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Large-scale aircraft observations of ultra-fine and fine particle concentrations in the remote Siberian troposphere: New particle formation studies

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Ultra-fine particle number concentrations were measured over Siberia during two large-scale airborne measurement campaigns in April and September 2006. During both campaigns, an aircraft flew between Novosibirsk and Yakutsk, collecting every 200 km vertical profiles up to 7 km. This dataset was completed by 5 years of monthly profiles above Novosibirsk. Particle number concentration was measured in the size ranges 3–70 and 70–200 nm, along with other tracers. Free troposphere (FT) particle concentrations (N_{3-200}) varied between 60 and 460 cm^{-3} , inferior to boundary layer concentrations (100–7000 cm^{-3}). In April, high concentrations of $\sim 500 \text{ cm}^{-3}$ were observed in a polluted air mass recently uplifted at 5–6 km altitude over eastern Siberia, with no sign of significant new particle formation. In September, particle concentrations decreased with altitude, but with a steeper gradient in N_{70-200} compared to N_{3-70} , the latter accounting for 90% of the total particle concentration in the free troposphere at 6–7 km altitude. Because ultra-fine particles presumably have short lifetimes, these observed particles could have been formed in situ in the clean Siberian atmosphere. Two cases of possible nucleation with high concentration and N_{3-70}/N_{70-200} ratios are reported for the September campaign, in the upper troposphere and in cloud outflow in the mid-troposphere. In the seasonal analysis, a FT N_{3-70} maximum is found in July–August between 6 and 7 km altitude, with N_{3-70} accounting for $\sim 90\%$ of N_{3-200} supporting the hypothesis of in situ formation in the FT. A secondary FT maximum of N_{3-70} was identified later in autumn. In the boundary layer, seasonally maximum N_{3-70} concentrations were found over Novosibirsk in May and September, but not in summer, possibly due to scavenging by precipitations and a large condensational sink from biomass burning aerosols. Our dataset has a limited size resolution and no speciation capability; more investigation is thus required to understand the conditions leading to in situ nucleation processes in the Siberian air shed.

1. Introduction

Aerosols play an essential role in the Earth system and impact the Earth energy balance due to their ability to interact with light and to nucleate clouds (Charlson et al., 1990). Free tropospheric particles are either uplifted from the boundary layer or formed in situ from gaseous precursors. Gas-to-particle conversion occurs as two competing processes: condensation on pre-existing particles or as nucleation. Binary homogeneous nucleation of sulphuric acid and water vapour is a dominant nucleation mechanism. High nucleation rates have also been observed at ultra-low sulphuric acid concentration (Covert et al., 1992; Hoppel et al., 1994; Weber

et al., 1996), suggesting ternary formation of new particles, e.g. with ammonia, is also a significant nucleation mechanism (Korhonen et al., 1999). Also, ion-induced nucleation (e.g. Lee et al., 2003) or activation of existing clusters (Kulmala et al., 2006a) can be important. Ultra-fine particles either grow or evaporate. Particles with size $\sim 10 \text{ nm}$ have a lifetime of about 2 h in the free troposphere (Williams et al., 2002), increasing with decreasing condensational sink.

Freshly nucleated aerosols have been observed at many locations in the free troposphere (FT) (Covert et al., 1992; Weber et al., 1996; Clarke et al., 1998, 1999; Nyeki et al., 1999; de Reus et al. 2000; Twohy et al., 2002; Lee et al., 2003; Minikin et al., 2003; Young et al. 2007; Benson et al., 2008). Twohy et al. (2002) in particular observed high condensation nuclei concentration up to 45,000 cm^{-3} from nucleation in the outflow of a mesoscale convective system. These observations are summarized in Table 1.

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Table 1

Observed concentration distribution in the FT from the literature, mostly during nucleation events.

Reference	Concentrations (cm ⁻³)	Size range (nm)	Location	Latitude	Altitude
Schröder and Ström, 1997	50	7–18	West Europe	Mid lat	LT
–	300	7–18	–	–	UT
–	1500	>18	–	–	FT
Nyeki et al., 1999	[35–550]	5–15	Germany	46° N	FT
–	[1000–2000]	>15	–	–	–
De Reus et al., 2000	[50–100]	6–18	Tenerife	29° N	FT
–	[500–1000]	>18	–	–	–
Hermann et al., 2003	[50–500]	4–12	Europe	Mid lat	UT/LS
–	[1000–6000]	>12	–	Mid lat	UT/LS
Minikin et al., 2003	1400 [450–15000]	>5	NH	NH	UT
–	770 [290–9600]	>14	–	–	–
–	40 [16–90]	100–1000	–	–	–
Twohy et al., 2002	Up to 45000	>25	Central US	Mid lat	UT
Lee et al., 2003 ^a	330	4–9	NH	Mid & high lat	UT/LS
– ^b	60	4–9	–	–	–
–	200–500	4–2000	–	–	–
Young et al., 2007 ^c	700–3960	4–9	US	Mid lat	UT/LS
Benson et al., 2008 ^a	200–400	4–9	US	Mid lat	FT
– ^a	250–750	4–2000	–	–	–
This study ^d	50–350	3–70	Siberia	Boreal	FT
– ^d	90–580	3–200	–	–	–
– ^e	50–1100	3–70	–	–	–
– ^e	75–1500	3–200	–	–	–

Range is given between square brackets.

^a Average concentration reported in the presence of identified nucleation.

^b Average concentration reported in the absence of identified nucleation.

^c within tropopause fold.

^d April 2006 campaign, all data.

^e September 2006 campaign, all data.

Observations of nucleation in the clean continental free troposphere remain sparse. Leaitch and Isaac (1991) argued that continental FT surveys are likely to be affected by lateral advection of DMS-rich marine air.

No extensive study has been reported for the clean continental free tropospheric air shed of Siberia. Previous boundary layer studies in this region highlighted the contribution of anthropogenic and biogenic emission to aerosols concentrations (Bashurova et al., 1992; Koutsenogii et al. 1993; Koutsenogii, 1997; Vartiainen et al. 2007; Heintzenberg et al., 2008; Dal Maso et al., 2008). Most of Siberia exhibits deep atmospheric boundary layers in summer, as the relatively dry boreal conifer forests tend to produce high sensible heat fluxes (Baldocchi et al., 2000). The diurnal growth of a deep convective boundary layer will ensure vigorous ventilation of surface emissions such as biogenic volatile organic compounds (Lathiere et al., 2006) in summer, which may in turn favour nucleation (O'Dowd et al., 2002).

We present new aerosol size distribution data from two continent-wide campaigns carried out over Siberia (between 55° N and 63° N) in September and April 2006 and from 5 years of monthly vertical profiles over Novosibirsk. The question of a nucleation signal in ultra-fine aerosols concentration in the Siberian FT, and its seasonal variation, is addressed.

Particle number concentrations are measured in the ranges 3–70 nm and 70–200 nm. Ultra-fine particle number concentration in the 3–70 nm range (N_{3-70}) accounts for most of the total number of particles in remote clean continental air (Seinfeld and Pandis, 1998), but only for a negligible fraction of the aerosol mass. Particle

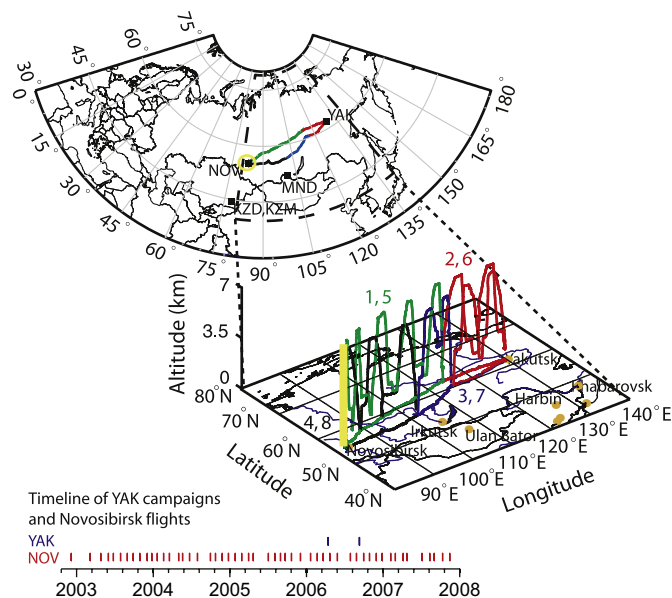


Fig. 1. Flight route during the campaigns. The April 2006 and September 2006 campaigns followed the same route. Each of these two campaigns was split into the same four flights (in the top panel, numbered 1 to 4 in April and 5 to 8 in September 2006 and differentiated by colours: green, red, blue and black). Novosibirsk profiles site is also shown in yellow. Bottom panel shows the timeline of measurements (red: regular profiles in Novosibirsk, blue: YAK intensive campaigns).

concentration in the 100–1000 nm size range is proportional to aerosol total surface and hence to the gaseous precursors aerosol surface sink. This coarse size resolution information is used to assess the large-scale distribution of ultra-fine particles in the free troposphere.

CO enhancement is used here as a tracer of pollution from anthropogenic sources and biomass burning. SO₂ was not directly measured during our campaigns so we instead use CO as a proxy of potential gaseous precursors such as SO₂. CO and SO₂ have common anthropogenic sources, such as combustion of coal and other fuels, also emitting submicron particles of black carbon and organic carbon (e.g. Ohara et al., 2007). CO enhancements may not necessarily be correlated with enhancements in SO₂ because of their different lifetimes against reaction with OH (1 month or more and a few days respectively; Seinfeld and Pandis, 1998).

2. Methods

The YAK-AEROSIB campaigns have been conducted in April and September 2006 (Paris et al., 2008) using a chartered Antonov-30 operated by the Institute of Atmospheric Optics in Tomsk. The two campaigns were carried so as to allow maximum sampling of the troposphere below 7 km, with a see-saw flight pattern combining 26 vertical profiles between 0.5 and 7 km of altitude spaced horizontally by ~300 km (Fig. 1). Each campaign consists of four flights, numbered 1–4 during the April campaign and 5–8 during the September campaign. Campaign measurements are completed by monthly vertical profiles near Novosibirsk (NOV in Fig. 1 top panel; Arshinov and Belan, 2004) collected between December 2002 and November 2007. The monthly flights use the same aerosol instrument and aircraft as the intensive campaigns. The NOV profiles were always collected between 1200 and 1600 local time, from the ground to 7 km altitude.

Ultra-fine particle concentration in the diameter range from 3 to 200 nm are measured using an 8-channel automated diffusion battery (ADB; designed by ICKC SB RAS, Novosibirsk; Reischl et al.,

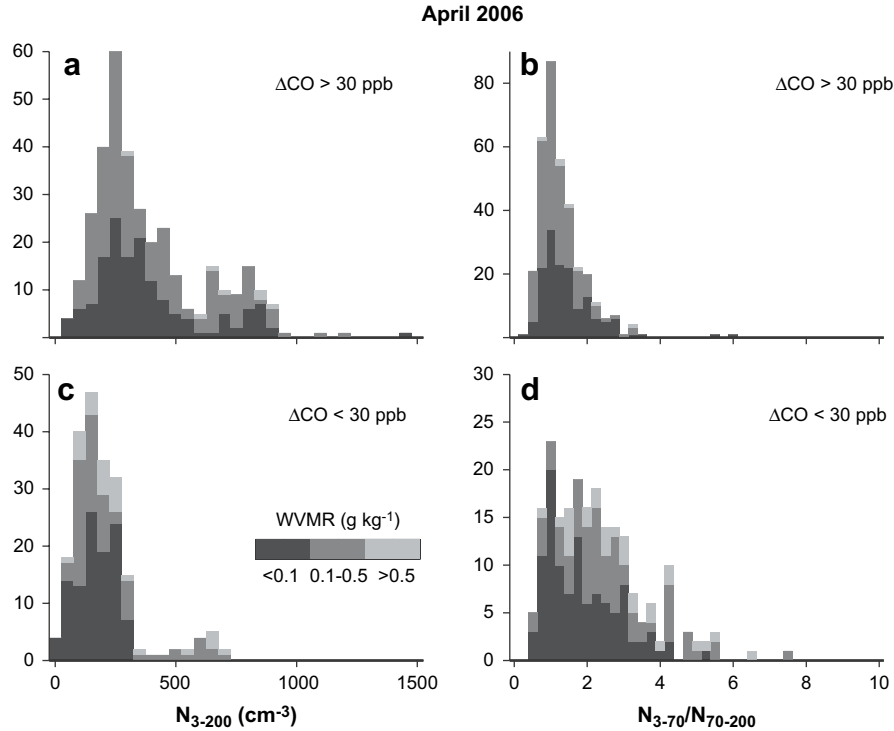


Fig. 2. (a) Free troposphere ($z > 3$ km) particle number concentration frequency distribution in the April 2006 campaign for samples with $\Delta\text{CO} > 15$ ppb (polluted air). ΔCO is defined in section 3.3. (b) R ratio in polluted air. (c) Particle number concentration in clean air ($\Delta\text{CO} < 15$ ppb). (d) R ratio in clean air. Grey shading represent the water vapour mixing ratio.

1991; Ankilov et al., 2002a, b) coupled to a condensation particle counter (Arshinov and Belan, 2004; Arshinov et al., 2007). Air is sampled using a quasi-isokinetic inlet (Zuev et al., 1992) at the front side of the aircraft. The ADB system has been specially designed for airborne applications with regulated flow rate, compensating for outside pressure variations and minimizing particle losses. In order to reduce integration (scanning) time and hence improve precision, only two channels of the battery are used in the instrument. This also improves the total particle concentration retrieval. Size response of the instrument is homogeneous over the 3–200 nm size range (Ankilov et al., 2002b) and for different aerosol compositions. 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. All concentrations are reported at standard pressure and temperature (STP) conditions.

The CO airborne analyser used for the YAK-AEROSIB campaigns is described in Nedelec et al., 2003. It is an automated instrument modified from the commercial infrared absorption correlation gas analyser (Model 48C, Thermo Environment Instruments, USA) to reach an accuracy of 5 ppb or 5% for 30 s integration time. Periodical (every 20 min), in-flight accurate zero measurements were performed during the campaigns. The O_3 instrument is derived from the commercial Thermo 49 instrument based on UV absorption with a precision of 2 ppb or 2% (e.g. Paris et al., 2008).

3. Results and discussion

3.1. Particle concentrations during the intensive campaigns

The total (sizes 3–200 nm) particle number concentration measured during the YAK-AEROSIB campaigns had a modal distribution of 150 cm^{-3} . In the April 2006 campaign, the median concentration was 190 cm^{-3} , and ranged from 90 to 580 cm^{-3} (10th and 90th percentile respectively; Fig. 2). One percent of the data

exhibited concentrations higher than 10^4 cm^{-3} , mostly in the vicinity of airports (at take-off and landing). Above 3 km, April concentrations are in the range $80\text{--}460 \text{ cm}^{-3}$. The distribution of ultra-fine aerosols has a maximum in the boundary layer (BL) where anthropogenic and biogenic emissions of aerosols and precursors are injected. In the lowest 1 km, in a very stable and shallow boundary layer, the April concentration ranged between 10^2 and $7 \times 10^3 \text{ cm}^{-3}$.

During the September 2006 campaign, the particle number concentrations observed were in the range $75\text{--}1.5 \times 10^3 \text{ cm}^{-3}$, (again, respectively 10th and 90th percentile; Fig. 3), with a median of 215 cm^{-3} and skewed towards lower concentrations. Above 3 km, typical September concentrations are between 60 and 340 cm^{-3} , and 1% of the data is above $3 \times 10^3 \text{ cm}^{-3}$. In the September BL (approximated by the lowest 2 km), variability was less important than the lowest 1 km in April, with a range $350\text{--}2 \times 10^3 \text{ cm}^{-3}$.

Fig. 4 shows three examples of the distribution of particle number concentration in vertical profiles, each profile being averaged over a given flight (6 ± 2 h). Also shown are the CO and water vapour mixing ratios. The September flights (shown in Fig. 4c; flight 8, 10 September 2006) exhibit larger N_{3-200} value in the BL or immediately above the BL. The strong sensible heat flux from the forest mixes efficiently biogenic volatile organic compounds (VOC) emitted by the vegetation (O'Dowd et al., 2002; Lathi  re et al., 2006; Tarvainen et al., 2007) into deep convective BL (Baldocchi et al., 2000), lofting these compounds up to the entrainment zone, where they can reach into the FT by night (Stull, 1988). Conditions favourable to nucleation were met during the September campaign. No local, fresh biomass burning plume was observed at that time. Only one aged fire plume from (agricultural) biomass burning transported from Kazakhstan and the Caspian Sea area was measured, which exhibited the highest CO concentrations of the campaign. In the September data above 4 km altitude, N_{3-70} is

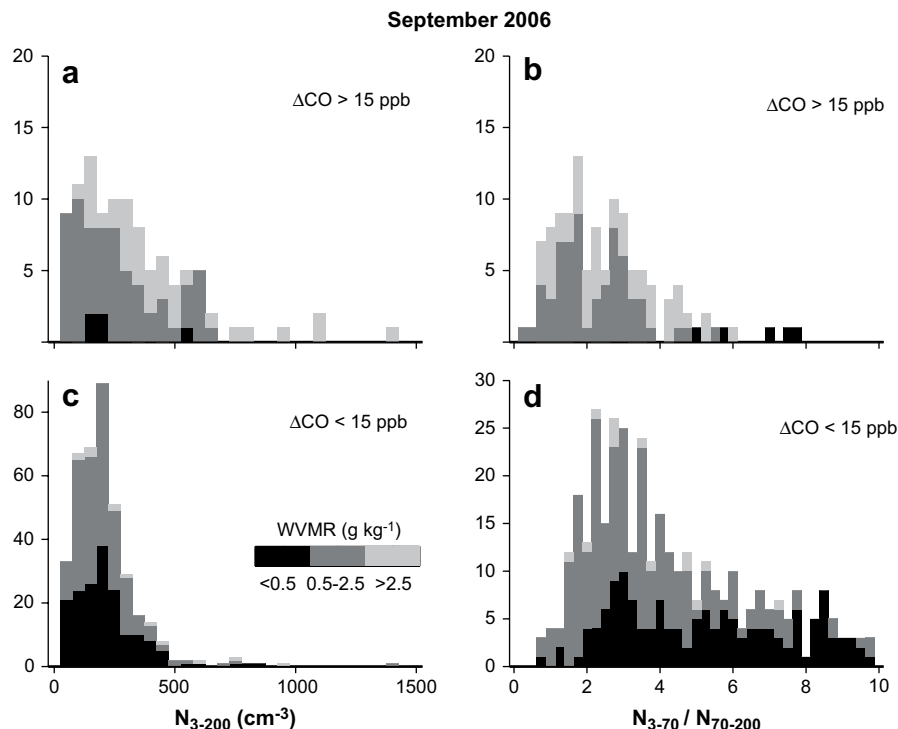


Fig. 3. (a) Free troposphere ($z > 3$ km) particle number concentration frequency distribution in the September 2006 campaign for samples with $\Delta\text{CO} > 15$ ppb (polluted air). (b) R ratio in polluted air. (c) Particle number concentration in clean air ($\Delta\text{CO} < 15$ ppb). (d) R ratio in clean air. Grey shading represent the water vapour mixing ratio.

higher by up to one order of magnitude than N_{70-200} . On the contrary, the April flights (April 12, 2006, Fig. 4a) has concentrations of N_{3-70} and N_{70-200} particles approximately equal, from the ground to flight ceiling.

3.2. Size distribution and the limitation of size resolution

Here we rely on the relative concentration of particle in the two size ranges to discriminate fresh particles (roughly, nucleation and Aitken modes) from condensation nuclei (CN). In the troposphere, under specific conditions of humidity, temperature and incoming radiation, nucleation yields a larger number concentration of ultra-fine particles relative to concentration nuclei. Studies on airborne aerosol measurements (using finer size resolution instruments) proposed nucleation event to be characterized by a modal distribution increased towards the finer sizes with specific criteria (see e.g. Covert et al., 1992; Lee et al., 2003; Young et al., 2007). We speculate here that a large increase in the ratio $R = N_{3-70}/N_{70-200}$ is sufficiently discriminative of freshly nucleated particles. The median of the R value is 1.5 for the April campaign (see Fig. 2), and 2.5 for the September campaign (see Fig. 3). In the September 2006 campaign 20% of the data had $R > 5$, and 5% had $R > 10$. In the free troposphere above 4 km altitude, 65% of the data had $R > 5$ and 10% had $R > 10$. No more than 4% of the data reached $R > 5$ in the April campaign. 11% of the September FT data had both $R > 5$ and $N_{3-70} > 100 \text{ cm}^{-3}$, which can be considered a possible criteria for the assessment of recently nucleated particles in the FT. Only 2% of the April data responded to this criterion.

3.3. Long range transport of surface tracers and particle concentrations: April 2006 case

High particle concentrations coincided with CO enhancements (Figs. 2 and 4a) in the mostly dry April troposphere, irrespective of altitude. During flights 2 and 3 in April 2006 uplifted boundary

layer air from polluted regions of China was identified in the mid-troposphere in Siberia above 5 km by Paris et al. (2008). Elevated CO concentrations were observed in this uplifted pollution (Fig. 4a), alongside with an absence of O_3 enhancement. The presence of Chinese pollution suggests the possible presence of coarse particles (anthropogenic and dust from dust storm in China), which could explain the absence of O_3 enhancement. Additional gas to particle conversion may occur in this plume.

In our sampled air mass from China, the observed N_{3-200} had median 300 cm^{-3} and third quartile $\sim 500 \text{ cm}^{-3}$, and was higher than in surrounding cleaner air (between 10 and 80 cm^{-3}), but remained low, pointing to a condensational growth of existing aerosols. At 5 km altitude in the uplifted pollution plume, N_{3-70} was higher than N_{70-200} , possibly reflecting a distinct layer where fresh nucleation occurred in the recent air mass history.

In the flights of April 2006, outside the well-defined uplifted pollution plume described above (which was encountered during Flights 2 and 3), the FT is characterized by very low N_{3-200} ($\sim 10^2 \text{ cm}^{-3}$) even in polluted air masses with CO excess (ΔCO) of ~ 100 ppb (Fig. 5). ΔCO is defined relative to the clean-air background according to Paris et al. (2008). There was no clear relation between ΔCO and the R ratio in the April 2006 campaign dataset. Within the clean FT air masses, the R ratio remained close to unity, suggesting a flat or symmetric size distribution. Over eastern Siberia (Flights 2 and 3), the R ratio is 0.99 ± 0.05 in air masses having $\Delta\text{CO} > 50$ ppb (associated with Chinese pollution), whereas it is 1.28 ± 0.11 for surrounding air. This observation suggests that new particle formation must have been very limited in a recent history of the sampled air mass, possibly with preferential sink on pre-existing particles.

Using CO as a surface tracer suggests that this air mass was exposed to large concomitant sources of both CO and SO_2 (Ohara et al., 2007). A literature survey suggests likely CO/ SO_2 ratios in the outflow of NE Chinese boundary layer. During a Chinese pollution export event over Taiwan, Lin et al. (2007) show well correlated SO_2

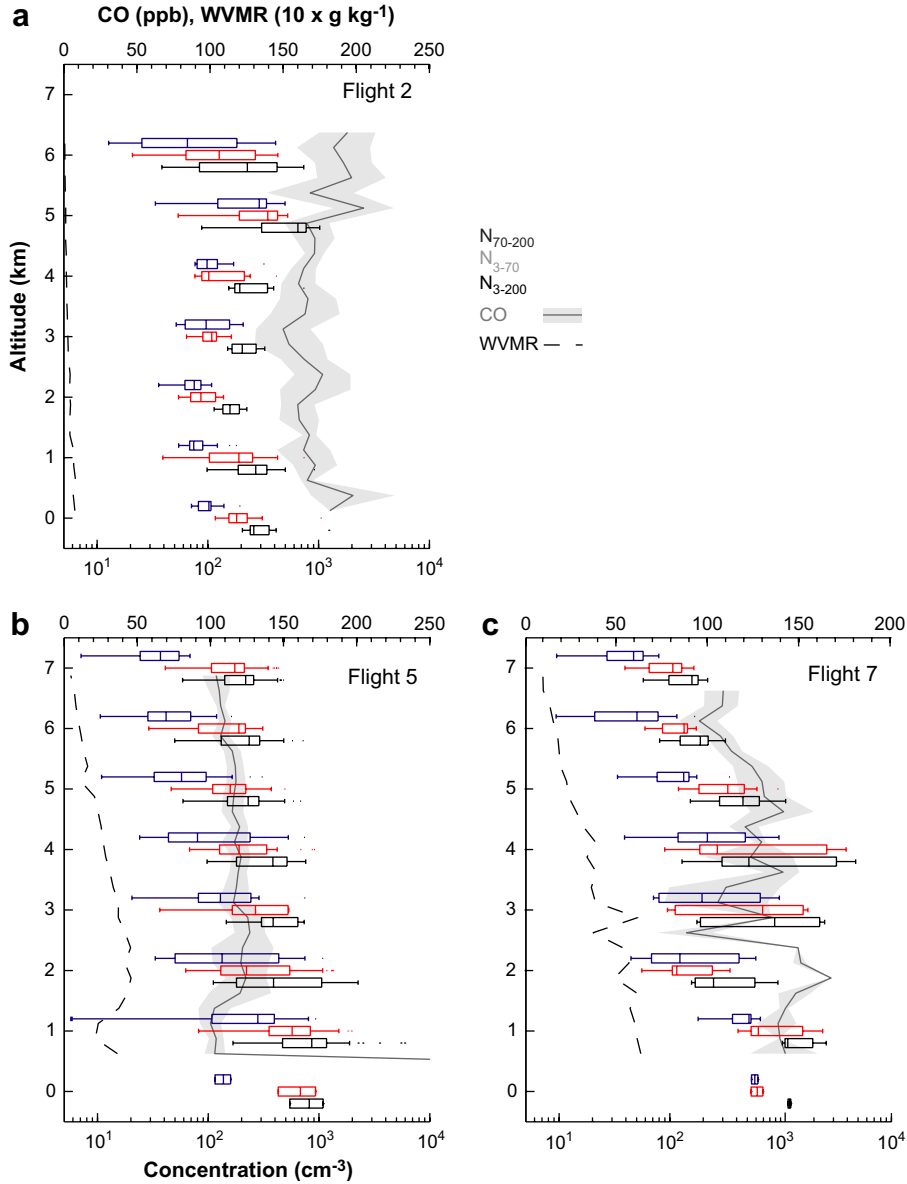


Fig. 4. Vertical profile of particle number concentrations and trace gases for (a) Flight 2 (12 April 2006), (b) Flight 5 (7 September 2006), and (c) Flight 7 (8 September 2006). Blue box plots represent the N_{3-70} concentrations, red box plots the N_{70-200} , black box plots the total N_{3-200} . The box-and-whiskers indicate the median, first and third quartile of the data in each 1 km altitude bin. Bars extending from the box encompass outliers residing at less than 1.5 inter-quartile range, more extreme outliers being marked as points. The dark grey line represents CO mixing ratio, with 1 standard deviation shaded in grey. The dashed line shows the water vapour mixing ratio. Each profile shown here is averaged over between 4 and 8 ascents and descents.

and CO measurement, with ratios of about $0.01\text{--}0.02 \text{ ppb}(\text{SO}_2)/\text{ppb}(\text{CO})$ after ~ 1 day of transport. By comparison, emission ratios from Ohara et al. (2007) are in the range $0.08\text{--}0.10 \text{ mol}(\text{SO}_2)/\text{mol}(\text{CO})$. In the polluted air that we sampled in April 2006, the 50–60 ppb CO enhancements could therefore be translated into SO_2 enhancements of about 0.5–1.2 ppb. In the immediate outflow of China in April Buzorius et al. (2004) found elevated CN concentration, coincident with high SO_2 concentrations (10 ppb), and alternating with layers of high CO and coarser particles acting as condensational sink, similar to the Chinese outflow described above.

3.4. Upper troposphere particle formation: September 2006 case

In September 2006, increasing R ratio was observed with decreasing CO enhancements (Figs. 3 and 4c). Dry FT air (4–8 km) has a lower concentration of particles but is characterized by R ratio

values up to 10. Due to a lower amount of anthropogenic pollution (for which we use ΔCO as a proxy) in the Siberian air shed at that time, the formation of new particles is able to dominate over the condensational sink.

Fig. 5 shows measured particle concentrations as well as relative humidity, CO and O_3 for Flight 5 on 7 September. Note the elevated CO concentrations ($\Delta\text{CO} \sim 60$ ppb, with large O_3 concentrations of 55–60 ppb) at about 2 km altitude at 0400–0415 UTC, resulting from the encounter with a biomass burning plume originating from Kazakhstan (Paris et al., 2008). The plume was encountered while crossing a warm front with heavy cloud cover (the cloud cover corresponding to the two sharp drops in particle concentration). The observed CO background in late summer is between 90 and 110 ppb, denoting a rather clean atmosphere. Particle concentrations are highly variable throughout the flight but two regimes emerge: high particle concentrations (a few thousand cm^{-3}) in the

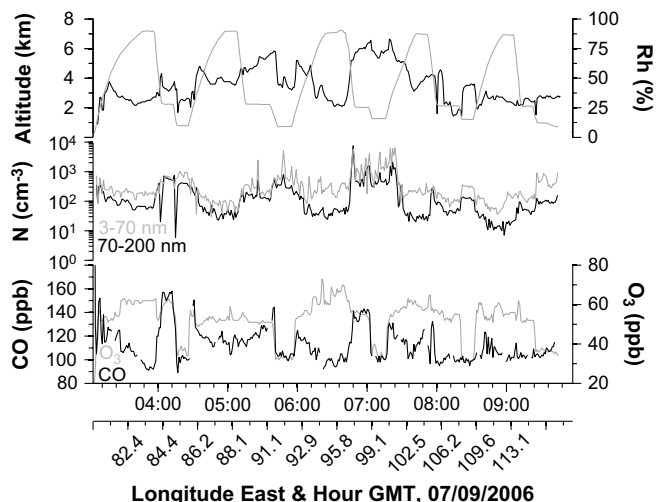


Fig. 5. Time series of relative humidity, altitude, and particle, CO and O₃ concentrations for Flight 5 on 7 September 2006.

BL (identified by high humidity and very low O₃ concentrations); and two occurrences (time between 0600 and 0800) of a stratospheric influence (marked by O₃ concentrations up to 70 ppb) in the upper troposphere, where N₃₋₇₀ is of the order of 100–200 cm⁻³ against N₇₀₋₂₀₀ concentrations of 10–20 cm⁻³. This results in an *R* ratio value of about 10, denoting the influence of new particle formation. Surface tracer CO is very low during these cases, further confirming the isolation from surface sources and suggesting ion-induced or ion-mediated nucleation as a contribution to new particle formation (Lee et al., 2003). In order to test the possibility that we observe export of particles that have nucleated in the BL we have investigated the sensitivity of the *R* ratio to surface emissions influence within a characteristic lifetime of 48 h. Using the FLEX-PART Lagrangian particle dispersion model (Stohl et al., 2005; Paris et al., 2008, results available online at <http://zardoz.nilu.no/~andreas/YAK/>) throughout the September 2006 campaign it was found that on average, in the air sampled above 4 km, less than 1% of the sampled “air parcels” have resided in the lowest 300 m of the troposphere, and hence would have been possibly influenced by surface emissions within the last 48 h. Due to their rather short lifetime, this rules out the vertical transport of particles originally nucleated in the BL to the FT. As a result our case study suggests that fresh particle nucleation occurred in the remote continental upper troposphere, with air well isolated from the BL. This could be attributed to the specific conditions (steep temperature and humidity gradients) in tropopause folds (Young et al., 2007).

3.5. New particle formation in mid-tropospheric cloud outflow: September 2006 case

The aircraft flew over convective clouds between 0750 and 0815 (Fig. 6) at 4 km altitude during Flight 7 (8 September 2006). The convection is reflected by a local increase in relative humidity to ~80%, and in CO concentrations to ~150 ppb, about 50 ppb above tropospheric background concentrations. Immediately past the outflow of the convection, concentrations of N₃₋₇₀ increase by one order of magnitude up to 4000 cm⁻³ while N₇₀₋₂₀₀ increases only up to 700 cm⁻³. As a result the *R* ratio increased from unity at 0800 to about 6. Ruling out size-dependent effects in aerosol load scavenging during convection, this strongly suggests the occurrence of new particle formation in the cloud outflow. According to FLEX-PART up to 90% of the air parcels sampled with highest *R* ratio values had resided in the BL in the last two days prior to sampling,

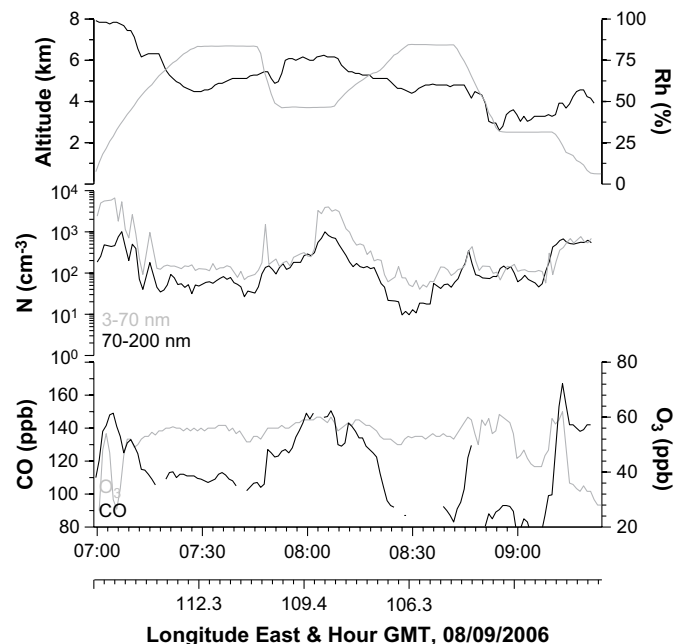


Fig. 6. Time series of relative humidity, altitude, and particle, CO and O₃ concentrations for Flight 7 on 8 September 2006.

confirming the influence of uplifted BL air in the measurement. The contribution of surface tracer such as CO is suggestive of a concomitant uplift of elevated SO₂ or insoluble organic compounds concentrations, potentially participating in the new particle formation event. Twohy et al. (2002) described a similar event although in their case it was observed in the upper troposphere, and fine particle concentrations were much higher.

3.6. Seasonal variation

Fig. 7a and b show respectively the seasonal evolution of N₃₋₇₀ and N₇₀₋₂₀₀ derived from the 5 years of monthly vertical profiles in Novosibirsk (Arshinov and Belan, 2004). Median ultra-fine particle (3–70 nm) concentrations range between ~300 cm⁻³ in summer and 30 cm⁻³ in winter. Fine particles (70–200 nm) range between 10 and 30 cm⁻³ in winter and summer, respectively. Variability of ultra-fine particles is higher in June and July, suggesting a shorter lifetime (Williams et al., 2002). The most important feature of Fig. 7 is the difference in concentration between the BL and the FT. The BL height seasonal variation is clearly visible in the data, except for an apparent drop in particle concentration between June and August. The concentrations are maximal in May and September (with both N₃₋₇₀ and N₇₀₋₂₀₀ > 500 cm⁻³). Kulmala et al. (2004) and Lyubovtseva et al. (2005) observed spring and autumn maxima in nucleation events frequency at the Hyytiälä station in Finland.

Our data do not allow explanation of the summer drop in BL particle concentrations. In summer, conditions typically include a high BL top, higher biogenic precursors emissions (Lathi  re et al., 2006) and higher growth rates of ion clusters and ultra-fine particles (Hirsikko et al., 2005). In Siberia the months of June, July and August account for 50–60% of the annual precipitation (Soja et al., 2004), leading to scavenging in the lower troposphere, and are also the period for highest evapo-transpiration. Lower fine particle concentrations in summer could additionally be influenced by the presence of coarser carbonaceous particle from forest fires (Soja et al., 2004), which could increase the condensational sink.

In the free troposphere there is a significant maximum of ultra-fine particles in July and August, which could be in situ formation.

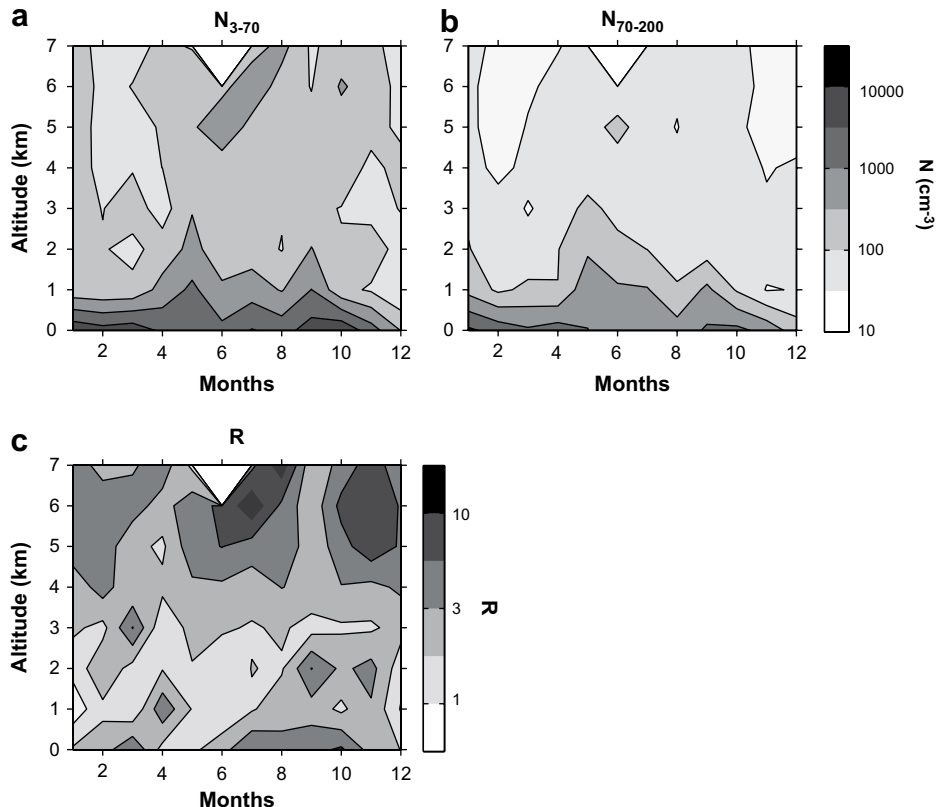


Fig. 7. Seasonal variation of the particles number concentrations and ratio R of ultra-fine (3–70 nm) and fine (70–200 nm) in the troposphere obtained from regular vertical profiles near Novosibirsk. (a) Total concentration in the 3–70 nm range. (b) Total concentration in the 70–200 nm range. (c) R ratio. Greyscales are logarithmic. There were not enough good data in June at 7 km to provide a robust value.

Fig. 7c shows the seasonal and vertical distribution of the R ratio. The FT median size ratio R shows a large variability from 0.1 to 10, possibly reflecting sporadic fresh particle nucleation. This ratio is maximum in July between 5 and 7 km altitude (R inter-quartile range being here 4–12), which suggest summer maximum in situ particle formation, despite wet scavenging by widespread continental convection. A secondary maximum occurs in November, whose origin is unclear. At 5 km there is a minimum in the R ratio in April, which could be compared to our case study of Asian pollutant export to Siberia during dust storms outbreak (Paris et al., 2008). This minimum is therefore possibly explained by an increased condensational sink preventing new particle formation.

Our September case study identified that uplift of BL air by convection in the mid troposphere and UT/LS conditions can enhance ultra-fine particle concentrations in the FT. These conclusions could explain the summer maximum in the R ratio at 5–7 km altitude. Alternatively, this finding could also be explained by the hypothesis by Kulmala et al. (2006b) that deep convection of water-insoluble trace gases to the FT are a possible in situ nucleation source in the absence of sulphuric acid.

4. Summary and conclusions

Fine (7–200 nm) particle number concentration measurement during two intensive airborne campaigns in the troposphere above Siberia between Novosibirsk and Yakutsk (the YAK-AEROSIB project) and monthly vertical profiles near Novosibirsk have been presented. During the campaigns, low concentrations on the order of 200 cm^{-3} have been observed in the free troposphere (FT). Our data are consistent with a possibly widespread gas-to-particle conversion in the free troposphere. In April, no strong excess of fine

particles was found in a filament of polluted air originating from NE China, suggesting a dominant effect of condensational sink.

In September 2006, particle concentrations decreased with altitude to reach $\sim 50\text{--}200 \text{ cm}^{-3}$ at 7 km. N_{70-200} exhibited a gradient steeper than that of N_{3-70} . N_{3-70} accounts for 90% of the total particle concentration in the free troposphere at 6–7 km altitude. Two cases of possible nucleation with relatively high concentrations ($\sim 200 \text{ cm}^{-3}$) and N_{3-70}/N_{70-200} ratios are reported for the September campaign, in the upper troposphere and in cloud outflow in the mid-troposphere. The latter case led to concentrations of $\sim 4000 \text{ cm}^{-3}$ in association with surface emissions as denoted by elevated CO concentrations.

In the seasonal analysis, we found high concentrations ($>1000 \text{ cm}^{-3}$) and N_{3-70}/N_{70-200} ratios in summer in the mid- and upper troposphere, consistent with in situ nucleation. This could be related to either one of the two processes described by our case studies. The summer precipitation maximum, and the high altitude of this maximum could also support the Kulmala et al. (2006b) hypothesis of in situ FT nucleation from water-insoluble precursors. Maximum concentrations in the BL are observed in spring and autumn.

Our aircraft was not equipped to provide high-resolution size distribution or precursors' concentration measurements. Therefore our preliminary results strongly suggest, but cannot diagnose unambiguously, fresh nucleation. Instead, this calls for new, targeted investigation in the clean, continental air shed above Siberia.

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