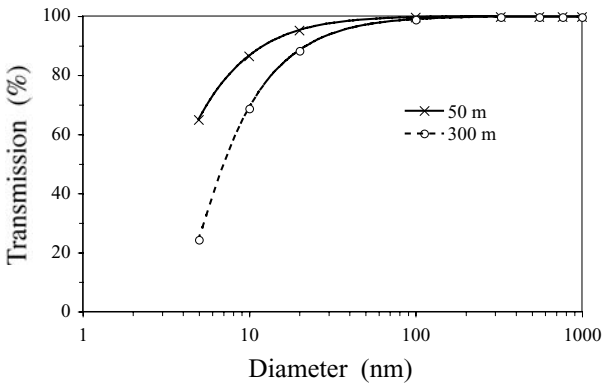

Fig. 1. Size-dependent transmissions of the 50 and 300 m aerosol inlets at the ZOTTO facility (after Birmili et al., 2007, Fig. 7).

Along with the switching frequency between the 300 and 50 m height levels, particle mobility distributions are recorded at about 6-min intervals. After inversion of the mobility distributions according to Stratmann and Wiedensohler (1996) the number size distributions was corrected for transmission losses with the results of the pre-experiment inlet calibration described earlier (Birmili et al., 2007). For completeness, the respective transmission curves for 50 and 300 m are given in Fig. 1. For further analyses 1-h to daily averages are formed.

2.4. Quality assurance

Before shipping to the ZOTTO site, the DMPS instrument was compared to an identical instrument at the World Calibration Center for Aerosol Physics at IfT Leipzig. The comparison revealed only small differences between both instruments, with a maximum deviation in concentration of 20% at the lower particle end. In order to provide reliable particle number concentration measurements after shipping and installation, measurements of total particle concentration using a CPC (TSI model 3010) were conducted over a limited period of 3 d during the set-up period in ZOTTO in parallel to the DMPS system. The comparison between CPC and DMPS total particle concentrations was subsequently used to correct the particle size distributions.

3. Results

3.1. Aerosol statistics

3.1.1. Total number and volume concentrations. The DMPS was operated from September 23, 2006 to June 1, 2007. For

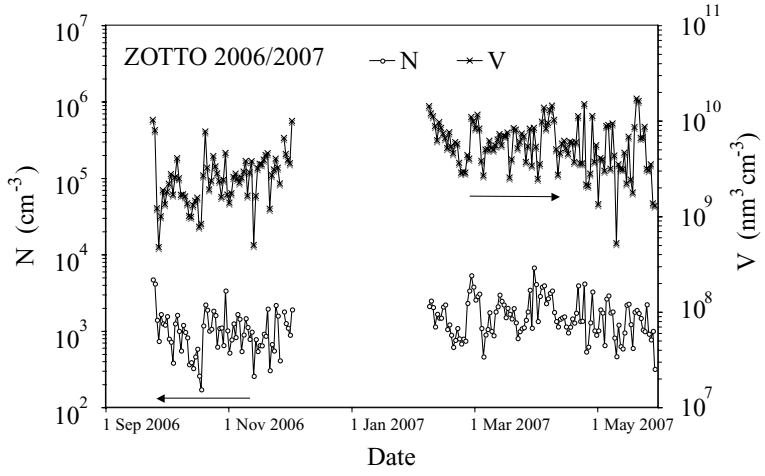

Fig. 2. Time-series of daily averages of total number (N, cm^{-3}, left-hand scale) and total volume (V, nm^3 cm^{-3}, right-hand scale) in 300 m height at the ZOTTO facility from September 9, 2006 to May 31, 2007. The break in the time series is due to butanol shortage at ZOTTO for want of transportation during December and January.

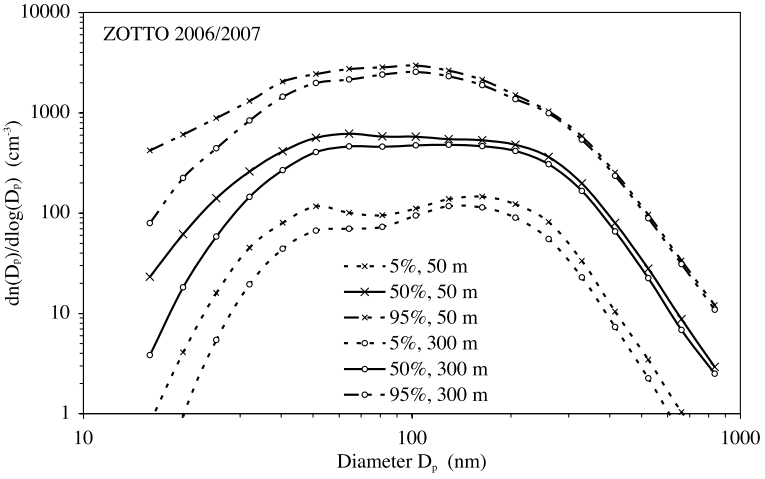

Fig. 3. Grand average percentiles (5, 50 and 95%) of 1-h-average number size distributions taken at 50 and 300 m height at the ZOTTO facility from September 9, 2006 to May 31, 2007.

want of butanol its operation ceased between December 2, 2006 and February 7, 2007. A first overview of the results in terms of daily-average total number (N) and volume (V) concentrations is shown for 300 m in Fig. 2. Interestingly, the total number concentrations were quite variable within two orders of magnitude. The concentrations at ZOTTO are mostly around a few thousand per cm^3. Both particle number and volume indicated a possible maximum in late winter, which could be associated to the well-known Arctic haze phenomenon (e.g. Heintzenberg, 1989).

3.1.2. Particle size distributions. The grand average size distributions, on an hourly basis, at both levels are shown in Fig. 3. A few differences between both levels are visible with respect to the shape of the number size distributions, mostly at the smallest sizes.

3.1.3. Ratio between measurements at height 300 and 50 m. A novel aspect of the ZOTTO-measurements is aerosol sampling at two levels, $h = 300$ and 50 m. The comparison of the size distributions at both levels yielded systematically higher particle numbers and volumes at 50 m compared to 300 m.

On a daily basis the 5, 50 and 95% percentiles of the ratios N_{50}/N_{300} and V_{50}/V_{300} are 1.0, 1.2, 1.9 and 1.0, 1.2, 1.8, respectively. Significant deviations from the value one only occur below about 50 nm, that is, the size range of newly formed particles. One might suspect that this finding indicates the influence of local contamination due to combustion sources near ZOTTO that more likely reach the 50 m level than the 300 m level. However, the highest ratios dn_{50}/dn_{300} occur during the 5% of lowest concentration, which speaks against local contamination as a possible cause.

3.1.4. Modal aspects of the size distribution. To explore the modal characteristics of the size distributions, notably showing up in the fifth percentile of the data (cf. Fig. 3), hourly averages of the number size distribution were fitted with a maximum of four lognormal distributions in the range of modal diameters between 10 and 800 nm, that is, somewhat beyond the size range of the data in order to fit even structures close to the data limits. We used an iterative fitting procedure with a modification of the method presented by Dal Maso et al. (2005). Within a given average relative deviation between the 18 fitted and the respective

original number concentrations per size distribution the number of fitted modes was minimized while adjusting the remaining modes to maintain the quality of fit. Seventy-four percent of the fitted distributions required four modes to be fitted within ±12% average relative deviation. The respective numbers for three, two and one mode were ±12, ±8 and ±1%.

A histogram of the probability distribution function (PDF) of modal diameters at the two inlet heights is drawn in Fig. 5. At 50 m four relative maxima of occurrence are visible in the classes 14–20, 27–38, 74–105 and 207–290 nm diameter. The absolute maximum of the PDF is given in the accumulation mode range (207–290 nm); this finding confirms the accumulation mode as the most constant and, therefore, most long-lived component of the particle number distribution. This observation is not unique; Over the European boreal forest Tunved et al. (2005) found a marked maximum in the PDF of lognormal modal diameters as well in the accumulation mode range. The positions of the relative maxima in PDF are the same at 300 m. The first maximum is lacking at the higher level, however, and the peaks are somewhat broader (except for the accumulation mode at 240 nm). As in the discussion of Figs. 3 and 4 the PDF in Fig. 5 shows that nucleation mode particles appeared more frequently at the lower level, whereas the next higher mode at 33 nm (possibly an aged nuclei mode) appeared more frequently at 300 m. Preliminary statistics of the parameters of the four lognormal modes fitted to 1-h average number size distributions are collected in Table 1.

3.1.5. Comparison with previous work. Total number (N_{300}) concentrations at 300 m height ranged between 400 cm^{-3} (5%) and 4000 cm^{-3} (95%) with a median of 1200 cm^{-3}, which is rather high for a nearly uninhabited boreal forest region during the low productivity period of the year. The corresponding volume (V_{300}) concentrations ranged between 1 and 11 μm^{-3} cm^{-3}. Assuming a particle density of 1.5 g cm^{-3} the 95% volume percentile would correspond to 17 μg^{-3} m^{-3}, which is near the Central European background level (Heintzenberg et al., 1998).

Previous measurement of atmospheric particle number distributions in the region of Siberia have been scarce, and generally been conducted only during shorter periods. Measurements for 2 weeks at Lake Baikal in July 1991 and for 1 month in June–July 1992 30 km south of Novosibirsk were combined by Koutsenogii and Jaenicke (1994) to a 'mean aerosol size distribution for Siberia'. Two of the lognormal modes that they derived fell into the diameter range covered at ZOTTO for which they derived the following parameters number, geometric mean diameter and geometric standard deviation: 2900 cm^{-3}, 1.65 and 29 nm for an Aitken mode and 0.3 cm^{-3}, 2.4, 400 nm for a large accumulation mode. Whereas the total number concentration reported by Koutsenogii and Jaenicke (1994) falls into the range observed in our work, their modal parameters diverge significantly from our findings reported in Table 1.

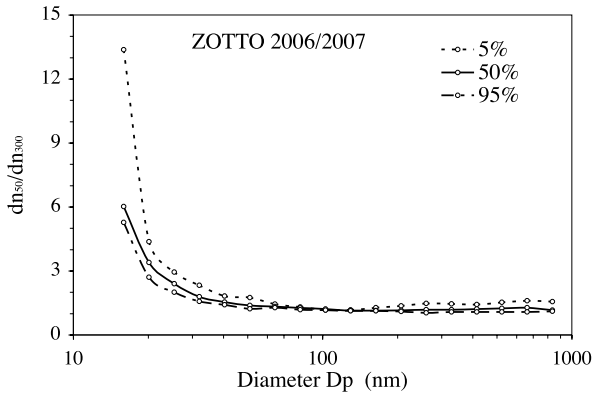

Fig. 4. Size-dependent ratios of one-hour-average-number concentrations measured in 50 and 300 m height at the ZOTTO facility from September 9, 2006 to May 31, 2007.

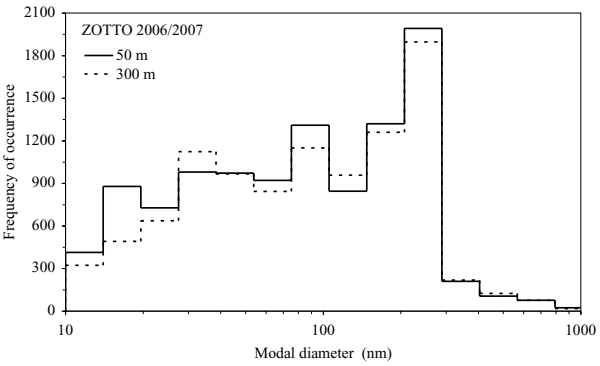

Fig. 5. Frequency distribution of modal diameters of 1-h-average number size distributions taken at 50 and 300 m height at the ZOTTO facility from September 9, 2006 to May 31, 2007. The size distributions were fitted with a maximum of four lognormal modes in the range 20–800 nm modal diameters.

Vartiainen et al. (2007) report on particle size distribution and ion measurements on board of a railway carriage, thereby performing a longitudinal measurement along the corridor of the Trans-Siberian railway, about 500 km south of ZOTTO. For rural, clean conditions (i.e. after removal of anthropogenic influences), they report median particle number concentrations for the accumulation mode ($D_p > 90$ nm; 733 cm^{-3}), the Aitken mode (25 nm $< D_p <$ 90 nm; 911 cm^{-3}), and the nucleation mode ($D_p < 25$ nm; 617 cm^{-3}), which broadly correspond to the total number concentrations found in our work. Since the measurements by Vartiainen et al. (2007) were conducted before the installation of ZOTTO (i.e. no concurrent measurements are available yet), a direct comparison of the two data sets is only of limited use.

Long-term measurements from the boreal forest region have so far been available only in European parts of Eurasia: Mäkelä et al. (2000) reported statistical lognormal parameters at the rural background site Hyytiälä located near 62° N in Finland, that is, at a latitude comparable to ZOTTO: accumulation mode (178 cm^{-3}, 1.47, 187 nm), Aitken mode (572 cm^{-3}, 1.64, 61.6 nm), nucleation mode (283 cm^{-3}, 1.42, 13.6 nm); their

Table 1. Statistics of log-normal fits (four) to 1-h averages of number size distributions measured in 50 and 300 m height at the ZOTTO facility September 9, 2006–May 31, 2007, P,percentile; $N_{1–4}$, total number (cm^{-3}); $DG_{1–4}$, geometric mean (modal) diameter (nm) and $\sigma_{1–4}$, geometric standard deviation

Height (m)	P (%)	N_1	DG_1	σ_1	N_1	DG_2	σ_2	N_3	DG_3	σ_3	N_4	DG_4	σ_4
50	5	4.0	10	1.1	5.0	22	1.1	112	52	1.2	9.0	110	1.2
50	50	20	16	1.2	142	31	1.3	833	75	1.6	221	208	1.5
50	95	1200	19	2.1	1470	48	1.9	3390	96	2.2	2210	374	1.9
300	5	2.0	10	1.1	2	22	1.1	84	52	1.2	6.0	110	1.2
300	50	14	17	1.1	50	30	1.2	597	74	1.5	212	201	1.5
300	95	337	20	2.3	765	48	1.7	2360	97	2.1	1870	378	1.9

parameters for the accumulation and Aitken modes are remarkably close to the values observed at ZOTTO (Table 1).

To date Tunved et al. (2003, 2005) provided the most comprehensive set of size distribution parameters spanning several years of observations at the Scandinavian atmospheric research stations Aspvreten (Sweden), Värrio (East Lapland), Pallas (West Lapland) and Hyytiälä (Southern Finland). In different air masses they found accumulation mode diameters mostly between 150 and 300 nm, Aitken mode diameters between 44 and 55 nm, and total number concentrations between 600 and 3000 cm^{-3} in continental air masses. The Scandinavian observations show a significant decrease in total particle number concentration from south to north, more pronounced in the Aitken mode than in the accumulation mode. Tunved et al. (2006) showed that the production of secondary aerosol from biogenic precursors can make up the major amount of particle mass in the northern regions of Scandinavia.

Finally, a comparison with the long-term size distribution statistics of Birmili et al. (2001) at Melpitz, Germany reveals similarities and differences in the comparable size range >10 nm. The smallest PDF maximum at ZOTTO corresponds to the aged nucleation category of Birmili et al. (2001). The next higher mode at ZOTTO does not frequently occur in the Central European source region about Melpitz. The 90 nm maximum at ZOTTO comes closest to the Aitken maximum in northern continental air masses at Melpitz (76 nm), whereas the accumulation maximum at ZOTTO lies in the range of accumulation modes one (160–240 nm) in northern continental air masses at Melpitz.

3.2. Trajectory statistics and case studies

The unique aerosol sampling point in 300 m height over the Siberian forest raises the question: What aerosol is being sampled at this site? As an answer to this question a 3-yr statistical study of back trajectories from the two inlet points at ZOTTO was conducted. With the HYSPLIT model (http://www.arl.noaa.gov/ready/hysplit4.html) 144 h back trajectories were calculated every 12 h for the years 2004–2006. For case studies of air mass analysis additional back trajectories were calculated every 3 h during the DMPS-measuring period.

The trajectory points of *ca*. 2500 trajectories at each arrival height were counted in a matrix covering the latitudes 35°N–90°N and longitudes 20°E–130°E with 2.5° × 2.5° resolution irrespective of their height and of model-calculated precipitation along the trajectory. The number of counts in each grid cell was divided by the total number of trajectory points yielding relative frequencies of occurrences. The geographic area (in terms of grid cells) that contained 95% of all trajectories is depicted in Fig. 6 for both arrival points and trajectory lengths of 24, 48, 96 and 144 h. The trajectories cover large fractions of the Russian Federation and the Kazakhstan Republic. Table 2 gives rough estimates of the percentage of both countries that are covered by the 95%-area of the back trajectories from ZOTTO. This table also contains average heights above ground and average trajectory lengths in km. The full 6-d-trajectory lengths at the 50 and 300 m arrival points cover roughly 30 and 50%, respectively, of the Russian territory.

From a combination of the trajectory statistics with information on anthropogenic emissions more information can be gleaned about the aerosol sources that can be observed with the ZOTTO facility. As proxy of information on the emission of combustion aerosols the most recent CO-emission database EDGAR (http://www.rivm.nl/bibliotheek/rapporten/771060002.html) was used with annual northern hemispheric CO-emissions north of 35°N on a 1° × 1° grid for the year 2000. A PDF of these distributions is plotted in Fig. 7. By counting the number of times that back trajectory points hit individual CO-emission points of this grid one can estimate the probability that measurements at ZOTTO will be affected by the respective emissions. The related pdfs of CO-emission points that have been hit by back trajectories to the 50 and 300 m arrival heights are also drawn in Fig. 7. The CO-emissions that are 'seen' by ZOTTO are mostly in the range 10^4–10^7 kg p.a. However, the probabilities of seeing these emissions are on the order of 1% or less with higher emissions than 10^7 kg p.a. having much lower probabilities than 1%.

Beyond the statistics two case studies were made with the help of the high-density back trajectories during the actual measuring period. For that purpose all times with V at 300 m less or equal the 5% percentile of V and all times with volume concentrations

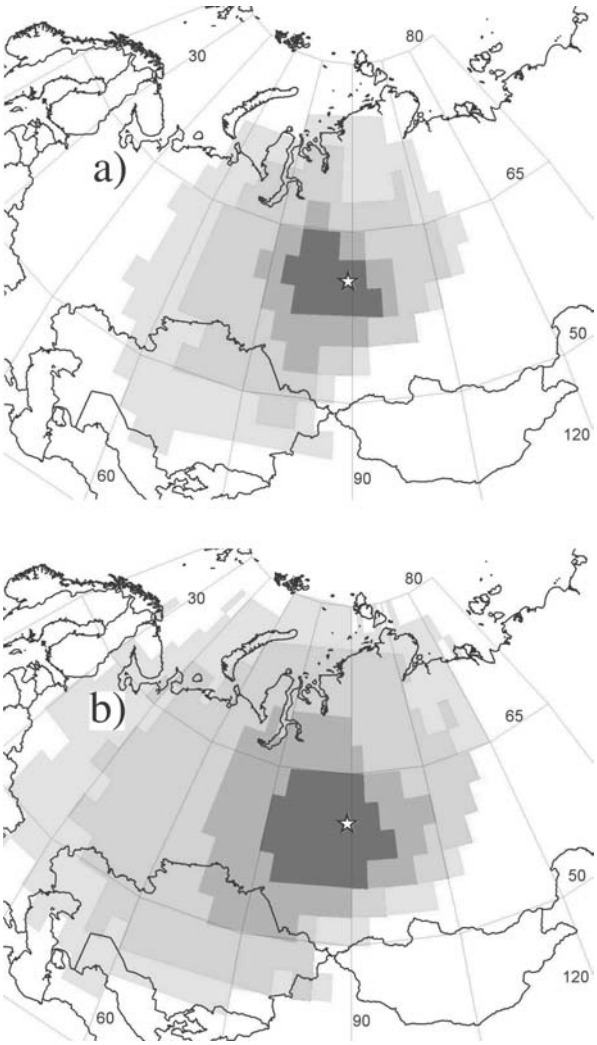

Fig. 6. Areal coverage of back trajectories arriving at 50 m (a) and at 300 m (b) at the ZOTTO facility (marked with a star) with the lengths 24, 48, 96 and 144 h (in decreasing shades of grey).

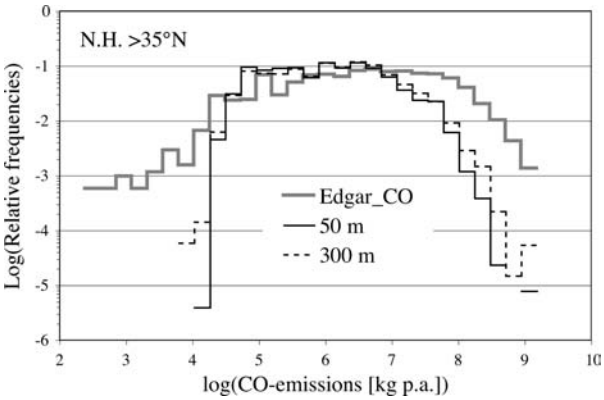

Fig. 7. Relative frequencies of occurrence of CO-emissions between 10^2 and 10^{10} kg p.a. north of 35°N taken from the EDGAR database (http://www.rivm.nl/bibliotheek/rapporten/771060002.html).

greater or equal 95% percentile of *V* were noted. All back trajectories within 3 h of the respective arrival times were collected in Fig. 8. Each trajectory, however, was utilized only once. Only trajectory positions with calculated heights above ground of 1000 m or less were plotted. When HYSPLIT indicated precipitation, the trajectory was terminated, assuming that wet scavenging would disconnect the aerosol properties measured at ZOTTO from air masses further upstream of the respective trajectory.

Back trajectories for the cleanest 5% of all cases were largely restricted to a narrow latitude band between 55°N and 70°N with a rather wide total longitudinal coverage from 60°E to nearly 120°E and stayed clear of larger emission centres (cf. Fig. 8a). Most of these trajectories were limited by precipitation. With two clear exceptions the air with the highest 5% in particulate volume came mostly from latitudes south of ZOTTO, reaching down to 50°N where many emission centres are located in the Transsiberian railway region (cf. Fig. 8b). These two exceptions reached back to the two industrial centres at Vorkuta and Noril'sk.

Table 2. Trajectory statistics for the time period January 1, 2004–May 31, 2007 based on three-dimensional back trajectories calculated at every 12 h to arrival heights 50 and 300 m with the NOAA HYSPLIT model (http://www.arl.noaa.gov/hysplit). Average height above ground (m), average length (km), and areal coverage (%) of the Russian Federation and the Kazakhstan Republic of back trajectories with lengths 24, 48, 96 and 144 h from the arrival points on 50 and 300 m height at the ZOTTO facility

Arrival height (m)	Trajectory length (h)	Average height above ground (m)	Average length (km)	Russian Federation (%)	Kazakhstan Republic (%)
50	24	48	300	3	0
50	48	57	600	8	0
50	96	89	1300	21	20
50	144	130	2100	32	62
300	24	300	540	6	0
300	48	320	1100	18	1
300	96	350	2200	39	62
300	144	360	3400	53	87

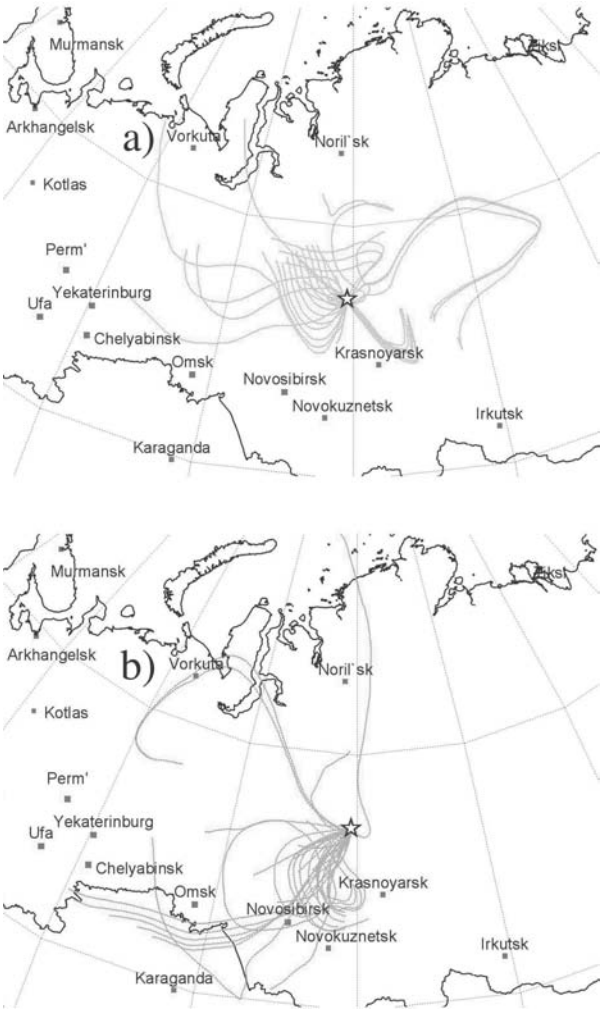

Fig. 8. 48 Back trajectories to 300 m at the ZOTTO facility during the time period September 9, 2006–May 31, 2007 for total volume (V) concentrations less or equal the 5% percentile of V (a). ZOTTO is marked by a star. The relative size of the cities is indicated by the size of the square marking their position. 38 Back trajectories to 300 m at the ZOTTO facility during the time period September 9, 2006–May 31, 2007 for total volume (V) concentrations greater or equal the 95% percentile of V (b).

4. Conclusions

This report describes the unique setup for and the first results of aerosol measurements at the new long-term Tall Tower monitoring facility near Zotino, Siberia (ZOTTO). Through two inlets at 50 and 300 m aerosol particle number size distributions are measured since September 2006 in the size range 15–835 nanometer dry diameter. The present report covers data until the end of May 2007 with a 2-month interruption in December and January due to butanol shortage. Total number (N_{300}) concentrations at 300 m height ranged between 400 cm^{-3} (5%) and 4000 cm^{-3} (95%) with a median of 1200 cm^{-3}, which is rather high for a nearly uninhabited boreal forest region during the low productivity period of the year. Corresponding volume (V_{300}) concentrations ranged between 1 and 11 μm^{-3} cm^{-3}. With a particle density of 1.5 g cm^{-3} the 95% volume percentile would correspond to 17 μg^{-3} m^{-3}, which is near the Central European background level (Heintzenberg et al., 1998). Median ratios N_{50}/N_{300} and V_{50}/V_{300} were 1.2 indicating that near-surface aerosol sources were dominating the ZOTTO site.

With a 3-yr back trajectory statistics the question as to which aerosol can be sampled at ZOTTO was addressed. At 300 m the average length of 6-d back trajectories is 3400 km, roughly covering 50% of the territory of the Russian federation plus 90% of the Kazakhstan Republic. Corresponding figures at 50 m height are 30 and 60%, respectively. From a combination of the trajectory statistics and the EDGAR database on CO-emissions the probability of the ZOTTO-aerosol measurements 'seeing' northern hemispheric combustion aerosols was estimated to be 1% or less. The 5% of ZOTTO-aerosol data with highest particulate volume concentrations to date could mostly be related via their respective back trajectories to urban emission centres in the region of the Transsiberian Railway whereas the back trajectories of the respective cleanest 5% avoided these emission regions.

The high sampling points at the tower were expected to free the aerosol measurements largely from local contamination due to power generation and surface traffic. This expectation was borne out by the general level and stability of the measurements at both levels and, in particular by the finding that the ratio of ultrafine particle concentrations at 50 m to those at 300 m is highest for the lowest absolute number concentrations.

The modal structure of the measured number size distributions was explored after fitting 1-h average distributions with a maximum of four lognormal functions. 74% of the fitted distributions needed four modes. The respective numbers for three, two, and one mode were 12, 8 and 1%. Ultrafine modes below 20 nm were more frequently seen at 50 m height than at 300 m, whereas the latter height more frequently showed an aged nucleation mode near 30 nm. The positions of Aitken (≈80 nm) and accumulation modes (≈210 nm) were very similar at both inlet heights, the very sharp latter one being the most frequent of all modes.

The very encouraging first aerosol results over the Siberian forest let us expect exciting new findings during the summer period with frequent forest fires and secondary particle sources from vegetation emissions as discussed by Tunved et al. (2006). The field-proven feasibility of aerosol measurements through a 300 m inlet should encourage others to implement similar measurements at other tall towers in order to monitor the atmospheric aerosol over very wide regions from a single point. It would also be desirable to couple the continuous aerosol and trace gas measurements at ZOTTO with concurrent episodic measurements along the Trans-Siberian transect such as illustrated in Vartiainen et al. (2007).

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