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# Airborne measurements of nucleation mode particles II: boreal forest nucleation events

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## Abstract

Airborne measurements of nucleation mode aerosol concentrations during nucleation events over the boreal forest of southern Finland are reported. Three case studies are analysis in an attempt to characterise the spatial scales over which these events occur and to identify the source region for particle production. For the cases presented, there is no evidence of nucleation mode particles in the Free Troposphere. Nucleation mode particles are first detected in the surface layer as the nocturnal inversion breaks up and develops into the current-day's new boundary layer. In terms of spatial variability, significant variability in the concentration of nucleation mode particles was observed and was attributed to changes in the topography which comprised a mix of forest canopy and frozen lakes. Measurements over the Gulf of Bothnia indicated no nucleation mode over the sea and confirm that the scale of the events is associated with the boreal forest scale and that the new particles are produced directly above the forest canopy.

## 1 Introduction

The atmospheric aerosol system constitutes perhaps one of the most complex atmospheric systems to elucidate due to the many different formation and growth processes, sources and sinks, interactions with gas-phase chemical cycling of constituents, and interactions with clouds and the hydrological cycle. Furthermore, aerosols contribute to the direct and indirect radiative forcing of the earth's radiative budget and consequently influences climate (IPCC, 2007).

While much attention has been focused on the impact of anthropogenic aerosol emissions on climate and air quality (Hansson and O'Dowd, 2006), the natural aerosol system is of equal importance for two reasons. First, the impact of anthropogenic aerosols can only be quantified once the background natural system has first been quantified and secondly, the natural aerosol system is likely to be involved in feedback processes (Charlson et al., 1987) since their production rates are typically linked to

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biological or ecosystems such as plankton (O'Dowd et al., 2002, 2004) over the ocean and vegetation or forested regions over land where the boreal forest is a significant source of new particles formed through gas to particle conversion processes (Kulmala et al., 2004; O'Dowd et al., 2002; Truvnd et al., 2006).

5 The boreal forest region in southern Finland has provided an ideal natural laboratory to study the processes driving terrestrial new particle formation processes driven by organic vapour emissions and large international projects such as BIOFOR (Kulmala et al., 2001) and QUEST (Laaksonen et al., 2003) have brought multi-disciplinary resources together to elucidate the dominant processes.

10 While the current consensus (Kulmala et al., 2004) on the formation mechanism is that nucleation is driving by sulphuric acid – ammonia cluster nucleation, condensable oxidant products of volatile organic compounds such as terpenes are required to grow the stable embryos into operationally-defined aerosol particles of 3 nm (O'Dowd et al., 2002), and even to sizes larger than 100 nm where they can contribute to direct and indirect radiative effects. The mechanism of production has been significantly elucidated; however, the location of particle formation and the role of meteorology still remains an open question. For example, Raes (1995) have suggested that nucleation and initial growth takes place in the free troposphere and when these new particles are entrained into the boundary layer, they can grow rapidly into larger sizes. One study over forested regions in Germany also reported particle formation above the surface layer followed by downward mixing into the surface layer (Stratmann et al., 2002). However, mixing diagram analysis of production events over the boreal forest supported more the concept that the particle production occurred in the boundary layer (Nilsson et al., 2001).

25 This study reports on a specific component of the QUEST project aimed at elucidating the source region of new particle production over the boreal forest utilising aerosol measurements onboard two light aircraft deployed over the forest canopy.

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## 2 Experimental

The second QUEST intensive field study took place in March–April 2003 at the Hyytiälä research station. In support of the ground based measurements, two research aircraft were deployed to characterise the nucleation events in terms of source regions and spatial scales.

The GTK Twin Otter Geophysics Research Aircraft (O'Dowd et al., 2007) and the Microlight (Junkerman et al., 2005) were used for this airborne study. Meteorological sensors for air temperature, pressure and dew point and radiation were fitted as well as a suite of aerosol sensors. For nucleation mode aerosol measurements, a suite of TSI condensation particle counters (CPCs) were deployed to measure particle concentration at sizes larger than 3 nm, 6 nm and 10 nm. Particles larger than 3 nm were measured using the TSI 3025 ultrafine particle counter while concentrations larger than 6 nm were measured with a modified TSI 3010(m) and particles larger than 10 nm were sampled using a standard TSI 3010 CPC. This configuration of CPCs allows crude size distributions between 3–6 and 6–10 nm to be derived at 1 Hz. The 3025 was connected to a 7 times dilution flow to avoid saturation of counts during the more intense nucleation events. For more detailed size-resolved measurements, a nano-SMPS scanning over 30 s was used. Finally, a PHA-CPC was used to examine nucleation mode aerosol growth in butanol vapour to discriminate between organic or inorganic nucleation mode particle compositions. For a detailed summary, see O'Dowd et al. (2007).

The microlight aircraft of IMK-IFU, a motorized hand-glider (Junkermann, 2005), was equipped with sensors for meteorological parameters, temperature and dew point, different radiation measurements, both up and down welling, and a set of instruments for different sizes of aerosols. Large particles were detected with a Grimm 1.108 aerosol spectrometer and a FSSP, forward scattering spectrometer probe. For the measurement of small size aerosols two condensation particle counters (CPCs) a TSI 3010 with 10 nm cut-off size and a TSI 3025 with 3 nm cut-off were used similar to the installation on the Twin Otter.

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The Hyytiälä SMEAR II research station in southern Finland was the central location for the ground-based measurement component of QUEST and is shown in Fig. 1. Also, the operating region for the flights is outlined by the red box in the figure. The aircraft was located at Tampere airport and the flight operation area was typically upwind and to the north of Tampere and to the west of the Hyytiälä SMEAR II research station. The terrain is densely populated with forest (typically pine and spruce) and the region is also populated with many lakes and one large river seen orientated north-south and to the west of Hyytiälä. An overview of the QUEST project can be found in Laaksonen et al. (2003).

### 3 Results

During the period 24 March to 30 March 2003, 4 strong nucleation events were observed at Hyytiälä. These are illustrated in Fig. 2 where the size and concentration evolution is displayed for particles between 3 nm and 500 nm as measured by the dual DMPS system. These events comprised the QUEST II “golden day events”. In the context of airborne measurements, flights were undertaken on three of these events and are reported on here.

### 4 Synoptic scale meteorological conditions

For the three selected days, the synoptic weather conditions are illustrated in Fig. 3. For the first event presented on 25 March 2003, the area was subject to slack weather conditions with a gentle northerly air flow of the order of  $1\text{--}4\text{ m s}^{-1}$  between the surface and 70 m. On the 26, the area was subjected to more westerly airflow in between a warm front to the east and a cold front to the north and west. Wind speeds on this day were between  $1.5\text{ m s}^{-1}$  at the surface, increasing to  $8\text{ m s}^{-1}$  at 70 m. The cold front passed through on the 27 and had moved over Russia by the 28 leaving the whole

of Finland exposed to a polar air mass with north-westerly airflow and maximum wind speeds of  $8 \text{ ms}^{-1}$  at 70 m. Typically, these conditions lead to slightly unstable air and clear sky. The clear sky leads to strong nocturnal cooling and the development of a stable nocturnal boundary layer which breaks up mid-morning due to increased surface heat flux (Nilsson et al., 2001).

#### 4.1 Case study 1–25 March 2003

The typical flight plan after take-off from Tampere was to transit into the defined starting area, conduct a number of ascents/descents to determine boundary layer structure and the subsequent 10 min horizontal runs are conducted at flights levels determined by the boundary layer structure. Figure 4 illustrated the number of ascents (2), descents (4) and horizontal runs (5) conducted during this flight. Take-off time was 11:30 UTC and landing time was 15:00 UTC.

Figure 5 displays the vertical meteorological profiles (relative humidity and potential temperature) for the descent (left plate) into the operations area and an ascent (right plate) out of the operations area. During the descent, there was a three-layer structure observed from the profiles: a surface layer extends up to 400 m, a second layer resides between 400 and 800 m and the free troposphere at 1000–1100 m. During the ascent later in the flight, the mixed layer height had increased to 1350–1400 m.

Figure 6 shows the aerosol profiles for the same descent and ascent. During the descent, no nucleation mode particles were detected in the Free Troposphere, however, once the aircraft entered the main boundary layer, nucleation mode particles were detected as seen by both the increase in total particle concentration and significant particle concentration differences being observed between the three CPCs measuring at sizes larger than 3, 6 and 10 nm. Peak particle concentrations of  $2 \times 10^4 \text{ cm}^{-3}$  were detected at 400–500 m and the concentration throughout the boundary layer was quite variable. During the ascent out of the area, some 2 h later, particle concentrations were more well-mixed with the total particle concentration had reduced to the order of  $8\,000\text{--}10\,000 \text{ cm}^{-3}$ .

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Figure 7 displays the concentration of particles of sizes from 3–6 nm and 6–10 nm. The highest concentrations ( $>10\,000\text{ cm}^{-3}$ ) of particles in both size intervals were encountered in the mixed layer.

After the initial profiles to characterise the boundary layer structure, four 10 min horizontal runs were undertaken at 30 m, 300 m, 600 m and 900 m (Fig. 8). Also shown is a 10 min run at 1500 m (just above the main inversion). As mentioned previously, the highest concentrations of up to  $2\times 10^4\text{ cm}^{-3}$  were encountered in the lowest levels. One striking result of the horizontal runs is that there is a large degree of variability, suggesting that while the new particle phenomenon is clearly occurring over a large spatial scale, the event is not very homogeneous suggesting. The inhomogeneity is likely to result from the observed inhomogeneities in the local topography (forests and lakes) which can influence the surface heat fluxes driving boundary layer evolution and canopy fluxes of aerosol precursors. A similar degree of variability is seen in the meteorological parameters. Such variability can account for the lack of clear structure seen in the ascents and descents given the horizontal distance transverse during profiles.

#### 4.2 Case study 2–26 March 2003

The second flight conducted was a short flight on the 26 March. The vertical profiles of aerosol concentration and meteorological parameters during the descent into the operating area are shown in Fig. 9. During this flight, the Free Troposphere inversion was located about 1100 m–1400 m (the highest level flown on this day) and the main boundary layer again had a three layer structure. The surface layer, below 600 m, exhibited large concentrations of nucleation mode particles while no nucleation mode particles were seen in the layer from 650–850 m layer or from 850–1100 m. Two horizontal runs were conducted during this flight, one at about 120 m and the other around 400 m. Particle concentrations for both runs are displayed in Fig. 10. During both reciprocal runs, at the start and end of the runs, no nucleation mode is observed; however, in the central area of each run is clearly a nucleation region with peak concentrations at sizes larger than 3 nm of  $30\,000\text{ cm}^{-3}$  at 120 m and concentrations approaching  $25\,000\text{ cm}^{-3}$

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at 400 m. Approximately half the nucleation mode particles resided at sizes between 3 and 6 nm and the other half between 6 nm and 10 nm.

#### 4.3 Case study 3–28 March 2003

The third case study that we focus on was the nucleation event on 28 March 2003. Since one of the triggers of the nucleation events is thought to be the breakdown of the nocturnal boundary layer (Nilsson et al., 2001), precursors have been concentrated, and the subsequent dilution of the aerosol condensation sink as the surface layer mixes up into the complete boundary layer, this flight mission aimed to catch the onset of nucleation as the boundary layer was breaking up. In fact, three flights were conducted using the Twin Otter and the Microlight. The Twin Otter's first flight started at 10:00 UTC and continued until 13:00 UTC when it landed for refuelling at Tampere airport. The second Twin Otter flight was conducted from 15:00–17:00 UTC and the period in between was covered by the micro-light. As a result, we were able to achieve a total of 8 h sampling boundary layer evolution and associated nucleation events.

The flight levels, ascents and descents are shown in Fig. 11. The initial part of the flight conducted of series of surface layer horizontal runs (30 m) followed by an ascent to 1000 m and then a descent. The horizontal runs were started at the same fixed ground position and the ascent and descent were conducted while the aircraft was returning to this fixed ground position to start the next horizontal run.

Figure 12 shows the initial meteorological vertical structure with a well-mixed surface layer extending to 600 m. On this day, the Free Troposphere inversion was also located at the top of the surface layer. The evolution of the boundary layer throughout different times of the day is also shown in the same figure. The main feature is the development of the surface layer up to 800–900 m by 10:00 UTC. Particle concentration during these ascents is also shown in Fig. 12.

During the initial descent, the nucleation mode is most evident at the lowest level (just a few m above the canopy top). As the boundary layer develops, the nucleation more becomes evident throughout the boundary layer and the intensity increases. Peak con-

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centrations of  $30\,000\text{ cm}^{-3}$  were observed during the profile at  $\sim 11:15$  UTC. It should be noted that one should be careful in interpreting the profiles since these profiles cover significant horizontal distances over which variability is seen in both boundary layer structure, and consequently particle concentration.

Particle concentrations during the horizontal runs are shown in Fig. 13 for flight levels of 30, 150, and 300 m at different times throughout the morning. The first series of horizontal runs generally indicate the absence of nucleation mode particles; however, there are brief regions where nucleation is observed in small air parcels. These horizontal runs capture the onset of the nucleation event and illustrate that the nucleation event was just bubbling under as the surface layer expands vertically. As the boundary layer develops, the total particle concentration increases from background of  $2500\text{ cm}^{-3}$ , to  $6000\text{ cm}^{-3}$  as small scale air parcels start to nucleate, to  $25\,000\text{ cm}^{-3}$  as the nucleation event becomes fully developed.

During the latter part of the flight, the flight plan was adapted to evaluate the spatial scale of the nucleation event and, in particular, to evaluate whether or not nucleation mode particles could be observed over the sea or whether they were contained to areas over the forest canopy. The run out to the ocean showed a decrease in nucleation mode particles as one approached the coastline and during the run at 30 m over the sea (which was ice-covered), no nucleation mode particles were observed (Fig. 13), as indicated by the lack of difference in concentration for the 3 CPCs. Thus, these results confirm that the nucleation event is indeed linked to processes occurring over the forest canopy and does not occur over the sea.

#### 4.4 Where does particle production occur?

From the above three flights, there is strong evidence that nucleation occurs first in the surface layer. In fact, we can only say that new, 3 nm particles are first detected in the surface layer and seem to be associated with the break-up of the nocturnal boundary layer. We have compiled all vertical profile data of aerosol concentrations from the 24

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profiles conducted throughout the day on 28 March. These are interpolated to visualise the evolution and aerosol concentration and are shown in Fig. 14. The pattern which emerges is that the first occurrence of elevated particle concentration occurs at the very lowest levels, immediately above the forest canopy and that only later in the day, as the surface layer increases in height, do elevated particle concentrations appear at higher altitudes. The interpolated combination of 20 profiles strongly points to the source of new particles being just above the forest canopy.

To provide a more rigorous analysis of where particles production occurs, we can invoke an analysis tool called conserved-variable mixing diagram analysis. The principal of conserved-variable mixing diagram analysis is that during mixing of two distinct homogeneous air parcels or layers, conserved thermodynamic variables such as potential temperature and total water content will be mixed in a linear manner. Thus, if one plots a scatter plot of the conserved variables, two distinct clusters of points, characteristic of the individual parcel or layer thermodynamic properties, will emerge if there is no mixing between the two air parcels, but, if mixing occurs, a linear mixing line will emerge connecting the two clusters. The position on the mixing line represents the degree of mixing between the two original parcels occurring for each individual data point.

Such an analysis is conducted for the first ascent into the boundary layer, thereby determining the thermodynamic properties of the Free Troposphere and each layer in the boundary layer (Fig. 15). In this analysis, since the data were gathered in cloud-free conditions, water vapour mixing ratio is used instead of total water content. In addition to the descent data being plotted, the thermodynamic properties of the surface layer during the first 30 m run and the 150 m run (i.e. where the first occurrence of new particles were detected are) shown in the same diagram. The scatter plot between potential temperature and total water content indicate that for both horizontal runs, the data points form a distinct cluster and no mixing line connecting this layer to the residual or Free Troposphere layers is observed. By comparison, a mixing line between the residual layer and the Free Troposphere is observed. This indicated that

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there is little mixing between the surface layer and the residual layer during the onset of particle production. Also shown in Fig. 15 is the scatter plot of 3–10 nm particle concentration versus total water content. What is seen is that the particle concentration is independent of water vapour concentration and thus, is not conserved. In other words, while the water vapour mixing ratio remains more or less constant (i.e. minimal mixing), increases in particle concentration is observed in the surface layer.

This analysis strongly suggests that the production of particles does indeed occur in the surface layer before significant mixing has taken place. More so, since we measure 3 nm particles, actual nucleation and growth to 3 nm sizes, the original nuclei must have been formed in the surface layer approximately 30–60 min prior to particle detection.

## 5 Conclusions

Airborne measurements of nucleation mode aerosol particles were conducted over the Boreal forest during the spring (March) of 2003 and during a period of strong nucleation and growth events. Three events are presented with particle concentration increasing from  $\sim 3000 \text{ cm}^{-3}$  to  $>20\,000 \text{ cm}^{-3}$ . The three flights indicate that no particle production is observed in the Free Troposphere and that new particles are first detected in the surface layer about 30 m above the canopy. The vertical and horizontal distribution of new particles demonstrated considerable variability and is thought to be due to variability in land coverage between forests and lakes. Despite the variability over horizontal scales, the production and occurrence of new particles was confined to above the forest canopy as no new particle occurrence was observed upwind of the forest over the sea. Conserved variable mixing diagram analysis lends strong support to the conclusion that new particles are formed close to the forest canopy, in the surface layer, and not in the Free Troposphere, pointing to the forest as being the source of new particles.

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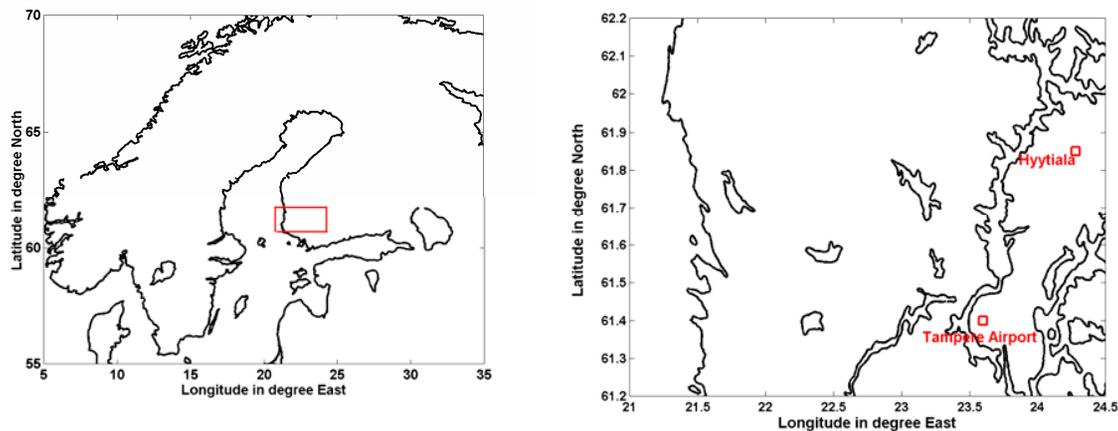
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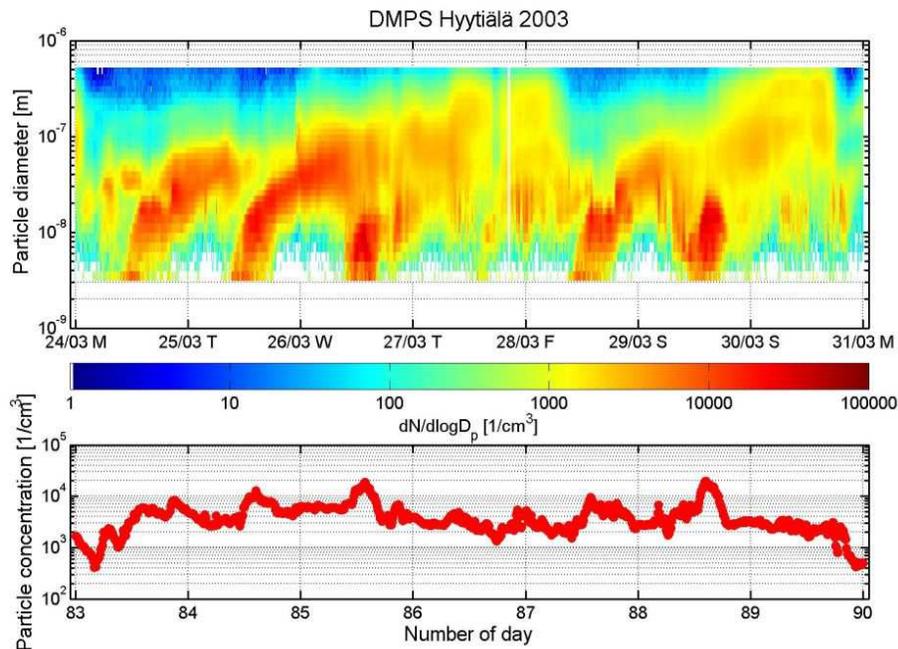


**Fig. 1.** Map of Finland with zoom box over the aircraft operations area.

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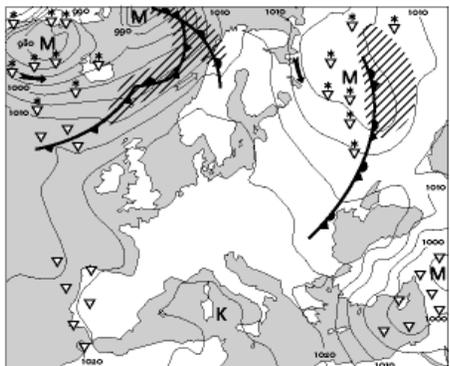
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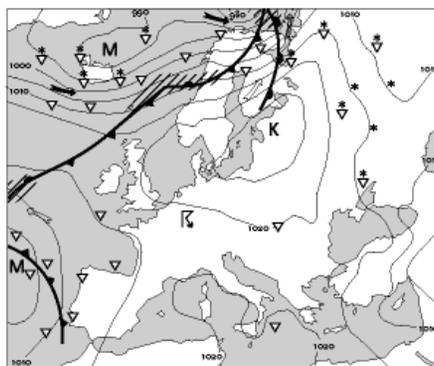
**Fig. 2.** Size distribution evolution during golden day nucleation events from 24 March to 30 March 2003.

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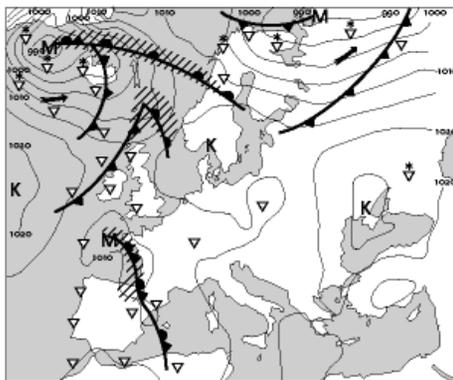
(a)



(b)



(c)



**Fig. 3.** Synoptic conditions on. **(a)** 25 March 2003, **(b)** 26 March 2003 and **(c)** 28 March 2003.

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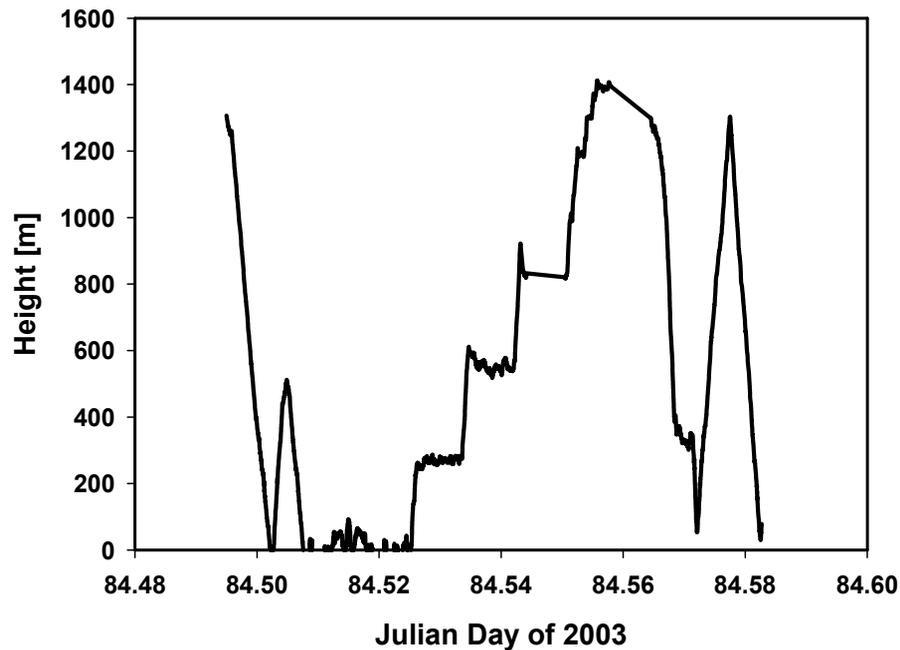
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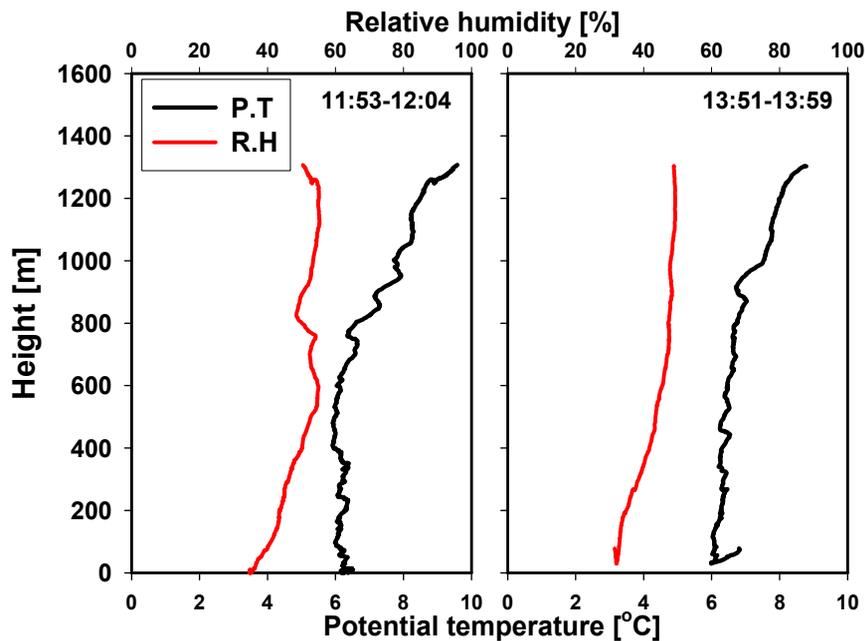
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**Fig. 4.** Flight levels for 25 March 2003.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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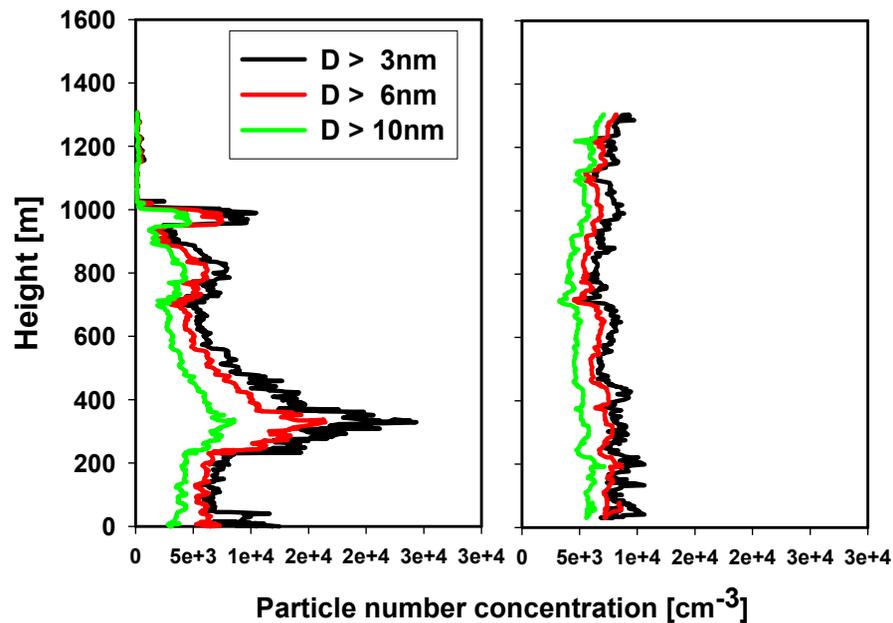


**Fig. 5.** Potential temperature and relative humidity boundary layer structure for 25 March 2003.

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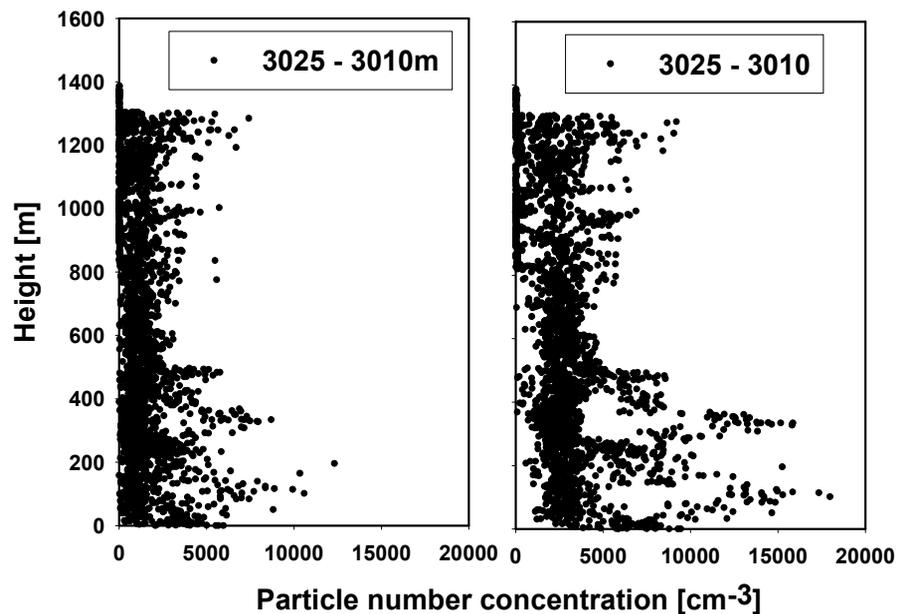


**Fig. 6.** Aerosol boundary layer structure for 25 March 2003. Total number concentration for sizes  $D > 3\text{ nm}$ ,  $6\text{ nm}$  and  $10\text{ nm}$ .

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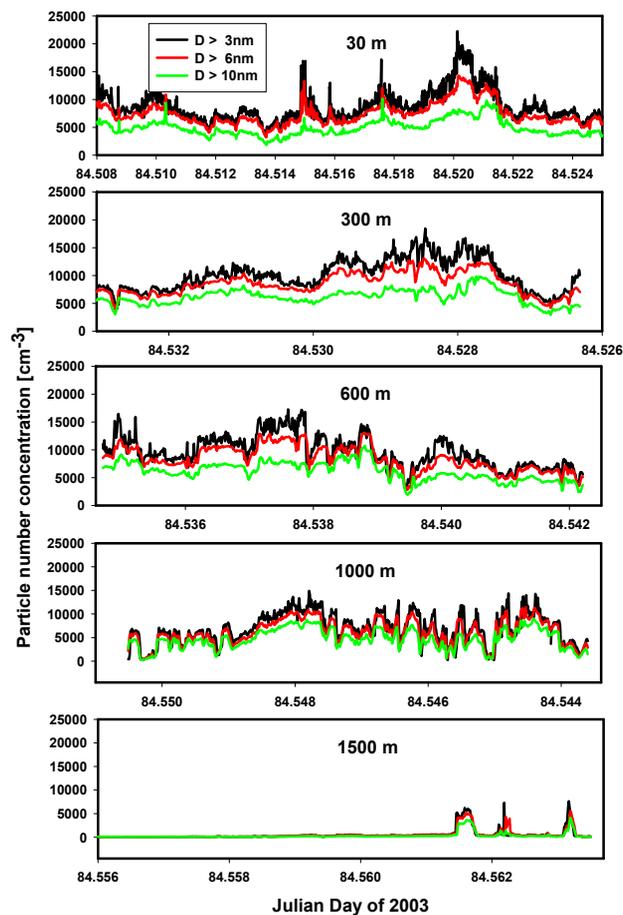


**Fig. 7.** (Left) Aerosol concentration for sizes  $3 \text{ nm} < D < 6 \text{ nm}$  and (Right)  $6 \text{ nm} < D < 10 \text{ nm}$ .

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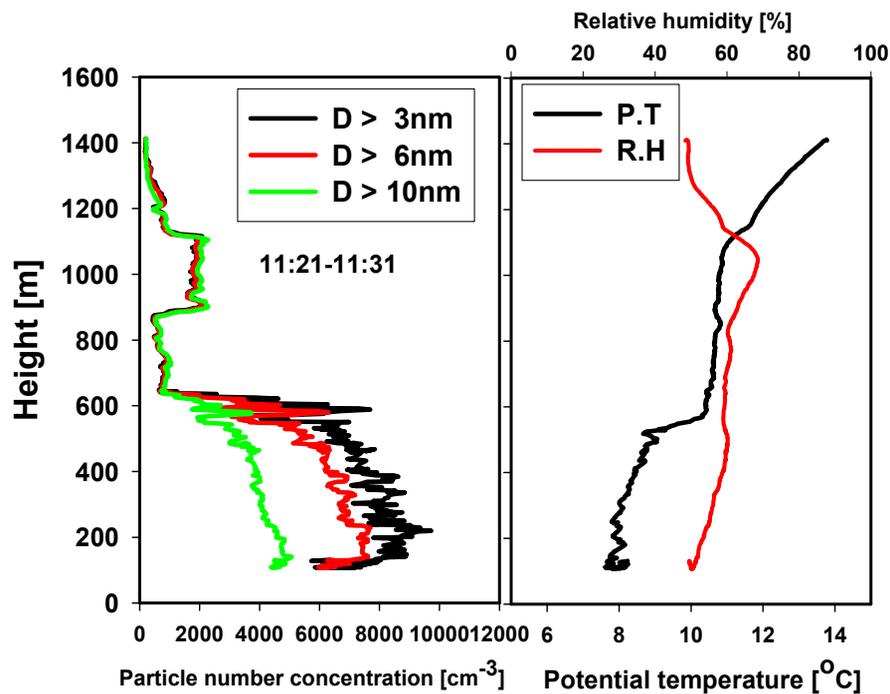


**Fig. 8.** Particle concentrations for 10min horizontal runs at selected altitudes on 25 March 2003.

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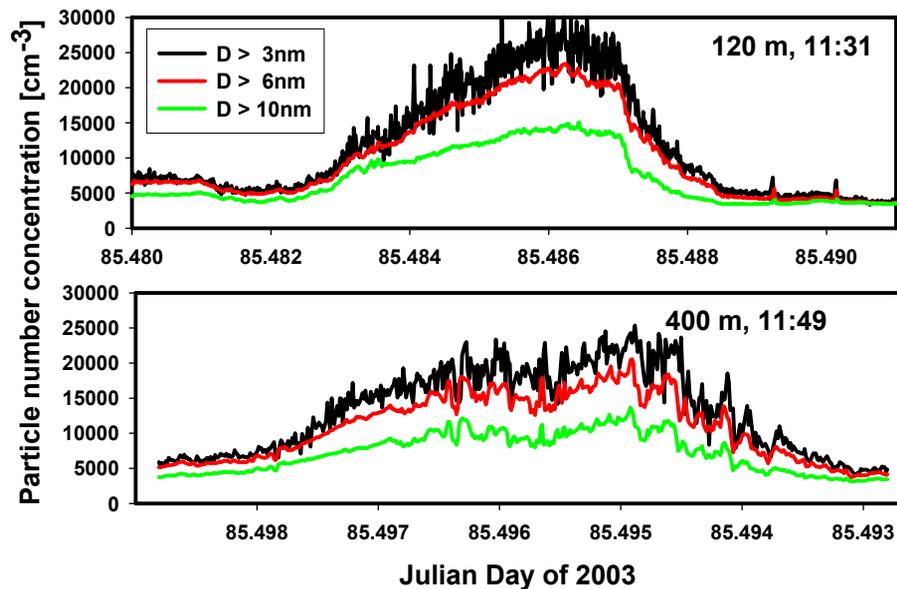


**Fig. 9.** Aerosol potential temperature and relative humidity boundary layer structure on 26 March 2003.

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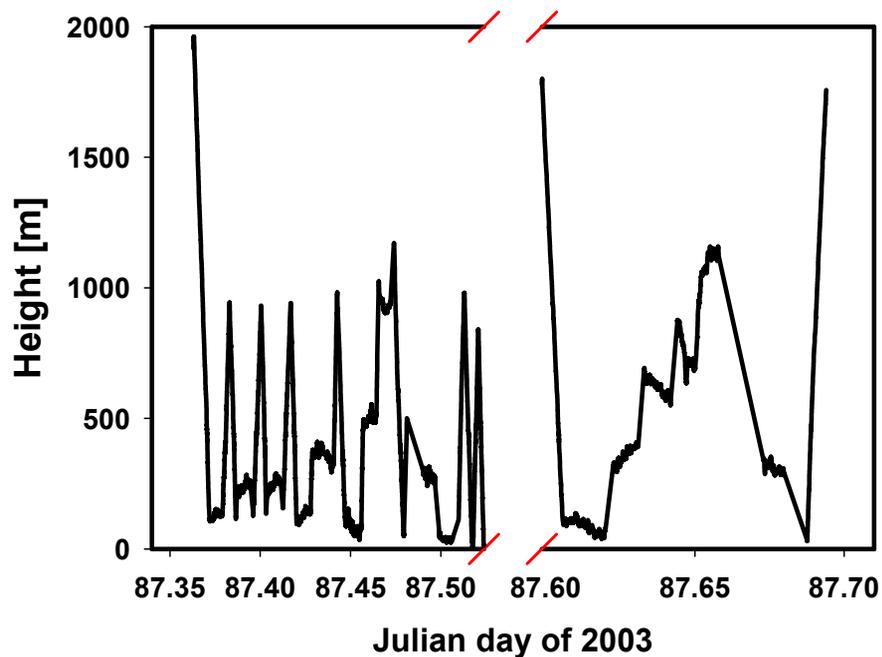


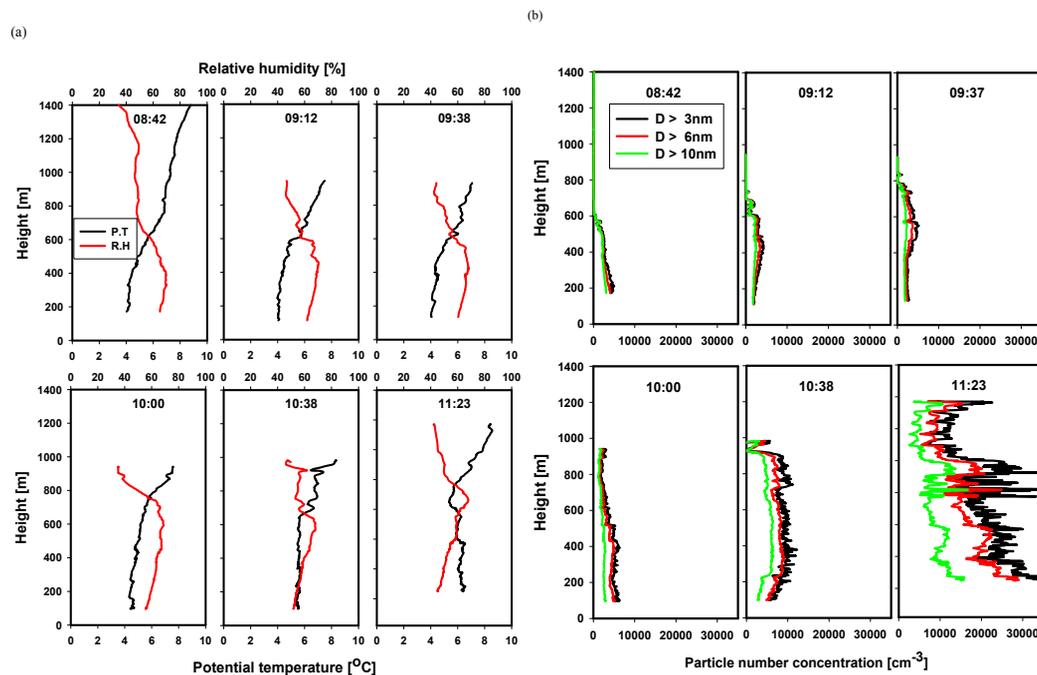
**Fig. 10.** Particle concentration and horizontal variability for two selected flight levels on 26 March 2003.

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**Fig. 11.** Flight levels for 28 March 2003.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



**Fig. 12.** (a) evolution of boundary layer structure, as determined by potential temperature and relative humidity, for 28 March 2003. (b) variation in aerosol vertical structure throughout the morning of 28 March 2003. Local time is also listed in each plot.

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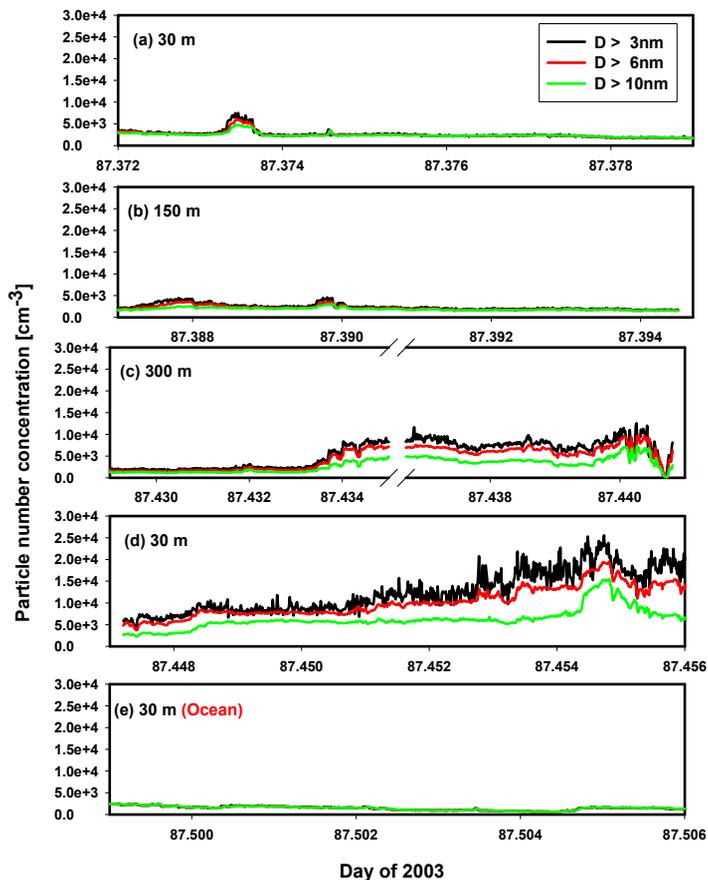
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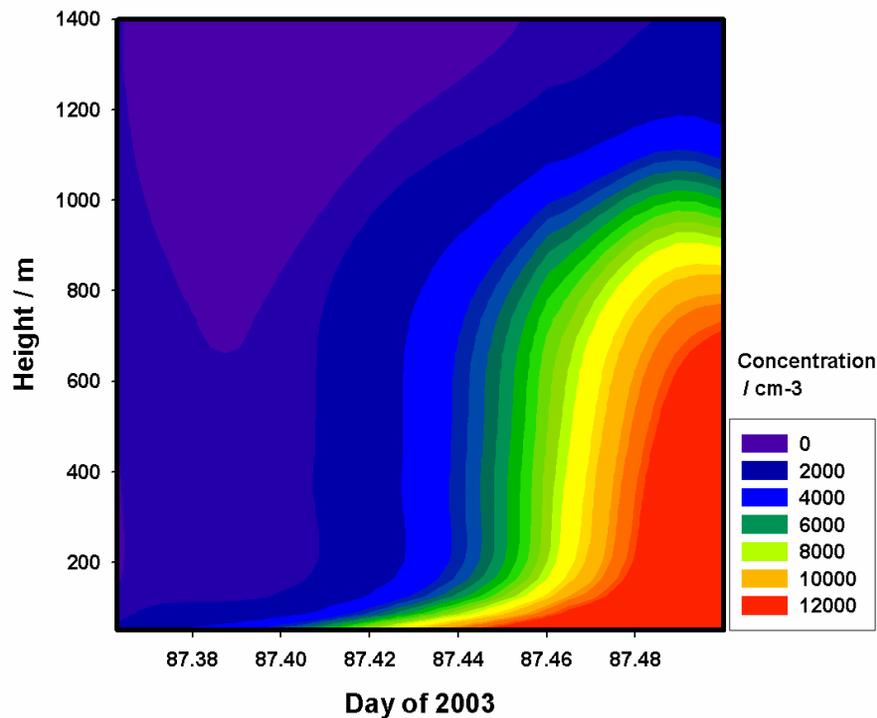


**Fig. 13.** Particle concentration at sizes  $D > 3$ , 6, and 10 nm for horizontal runs at 30, 150 and 300 m over the forest canopy. Also shown is the particle concentration for a 30 m horizontal run over the sea.

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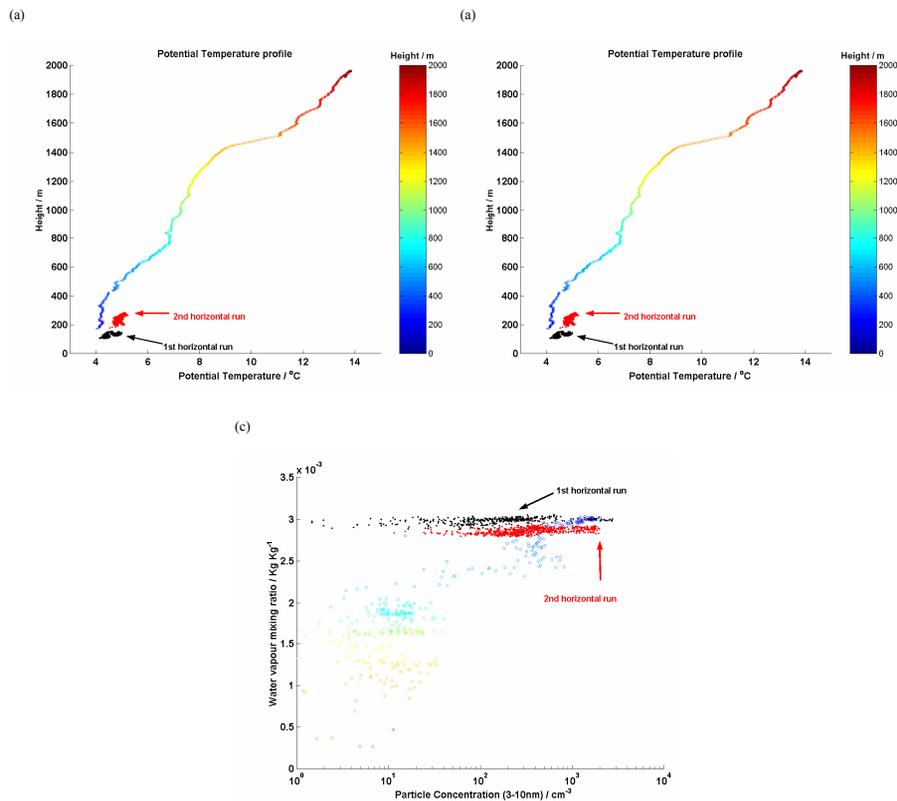


**Fig. 14.** Interpolated particle concentration at  $D > 3$  nm for 24 profiles conducted on 28 March 2003.

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**Fig. 15.** (a) Vertical profile of potential temperature during ascent and first two horizontal runs on 28 March 2003. (b) conserved mixing variable diagram for potential temperature and water vapour mixing ratio (c) mixing diagram analysis for nucleation mode particles (3–10 nm) and water vapour mixing ratio.

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