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Measurements of Ultrafine and Fine Aerosol Particles over Russia: Large-scale Airborne Campaigns

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In-situ measurements of ultrafine and fine aerosol particles obtained in the Siberian troposphere during large-scale airborne campaigns were analyzed. These measurements have been carried out in the framework of several international and Russian State projects aimed at to fill the gap in data on atmospheric components of climatic importance distributed in the Siberian air shed. In this paper we focus on particles with diameters from 3 to 20 nm to study new particle formation in the free troposphere (FT) over middle and high latitudes of Asia. In the upper troposphere number concentration of ultrafine particles varied between 20 and 3000 cm-3. The data obtained during YAK-AEROSIB/POLARCAT 2008 campaigns showed that remote Siberian troposphere is a relatively efficient source region for recently formed particles. Measurements carried out in the FT (3-7 km) showed that about 44% of them satisfied criteria of new particle formation. More favorable conditions are observed between 5 and 7 km (48%).

1. Introduction

Nucleation occurring in the free troposphere (FT) is a source of newly formed particles, whose eventual growth provides of the basis for formation of cloud condensation nuclei (CCN). Therefore processes of new particle formation (NPF) in the FT are of climatic importance due to their indirect radiative effect. The low temperatures and low surface area of preexisting particles observed in the upper troposphere favor nucleation and NPF. Recently nucleated aerosols have been observed in the FT widely (Kulmala et al., 2004; Lee et al., 2003; Young et al. 2007), and in spite of the fact that the most likely nucleation mechanisms are identified, there is no common agreement on their relative importance in the global atmosphere. Another problem is that studies in the remote continental FT remain sparse. The first large-scale observations in the clean free troposphere have been undertaken over Siberia in 2006 (Paris et al., 2009a). The main

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limitation of that investigation was a limited size resolution (with two size bins: 3-70 nm and 70-200 nm), so this study relied on the speculation that a large increase in particle number concentration combined with a high ratio $R=N_{3-70}/N_{70-200}$ reflect NPF. In this paper, we present new measurements that were carried out with a similar observational strategy but with an improved size resolution.

2. Experimental

The OPTIK-É Antonov-30 aircraft (http://iao.ru/en/resources/equip/plane/) was used as a research platform. It is a high-wing, two turbo-prop airplane, with one additional turbo-jet engine. This enables flying in a wide range of altitudes from 100 to 7000 m with speeds of 70–125 m s^{-1}. Its range ability is 3700 km, and maximum flight time is 8 hours.

2.1 Air samplers

The air sampling system consists of two independent samplers (Fig.1a) mounted on the starboard and larboard of the aircraft in the undisturbed zone in front of the propellers. Each one has seven inlets used for aerosol and trace gas sampling.

The forward-facing aluminum inlets 2 (Fig. 1) in combination with exhaust outlets 3 form the sampling system used for aerosol sampling and sizing. In order to maintain isokinetic conditions and suck the air without extra pumps, exhaust air is vented to the ambient atmosphere trough the outlets coupled with Venturi tube (Fig. 1b and 1c). Aerosol instruments for particle sizing and counting are connected to the inlets and outlets as shown in Fig. 1b and 1c. The 0.75 cm ID inlet 1 and outlet 3 of 1.6 cm ID (smaller one) are used to measure accumulation and coarse mode particles, and the 1.1 cm ID inlet 1 and outlet 3 of 2.8 cm ID for ultrafine and fine particle measurements. There is a bypass in the manifolds to reduce the pressure difference between the instrument inlets and outlets and diffusional losses of ultrafine aerosols in the air ducts as well.

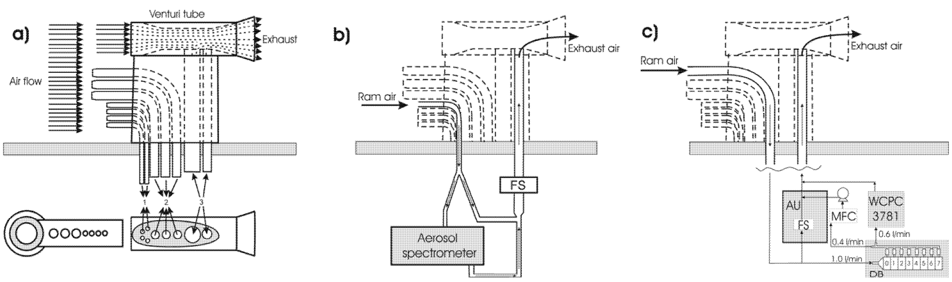

Figure 1. Isokinetic sampler with Venturi tube used to sample aerosols and trace gases: a – general view (gas/aerosol inlets 1, aerosol inlets 2, and exhaust outlets 3); b – an example of the optical aerosol spectrometer assembly (FS – flow stabilizer); c –the airborne diffusional particle sizer system: DB – 8-chanel synthetic-screen diffusion battery, AU – aspiration unit, MFC – mass flow controller.

2.2 Aerosol counting and sizing

Two devices are used for continuous aerosol measurements. The GRIMM (Grimm Aerosol Technik GmbH & Co. KG, Germany) Model 1.108 Aerosol Spectrometer is used to detect and count aerosol particles in 15 size ranges of 0.3, 0.4, 0.5, 0.65, 0.8, 1.0, 1.6, 2.0, 3.0, 4.0, 5.0, 7.5, 10, 15, and 20 µm. The Model 1.108 samples ambient air from the isokinetic sampler described above (Fig. 1b).

An 8-channel automated diffusion battery (ADB) designed at the Institute of Chemical Kinetics and Combustion SB RAS (Ankilov et al., 2002a, 2002b) is used to classify aerosol particles according to size in the diameter range of 3 to 200 nm. During campaigns of 2008, the ADB was connected to a dibutyl phthalate-based condensation particle counter (CPC). This CPC had a response time of about 20 s, so in order to minimize the time of one scan we used only three channels of the ADB to measure the aerosol number concentration of three size ranges: 3-6 nm, 6-21 nm, and 21-200 nm. The diffusional particle sizer system (ADB+CPC) was significantly upgraded in 2009, in particular the old CPC was replaced with a fast TSI (TSI inc., USA) Model 3781 Water-based Condensation Particle Counter (WCPC). This allowed the entire size distribution measurements to be carried in the diameter range of 3 to 200 nm and the total number concentration of particles as well. The inversion algorithm of the ADB data to particle size distribution was developed by Eremenko and Ankilov (1995). When calculating size spectra the counting efficiency of ultrafine particles of the Model 3781 WCPC is taken into account. An upgraded diffusional particle sizer (DPS) was used during airborne survey carried out in 2009, when the aircraft flew from Novosibirsk to Anapa for air quality study over Black Sea resorts.

Aspiration system has been specially designed for airborne applications with regulated flow rate, compensating for outside pressure variations and minimizing diffusional losses in the inlet manifold. Transmission efficiency for the airborne instrument is corrected for, and is ~ 0.997 in the 70–200 nm range and between 0.82 at 400 hPa and 0.89 at 1000 hPa for the 3–70 nm size range.

2.3 Flight patterns

During campaigns the flight pattern consisted of consecutive ascents and descents to make as many vertical profiles as possible (Fig. 2). Each profile was made between 0.5 (or minimum flight altitude allowed by topography) and 7 km. The measurements over the Lake Baikal were carried out in the atmospheric boundary layer (500-1000 m).

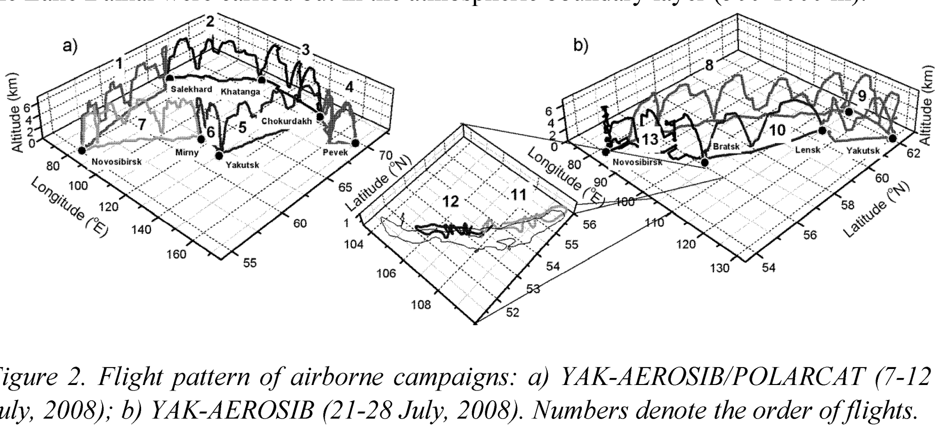

Figure 2. Flight pattern of airborne campaigns: a) YAK-AEROSIB/POLARCAT (7-12 July, 2008); b) YAK-AEROSIB (21-28 July, 2008). Numbers denote the order of flights.

3. Results and discussion

3.1 Particle concentrations over high latitudes

The investigation in the high latitudes has been done in the framework of YAK-AEROSIB project as a contribution to the summer 2008 POLARCAT programme of IPY (Paris et al., 2009b). The aircraft flew between longitudes 67° and 170°E, and latitude ranged from 65° to 72°N (flights 2-4, Fig.2). The total particle number concentration in the FT over this region varied in the range 120-1200 cm^{-3} (3-5 km) and 70-2200 cm^{-3} (5-7 km). According to Jaenicke (1993), the number concentration of the background aerosol aloft typical for polar FT is about 200 cm^{-3}. Higher concentrations observed during the campaign may denote new particle formation events occurring in the middle and upper troposphere, the more so as the number concentration of ultrafine particles (N_{3-6}) in some areas exceeded 1000 cm^{-3} and ranged from a few particles to 2000 cm^{-3}. There are several approaches to identify new particle formation events by specific criteria (Lee et al., 2003; Young et al., 2007). In this study we used similar criteria as Young et al. (2007): (1) N_{3-6}>10 cm^{-3}, (2) $N_{3-6}/N_{6-21}>1$ and $N_{3-21}/N_{21-200}>0.5$. So when one of the ratios R1 or R2 tends to the above limits the new particle formation is weakened. Data analysis done with the use of these criteria showed that more often NPF events occur in the upper troposphere (5-7 km). Thus, when the aircraft flew within 5-7 km layer between 106 and 116°E (Flight 3; 08.07.2008), the median of the N_{3-6}, R_1, and R_2 values were 182, 3.8 and 1.2 respectively. Arithmetic means and medians of particle concentration values are summarized in Table1.

3.2 Particle concentrations over middle latitudes

Measurements carried out in the middle latitudes gather more data since flights were performed twice in too weeks in this region. At mid-latitudes, observed in-situ concentrations of ultrafine particles in the FT were distinctly higher, and ratios R_1 and R_2 lower, than at high latitudes. For instance, during the second 7-km horizontal plateau (Flight 8; 21.07.2008) the medians of number concentration, R1 and R2 during NPF events were: N_{3-6}=403 cm^{-3}; R_1=1.5; R_2=1.7. Their maxima during these events were 1900 cm^{-3}, 5.0, and 4.7, respectively. Such difference relative to high latitudes can be explained by the influence of generally stronger convection, at mid latitudes. It means that, on the one hand, more fresh particles can form in the outflow regions of mesoscale convective systems (MCS) due to the last enrich the upper troposphere with aerosol precursors (Twohy et al.; 2002; Paris et al., 2009a), and, on the other hand, convection can transport preexisting Aitken nuclei from the lower troposphere that leads to the decrease of ratios R_1 and R_2.

Table 1. Mean and median number concentrations (cm^{-3}) observed in the FT over high latitudes (HL) and mid-latitudes (ML) during campaigns 2008.

Region		Mean			Median			Standard deviation		
		N_{3-200}	N_{3-6}	N_{6-21}	N_{3-200}	N_{3-6}	N_{6-21}	N_{3-200}	N_{3-6}	N_{6-21}
HL	3-5 km	237	77	42	212	35	42	161	148	23
	5-7 km	347	157	79	248	61	63	294	277	58
ML	3-5 km	682	226	147	410	81	87	830	539	203
	5-7 km	855	307	225	598	152	148	838	490	227

3.3 A case study of aerosol number size distribution in the free troposphere

Measurements of aerosol number size distribution in the FT over middle latitudes were carried out during another flight, from the West Siberia to the Black Sea coast (map not shown). The same diffusional particle sizer and optical particle counter as described above were used. Before the flight, both instruments have been tested with GRIMM aerosol generators (Model 7.811 and 7.822). Time resolution of DPS is 80 s.
These measurements with improved size resolution (section 2.2) are analyzed to confirm the results described above. Fig. 3 shows that number size distribution in the FT shifts towards smaller sizes in comparison with the one in the lower troposphere. The entire size spectra also provide the mean values used in characterizing aerosol size distribution. So, in addition to the above NPF criteria, the geometric mean diameter (D_{pg}) can be used. According to the properties of the log-normal distribution, D_{pg} is equal to the number median diameter. Thus, when the increase in the total number concentration is accompanied by the D_{pg} decreasing down to the nucleation size range denoting that in situ NPF has occurred, because half the particles lie below this diameter. Several such events observed in the FT during this flight can be seen from Figure 3.

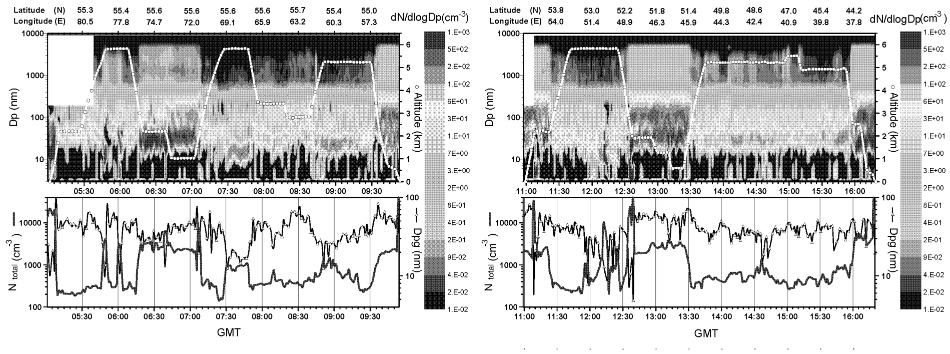

Figure 3. Number size distributions, total number concentration (N_{total}), and geometric mean diameter (D_{pg}) observed during the flight from Novosibirsk to Anapa (03.07.2009)

4. Summary and conclusions

Measurements of ultrafine and fine particles carried out in the free troposphere over high and middle latitudes of Russia have been presented. Concentrations of ultra-fine (N_{3-6}) particles observed in the upper troposphere ranged from a few particles to 2000 particles per cm^3.
Summarising the data obtained during two intensive measurement campaigns we can draw the conclusion that remote Siberian troposphere is a relatively efficient source of recently formed particles. Measurements carried out in the FT (3-7 km) showed that about 44% of them can be related to new particle formation. At the same time, more favourable conditions are observed between 5 and 7 km (48%).

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