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Long term particle size distribution measurements at Mount Waliguan, a high-altitude site in inland China

N. Kivekäs¹, J. Sun², M. Zhan², V.-M. Kerminen¹, A. Hyvärinen¹, M. Komppula³, Y. Viisanen¹, N. Hong⁴, Y. Zhang², M. Kulmala⁵, X.-C. Zhang², D. Geer⁴, and H. Lihavainen¹

¹Finnish Meteorological Institute, P.O. Box 503, 00101 Helsinki, Finland

²Key Laboratory for Atmospheric Chemistry of CMA, Center for Atmosphere Watch and Services, Chinese Academy of Meteorological Sciences, Beijing 100081, China

³Finnish Meteorological Institute P.O. Box 1627, 70211 Kuopio, Finland

⁴China GAW Baseline Observatory, Qinghai Meteorological Bureau, Xining 810001, China

⁵University of Helsinki, Dept. Physical Sciences, P.O. Box 64, 00014 Univ. of Helsinki, Finland

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Correspondence to: N. Kivekäs (niku.kivekas@fmi.fi)

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Particle number size distributions in size range 12–570 nm were measured continuously at Mount Waliguan, a remote mountain-top station in inland China. The station is located at the altitude of 3816 m above the sea level, and some 600 m above the surrounding area. The measurement period lasted from September 2005 to May 2007. The measurements were verified with independent CPC measurements at the same site. The average particle concentration in ambient conditions was 2040 cm^{-3} , which is higher than the values measured at similar altitude in other regions of the world. On average, the Aitken mode contributed to roughly half of the particle number concentration. The concentrations were found to be higher during the summer than during the winter. The diurnal variation was also investigated and a clear pattern was found for the nucleation mode during all seasons, so that the nucleation mode particle concentration increased in the afternoon. The same pattern was visible in the Aitken mode during the summer, whereas the accumulation mode did not show any level of diurnal pattern. Excluding the nucleation mode, the average day-time particle concentrations were not significantly higher than those measured at night-time, indicating no systematic pattern of change between planetary boundary layer conditions and free troposphere conditions. In air masses coming from east, the number concentration of particles was higher than in other air masses, which indicates that the air mass might be affected by anthropogenic pollution east of the station. Also other factors, such as active new-particle formation, keep aerosol number concentrations high in the area.

1 Introduction

In order to improve our understanding on the climatic and other influences of atmospheric aerosols, we need to have detailed information on the physical, chemical and optical properties of these aerosols (e.g. Diner et al., 2004). Since practically all aerosol properties vary greatly in both time and space, multiple approaches to tackle the prob-

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lem are required. These include large-scale model simulations, usage of various remote sensing techniques and in-situ measurements (e.g. Yu et al., 2006; Ghan and Schwartz, 2007; Remer et al., 2008; Kulmala et al., 2008). Of these, continuous in-situ measurements are currently the only means to get both versatile and accurate aerosol data with sufficient temporal resolution.

Most long-term aerosol measurements have been conducted in cities and rural areas in Europe and North America in locations that are relatively easy to access. The majority of such measurements have been based on the aerosol mass concentration, because this quantity is normally used in air quality standards. Particle number concentration or size distribution measurements are fewer in number. Very few long-term aerosol measurements have been made in South America, Africa or East Asia, and the conducted studies have usually focused on large cities in which air quality is the major issue. Any long term measurements in rural or background areas in those regions are rare, even though it is known that some large areas are exposed high level of aerosol pollution (East Asian brown cloud) (Lelieveld et al., 2001; Nakajima et al., 2007; Ramanathan et al., 2007a). Such high aerosol levels affect both climate and human health in those areas (Pandey et al., 2005; Lau and Kim, 2006; Oanh et al., 2006; Pathirana et al., 2007; Nakajima et al., 2007; Ramanathan et al., 2007b).

Continuous aerosol measurements have mostly been carried out at low altitudes. This is logical, as people tend to live in low altitude areas, and convenient, as stations are easier to be built and operated there. However, low-altitude measurements are easily affected by local aerosol sources and small-scale meteorological patterns. Therefore regional and large-scale concentration level of aerosol particles can be observed more reliably in measurements conducted at high altitudes.

High-altitude aerosol measurements can be made by using basically three different approaches. The first one is setting the instruments in aircrafts and then flying a pre-determined route going usually back and forth the same line in different altitudes (e.g. Reus et al., 2000; Keil and Wendisch, 2000). Another technique is to set the instruments in a hot air balloon, and fly it up and down for a couple of times, or to

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follow the same air parcel for some time as the balloon flies with the wind (Tobo et al., 2007; Laakso et al., 2007). Low-cost and low-weight aerosol instruments can also be attached to weather balloons to obtain vertical profiles of the aerosol properties in the atmosphere (Hoffmann, 1993). The only high-altitude measurement technique that allows continuous long-term measurements is to use a measurement site that is located high on a mountain, preferably on top, or in a high mountain saddle (Weber et al., 1996; Nyeki et al., 1998a).

The mountain top measurement data around the world have revealed quite variable aerosol concentrations both with the location and season. The boundary-layer air and free-tropospheric air can also in some cases be identified and studied separately, in which case we get information on the temporal pattern how these two types of air change around the mountain (Nyeki et al., 1998b; Nishita et al., 2008; Shaw, 2007).

In this work we have measured and analyzed almost two years of continuous aerosol size distribution and number concentration data from a mountain top observatory in central China. The objectives of these measurements have been i) to investigate the general level and seasonal variation of particle size distribution and number concentration in Asian continental background air, ii) to study whether the East-Asian brown cloud extends as far into inland China as Mount Waliguan, and iii) to find out whether differences in aerosol size distribution in planetary boundary layer air and in free troposphere air can be seen at Mount Waliguan.

2 Materials and methods

2.1 Measurement site

The Waliguan Baseline Observatory (36°17' N, 100°54' E, 3816 m a.s.l.) is one of 24 baseline observatories of World Meteorological Organization (WMO) Global Atmosphere Watch (GAW), situated on the top of Mt. Waliguan at the edge of northeastern part of the Qinghai-Xizang (Tibet) Plateau in a remote region of western China. The

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station is relatively isolated from major industrial sources and populated centers, locating about 90 km away from the Qinghai provincial capital Xining to the east. The closest major settlement is Qiapuqia with a population of 30 000, located 30 km to the west. The closest cities with more than 1 000 000 inhabitants are located east from the station: Xining at a distance of about 90 km and Lanzhou at the distance of 260 km.

The mountain rises about 600 m from the plains in the north and 1300 m from the Longyangxia reservoir lake in the south, some 12 km from the station. The surrounding area maintains its natural environment of sparse vegetation along with arid and semi-arid grassland and some desert regions. The population density within 25 km from the station is less than 6 people per km². Yak and sheep grazing is the main activity during summer, with small agricultural regions located in the lower valley area.

The annual average temperature is -0.9°C with a relatively strong seasonal variation. The monthly average temperatures vary from -10°C in mid-winter to 10°C in summer. The wind direction is mostly from east in summer and from west in winter. Yearly average wind speed is 4.6 m/s. The annual average precipitation is around 350 mm, with most of it occurring in summer (Zhou et al., 2004).

2.2 Instrumentation

As a part of the GAW network, the Mount Waliguan station is a platform for many different measurements. These measurements include meteorological parameters, greenhouse gases, reactive gases, surface ozone, precipitation chemistry and some aerosol parameters (GAWSIS). The aerosol light absorption coefficient has been measured with aethalometer and aerosol multiwavelength optical depth with pyrroheliometer since the establishment of the station in 1994. Besides these, the aerosol number size distribution has been measured during 2005–2007 with a differential mobility particle sizer (DMPS) and aerosol number concentration during 2006 with condensation particle counter (CPC).

The DMPS system used at Mount Waliguan consists of a 28-cm-long Hauke-type differential mobility analyzer (DMA) with a closed loop sheath flow arrangement, and

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a condensation particle counter (TSI model 3010) (Komppula et al., 2003). The DMPS measures particles in 30 discrete size classes, one size class at the time. The voltages of the DMA were set so that the geometric means of the size classes range from 10.0 nm to 500 nm, and are distributed evenly in logarithmic scale. The analysis of the raw data was based on programs and routines developed in the University of Helsinki (Aalto et al., 2001). However, the number concentrations of the two smallest size classes were found unreliable and were therefore removed, leaving the geometric mean diameter of the smallest size class to be 13.1 nm. If the logarithmic widths of the size classes at the end of the scale are assumed to be the same with the other size classes, this leaves us with capability to measure over the mobility diameter range of 12–570 nm. The DMPS system goes stepwise through the whole measured size range in approximately five minutes.

The DMPS instrumentation was established at the station on 19 August 2005 and was operational from there on to 9 May 2007. During this period there were two longer gaps when the measurements were not running due to instrumentation failures. These gap periods were from 16 January to 7 April 2006 and from 26 June to 10 September 2006.

The particle number concentration above the 10 nm diameter was also measured with a butanol based condensation particle counter (TSI model 3010). The upper limit of the CPC measurement size range is determined by the bends of the inlet channel at the station and inside the device. However, since the number concentration of supermicron particles is in almost always negligible compared to that of submicron size fraction, the CPC and DMPS systems should give approximately the same particle number concentration. This allows us to use the CPC data to confirm the DMPS is working correctly and vice versa. The CPC measurements were also suffering from problems at the station, and there are gaps in the CPC data as well. The CPC measurement data starts from 15 January and ends in 25 November 2006. The longest gap in the data was 18 May–13 August 2006.

FLEXTRA back trajectories from the site were calculated for each three hours. The

trajectories followed the air mass arriving path starting 120 h before the air mass reached Mount Waliguan. The trajectories consisted of longitude and latitude of the air parcel, as well as altitude from sea level and from the ground.

Meteorological parameters available for this study were the temperature, relative humidity, wind speed and wind direction. All the meteorological parameters were given as five-minute averages.

2.3 Data processing

The DMPS data was examined and bad data was removed, including the periods of clear local anthropogenic effects from the station or from the road to the station. These periods were characterized by sharp peaks of Aitken and accumulation mode particle number concentrations in the data. The total particle number concentration from each size distribution in the DMPS measurements was calculated as the sum of all size bins. All the DMPS data and meteorological data were converted into one-hour averages for data handling reasons. If there were less than four data points for any individual hour, the data for that hour was neglected. Also the CPC data was converted into hourly averages.

For each hourly-average size distribution, a number concentration was calculated separately for nucleation ($12 \text{ nm} < d_p < 21 \text{ nm}$), Aitken ($21 \text{ nm} < d_p < 95 \text{ nm}$) and accumulation ($95 < d_p < 570 \text{ nm}$) mode particles. The hourly average number size distributions were also normalized and classified into five groups according to the shape of the normalized size distribution. These distribution types were characterized by a dominating nucleation mode (type 1), grown nucleation mode (type 2), dominating Aitken mode (type 3), dominating Aitken and accumulation modes (type 4) and dispersed distribution with the Aitken mode being the dominant one and nucleation and accumulation modes being roughly equal (type 5). The five distribution types are presented in Fig. 1.

Trajectories were combined with the data in such a way that each trajectory was attached to the size distribution data at the hour corresponding to the arrival of the air parcel at Mount Waliguan. The same trajectory was also attached to the size distri-

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5 bution data of the previous hour and the following hour. Four sectors were used to
separate the trajectories into different groups (Fig. 2). These sectors were 315°–45°
(sector 1, northern grasslands), 45°–165° (sector 2, densely populated areas of China),
165°–270° (sector 3, highland areas) and 270°–315° (sector 4, western deserts). The
fraction of the time that each trajectory had spent in each sector was calculated. The
same calculation was performed for the air mass height, dividing the air masses into
high-altitude air ($p < 550$ hPa) and low-altitude air ($p > 550$ hPa). This refers to altitude of
approximately 4800 m a.s.l. (Brasseur et al., 1999). The air mass location (sector and
height) at one day, three days and five days before arrival at Mount Waliguan was also
10 investigated.

The diurnal pattern of aerosol number concentration of each mode was investi-
gated separately for spring (March–May), summer (June–August), fall (September–
November), and winter (December–February). For all seasons, the seasonal-average
number concentration of particles in each mode was calculated for each hour of the
15 day.

3 Results

3.1 General

In hourly average data, the ratio in the total particle number concentrations between
the CPC and DMPS was, on average, 0.96. In 50% of the cases, this value was be-
20 tween 0.85 and 1.18, and in 80% of the cases it was between 0.79 and 1.49 (Fig. 3).
However, the ratio occasionally reached values close to 5. These extreme cases were
typically observed when a large fraction of the particles measured by DMPS were in the
nucleation mode ($d_p < 21$ nm). This indicates that the counting efficiency of the mea-
surement devices is different for particles with a small diameter. The results presented
25 later in this paper are based on the data measured with the DMPS, unless otherwise
mentioned. The concentrations reported here are given at ambient pressure, unless

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otherwise mentioned.

The average number concentration of all (12–570 nm) particles during the whole measurement period was 2040 cm^{-3} at ambient pressure. The average concentrations of particles in each mode were 570 cm^{-3} for the nucleation mode ($12 \text{ nm} < d_p < 21 \text{ nm}$),
5 1060 cm^{-3} for Aitken mode ($21 \text{ nm} < d_p < 95 \text{ nm}$) and 430 cm^{-3} for accumulation mode ($95 \text{ nm} < d_p < 570 \text{ nm}$). The corresponding median values were 1390 cm^{-3} , 290 cm^{-3} , 690 cm^{-3} and 330 cm^{-3} . The average Aitken to accumulation mode particle number concentration ratio was 2.65.

The normalized size distributions were classified into five categories, as explained in the data processing section (Fig. 1). The categories follow the growth of new particles from recent nucleation (distribution type 1) to grown nucleation mode (type 2) and to Aitken mode (type 3). The type 4 distribution can also be reached as the particles grow in size, but it might contain a significant portion of primary particles as well. The type 5
10 distribution is more flat than the other ones, and also in most occasions lower in total number concentration. Such a distribution can be reached by aging the air mass and mixing different types of air. It can also be seen as a typical background distribution before a new particle formation or primary particle emission happens.

3.2 Comparison to other measurements

There are not many long-term aerosol measurements conducted in rural areas in East Asia, nor are there many measurements at such high altitude either. The best measurements to compare the results are those conducted at similar altitude (3454 m) at Jungfrauoch in the Swiss alps (Weingartner et al., 1999; Nyeki et al., 1998b), those by Nishita et al. (2007) at Mount Norikura (2770 m) in Japan, those by Shaw (2007) at the top of Mount Lemnon (2790 m) in Arizona, USA and several measurements at the
20 Himalayan region in India and Nepal.

When comparing the particle number concentrations measured at different altitudes, the measured values need to be calculated to the same pressure level to be compa-
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5 rable. In this study we have used the sea level pressure 1013 hPa. At this pressure the average particle number concentration at Mount Waliguan are 3280 cm^{-3} for the total particle number concentration, 920 cm^{-3} for the nucleation mode, 1710 cm^{-3} for Aitken mode and 690 cm^{-3} for accumulation mode. All of the values of the other measurements presented here have been either reported in sea-level pressure, or have been calculated into it based on reported average pressure at the measurement site, or based on the measurement site height, using a formula presented in Brasseur et al. (1999).

10 The general level of submicron aerosol number concentration at Mount Waliguan was significantly higher than that measured at Jungfraujoch. At Jungfraujoch the annual average number concentration of particles (10–750 nm) was 900 cm^{-3} (Weingartner et al., 1999), calculated by averaging the data over a period of one month and then fitting two log-normal modes to the averaged size distribution. At Mount Norikura the measurements covered the particle size range below 9–300 nm, and the measurements were conducted in late summer and early fall. The reported median number concentrations in that size range at Mount Norikura were $300\text{--}1300 \text{ cm}^{-3}$, depending on the type of air mass (Nishita et al., 2007). These values are closer to those measured by us, but still smaller. Shaw (2007) measured mean particle number concentration of 2000 cm^{-3} in the size range (12–390 nm) during late winter and spring (February–May) at night time, when the conditions are expected to represent the free troposphere. The data processing method used by Shaw (2007) was the same as used by Weingartner at Jungfraujoch (Weingartner et al., 1999). The daytime particle number concentrations (that represent the planetary boundary layer conditions) measured by Shaw (2007) were clearly higher than the night-time values, being more similar to the mean and median values found in this study.

25 Our measurements are best comparable those conducted by Komppula et al. (2008) in Mukteshwar at the height of 2180 m above sea level in the foothills of the Indian Himalayas. These measurements were carried out at the same time, and with a very similar instrumentation, as the measurements at Mount Waliguan. Komppula et al. (2008)

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reported an average particle (10–800 nm) number concentration of 3420 cm^{-3} . Ga-
jaranda et al. (2005) measured particle concentrations during years 1996–2003 at
three measuring sites at different altitudes (1150–2530 m above sea level) in the In-
dian Himalayas. They reported average particle concentrations (1–1000 nm) between
980 and 3450 cm^{-3} , being highest at the site located at lowest altitude and lowest at
the one located at highest altitude. The higher the altitude was, the lower was the
average particle number concentration. Bonasoni et al. (2008) reported an average
value of 880 cm^{-3} for the particle (10–500 nm) number concentration measured at the
Nepalese ABC-Pyramid observatory in the Khumbu valley in Nepalese Himalayas at
the height of 5079 m above sea level.

The total particle number concentration can be affected by primary particle emis-
sions as well as formation of new particles from gaseous precursors. A more relevant
quantity for estimating the effect of anthropogenic activity is the number concentration
of accumulation mode particles (N_{acc}). The reported diameter limits between Aitken
and accumulation mode particles are usually 90–100 nm, varying from one publication
to another. Concerning high-altitude sites, Shaw (2007) reported N_{acc} of 260 cm^{-3} at
Mount Lemnon (in free troposphere conditions) and Weingartner et al. (1999) a value
of 220 cm^{-3} at Jungfaujoch (average over all cases). At Mount Norikura Nishita et
al. (2007) measured values of N_{acc} around 100 cm^{-3} when the air mass had been af-
fected by recent precipitation, and $350\text{--}400\text{ cm}^{-3}$ when no precipitation had altered the
air mass during the previous days. For the lower altitude stations the average N_{acc}
values vary from 230 cm^{-3} (background) to 580 cm^{-3} (rural) to 1650 cm^{-3} (urban) in
Finland (Laakso et al., 2003; Dal Maso et al., 2008). In some megacities average val-
ues as up to $10\,000\text{ cm}^{-3}$ (Beijing) (Wehner et al., 2008) and $25\,000\text{ cm}^{-3}$ (New Delhi)
(Mönkkönen et al., 2005) have been reported for N_{acc} . In Mount Waliguan, the value of
 N_{acc} averaged 692 cm^{-3} and varied with the air mass direction. The lowest average val-
ues of N_{acc} were 362 cm^{-3} in western air masses and highest 606 cm^{-3} in eastern air
masses. These values would be expectable for a rural site at low altitudes, but higher
than the values measured at other high-altitude sites. This was the case especially in

the air masses coming from east to Mount Waliguan.

3.3 Temporal variation

Although there were major breaks in the particle number concentration measurements, some seasonal variation could be observed from the data (Fig. 4). The particle number concentration seemed to be higher during the summer months and lower during the rest of the year. There was also a long period with high particle number concentrations in December 2005 and January 2006, but no similar pattern was found in the next winter. The daily-average particle number concentration (measured with DMPS) varied from 500 to 6800 cm⁻³, while the hourly averages varied from 140 to 17 700 cm⁻³. The monthly-median particle number concentrations derived from the combined DMPS and CPC data varied from 1130 cm⁻³ (March) to 1920 cm⁻³ (June), the corresponding mean values being equal to 1520 and 2540 cm⁻³, respectively. The period with high particle number concentration during the winter 2005–2006 lifted the December and January average and median particle concentrations close to those measured in June. There was no aerosol data from the month of July because of the measurement period and gaps in the measurements.

During all seasons, there was a diurnal pattern in the number concentration of nucleation mode particles (Fig. 5), with an increase between the noon and around 17:00 Beijing time and a gradual decrease during the rest of the day. The Beijing time is used here because it is the official time of all China. The local solar time is 1 h and 20 min behind Beijing time. The increase in nucleation mode particle number concentration is probably caused by the frequent new-particle formation events, taking usually place earlier during the day like observed all around the world (Kulmala et al., 2004). During winter the phenomenon was not as clear as during the other seasons. The influence of nucleation on the number concentration of Aitken mode particles was clearly visible in the summer data (Fig. 6). The increase in Aitken mode particle number concentration took place a few hours later than in nucleation mode, as it takes time for the particles to grow into Aitken mode size. During the other seasons the new particles did not

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grow enough, or the new particle formation events did not occur frequently enough, to be seen in the seasonal-average diurnal pattern of the Aitken mode particle number concentration. In accumulation mode no diurnal patterns were seen in any season. Such mid-day maximum of accumulation mode particles as observed by Komppula et al. (2008) and Gajaranda et al. (2005) was not seen at Mount Waliguan.

Several studies conducted at mountain sites have shown a diurnal pattern of changes between air coming from the planetary boundary layer (PBL) during the day and from the free troposphere (FT) during the night (Nyeki et al., 1998b; Shaw, 2007; Nishita et al., 2008). In such a pattern, the particle concentrations are typically significantly higher in the PBL air during the day than in the FT air during the night. The division of observations into these categories can be done in several different ways. Shaw (2007) compared the difference in the potential temperature between mountain observations and observations conducted at the mountain base. Nyeki et al. (1998b) and Nishita et al. (2008) used synoptic or local meteorological conditions as criteria for selection. Besides these methods, also water vapor pressure and concentrations of some relatively long-lived trace gases produced in PBL, such as CO, could be useful criteria when looking at the type of air. Also aerosol particle size distribution in FT is expected to be more concentrated in the Aitken mode than in the PBL as there are fewer sources for accumulation mode particles in the free troposphere.

No trace gas data were available for this study, even though some trace gases are measured at the site. Neither were there other measurement sites at the mountain base, from which one could obtain the temperature for calculating the potential temperature difference. Water vapor pressure was investigated as a potential criterion for pattern in changes between PBL and FT air. The water is expected to enter the air mostly by evaporation from the surface, especially since there is a large lake in the vicinity of the measurement station. Water vapor was calculated from RH and T according to Buck (1981). Rapid changes in water vapor pressure were compared to rapid changes in the aerosol size distribution. Although there were sudden changes in both of these parameters, they were not found to coincide with each other. The typical

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diurnal pattern of water vapor pressure was actually quite opposite to that expected: when the temperature rose during the day, there was typically a decrease in the water vapor pressure.

Average and median particle number concentration of each mode for afternoon daytime (14:00–18:00 Beijing time) values and nighttime (02:00–06:00 Beijing time) values was also calculated for each season. The ratios between the daytime and nighttime values were also calculated in order to obtain the differences in the air masses (Table 2). This ratio for average values of nucleation mode particles were high throughout the year (1.75 in winter and 2.25–2.58 during other seasons). Similar behavior was not found when median values were used. The ratio of the median values varied from 0.89 (summer) to 1.25 (fall). These values indicate that the high average values come from individual very high nucleation mode particle number concentrations, probably resulting from strong new particle formation events during daytime. The Aitken mode particle number concentrations were typically a bit higher during nighttime than daytime, especially during summer, when the ratios of both average and median values were below 0.7. The accumulation mode particle number concentration during daytime was within $\pm 10\%$ of that during the nighttime almost throughout the year. These findings indicate that there is no systematic pattern of changes between PBL air and FT air at the site.

3.4 Trajectory analysis

The trajectory analysis of the data revealed that in almost two thirds of the cases, the air mass had spent more than 50% of the trajectory time in the western sector, even though it was the narrowest of all the sectors. In the remaining one third of the cases, the air masses were distributed roughly equally between the three other sectors. In only 7% of the cases did the air mass spend not more than half of the five day time in any of the sectors. When the same analysis was made according to the air mass location 24 h before arriving to the station, the dominance of the western sector was not that high, yet still more than 50% of the cases.

When the location of air mass 24 h before arriving at the station was used as the clas-

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sification criteria, particle number concentrations in air masses coming from the more densely populated eastern sector were higher than in other air masses (Table 1). Eastern air masses were also typically associated with particle number-size distributions of types 3 and 4 (see data processing). Distribution types 1 and 2 were significantly less abundant in the eastern air masses than in the other ones, indicating low frequency of new particle formation events. Air masses coming from south and west were more often associated with distribution types 2 and 5. Western air masses were associated with the lowest particle number concentrations (Fig. 7).

When the same analysis was made using the trajectories that had spent more than 50% of the 5-day time in a sector as the classification criterion, a similar pattern was found. In this way, however, the eastern air masses were not so clearly different from the northern and southern air masses. As the usual air mass moving direction in the area during the measurement period was from west to east, the air masses arriving at the site from east had often spent a significant portion of the trajectory time in other sectors, and only the last day or two in the eastern sector. By this way many of the trajectories arriving from the east got classified into some other sector. It also means that the eastern air masses were more probably affected by regional pollution sources (i.e. Xining, Lanzhou), rather than sources in the densely-populated coastal regions in China.

From here on, the trajectory analysis is based on the air mass location 24 h before arriving at Mount Waliguan. The average Aitken to accumulation mode particle number concentration ratio varied from 2.6 to 3.2 between the air masses. The highest values were obtained in air masses coming from west, and lowest values in eastern air masses. These values are very different from the results of Komppula et al. (2008) at Indian Himalayas, where the ratio was about unity. The ratios observed at Mount Waliguan are more similar to those observed at sites with much lower particle number concentrations. Typically, the Aitken to accumulation mode particle number concentration ratio is above unity when the particles are produced within the last couple of days. In more aged air masses this ratio is usually is around or below unity, as both particle

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growth and removal processes shape the distribution towards a dominating accumulation mode. There Aitken mode particles can be formed by combustion processes or from gaseous precursors, either natural or anthropogenic.

When a running 24 h average was used, both Aitken and accumulation mode number concentrations correlated weakly ($R^2 \approx 0.2-0.3$) with the time fraction the air mass had spent in the eastern sector, and anticorrelated with similar strength with the time fraction in western sector. The nucleation mode fraction (number concentration of nucleation mode particles divided by the number concentration of all measured particles) behaved in the opposite way. The correlations were a bit stronger when the number-size distributions were normalized. Other correlations between the modal number concentrations and air masses were not significant. The observed correlations support the interpretation that new particles are produced preferably in air masses arriving from the western sector. There is also some support for the long range transport of anthropogenic pollution from the east, but since the typical Aitken mode concentration is also high in eastern air masses, the air is more probably affected by regional sources within a few hundred km from the station.

4 Conclusions

Aerosol number-size distributions were measured continuously with a DMPS at Mount Waliguan observatory, inland China, at the height of 3816 m above sea level. The measured size range was 12–570 nm in diameter. The measurements started in August 2005 and ended in May 2007. There were two major gaps in the time series, each being 2.5–3 months in length. The data was checked with independent CPC measurements (particle diameter >10 nm) running parallel with the DMPS system and a good agreement was found between the instruments.

The average and median number concentrations of all particles (12–570 nm in diameter) were 2040 cm^{-3} and 1390 cm^{-3} , respectively. The concentrations at Mount Waliguan were higher than those measured in Europe, North America and Japan at

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similar altitudes. The particle number concentrations reported in this study are more similar to those reported at several alpine sites at Indian Himalayas. Accumulation mode particle number concentrations were higher than those reported elsewhere at similar altitude, and are closer to those reported in planetary boundary layer at rural locations.

The particle number concentration seemed to peak at summer but, because of the lack of data, this feature could not be verified. When averaged over the different seasons, daily patterns in the particle number concentration emerged. The number concentration of nucleation mode particles (diameter <21 nm) increased in the afternoon between noon and around 17:00 Beijing time, and gradually decreased for the rest of the time. This increase was also seen in the Aitken mode some hours later during summer, but not during the other seasons. No clear daily patterns were found in the accumulation mode number concentration during any season. The daily patterns in nucleation and Aitken modes are probably a result of the new particle formation events in the area. There was no systematic pattern of changes between boundary layer and free tropospheric air in any season.

A trajectory analysis revealed that air masses coming from the east (45° – 165°) had, on average, higher particle number concentrations than those coming from other directions. These particles were mainly in the Aitken and accumulation mode. The portion of nucleation mode particles was highest in air masses coming from western desert areas (direction 270° – 315°). The Aitken to accumulation mode number concentration ratio was around 2.6–3.2 in all air masses, which indicated that most particles were produced within the last one or two days, rather than being transported from over large distances.

These findings indicate that the site is probably not affected as much by the East Asian brown cloud (Ramanathan et al., 2007a) at the coastal regions of China when the air masses are coming from east, as by some anthropogenic pollution sources at shorter distances east of the station. Also the regional-scale aerosol formation seems to be intense, contributing to high particle number concentrations in other air masses

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as well.

Acknowledgements. This work was supported by Tor and Maj Nessling foundation and by CMA-FMI bilateral cooperation project, the National Natural Science Foundation of China (Grant No. 40575063), National Basic Research Program of China (Grant No. 2006CB403703, 2006CB403701). The authors would also like to thank the Mount Waliguan staff for operating and maintaining the instruments at the station.

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Table 1. The sectors in the trajectory analysis (air mass location 24 h before arriving to the site) and aerosol size distribution parameters attached to them. The percentages in the Distribution rows are the number of the cases with that type of distribution divided by the number of all cases with the arriving sector determined by the column.

Parameter	sector 1 North	sector 2 East	sector 3 South	sector 4 West	All data
Compass angle (°)	315–45	45–165	165–270	270–315	0–360
Number of cases	904	1586	1300	4928	8678
Distribution 1	7.9%	4.0%	8.0%	8.9%	7.8%
Distribution 2	13.1%	8.6%	23.9%	23.1%	19.6%
Distribution 3	26.5%	27.6%	16.4%	6.9%	14.2%
Distribution 4	26.5%	46.2%	18.9%	15.6%	22.9%
Distribution 5	26.0%	13.7%	32.8%	45.4%	35.9%
Mean N_{nuc} (cm^{-3})	581	577	692	555	570
Mean N_{Ait} (cm^{-3})	1149	1488	1238	868	1060
Mean N_{acc} (cm^{-3})	425	606	467	362	430
Mean N_{tot} (cm^{-3})	2120	2637	2356	1753	2040
Median N_{nuc} (cm^{-3})	277	222	368	304	290
Median N_{Ait} (cm^{-3})	826	1070	763	598	690
Median N_{acc} (cm^{-3})	379	559	319	306	330
Median N_{tot} (cm^{-3})	1533	1941	1511	1229	1390
Mean $N_{\text{Ait}}/N_{\text{acc}}$	3.18	2.76	2.90	2.58	2.65

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Table 2. The ratios of daytime (14:00–18:00) to nighttime (hours 02:00–06:00) average and median number concentrations of particles of each mode during each season.

Average				
Season	Nucleation	Aitken	Accumulation	Total
winter	1.75	1.04	1.02	1.19
spring	2.47	0.86	0.97	1.18
summer	2.25	0.67	1.04	0.96
fall	2.58	0.97	1.09	1.31
total	2.20	0.93	1.03	1.20
Median				
Season	Nucleation	Aitken	Accumulation	Total
winter	1.19	0.95	1.09	0.99
spring	0.90	0.88	0.97	0.91
summer	0.89	0.63	1.08	0.80
fall	1.25	1.03	1.11	1.15
total	1.08	0.97	1.06	1.05

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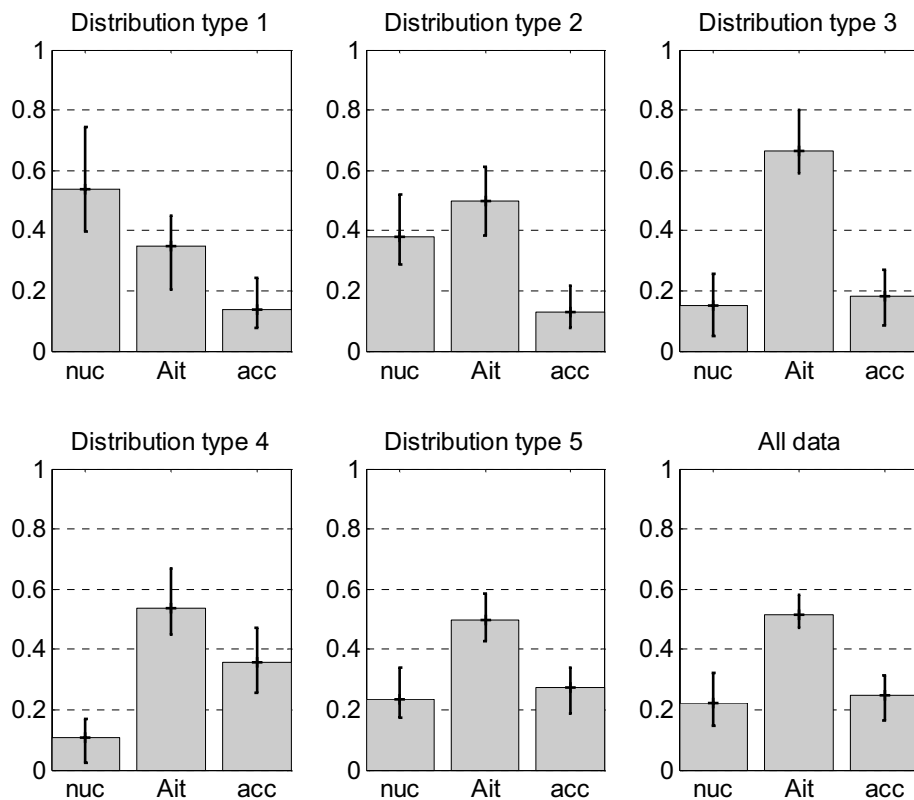


Fig. 1. Median relative concentrations of each mode in the five distribution types and in the total data set. The error bars show the 5 and 95 percentiles.

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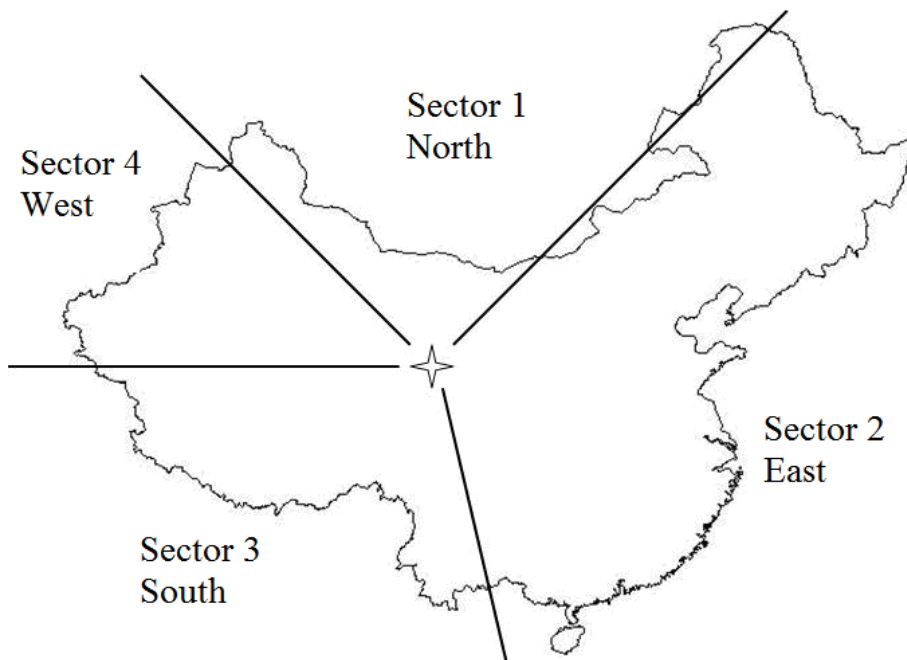


Fig. 2. The location of Mount Waliguan observatory, and the sectors according to which air masses were divided.

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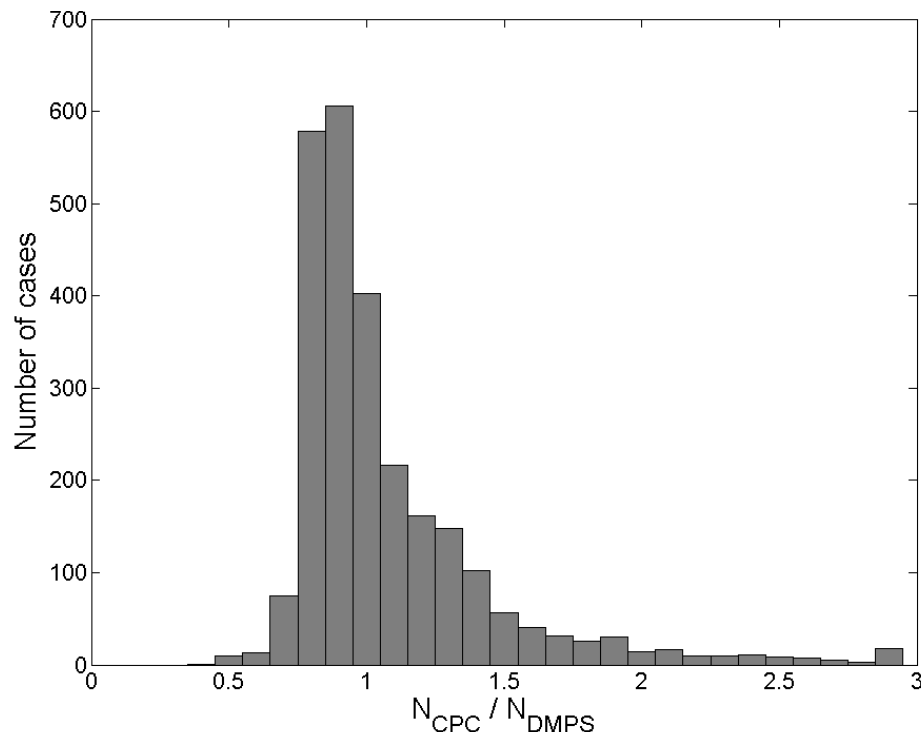


Fig. 3. A histogram showing the ratio of particle number concentration obtained from CPC to that obtained from the DMPS.

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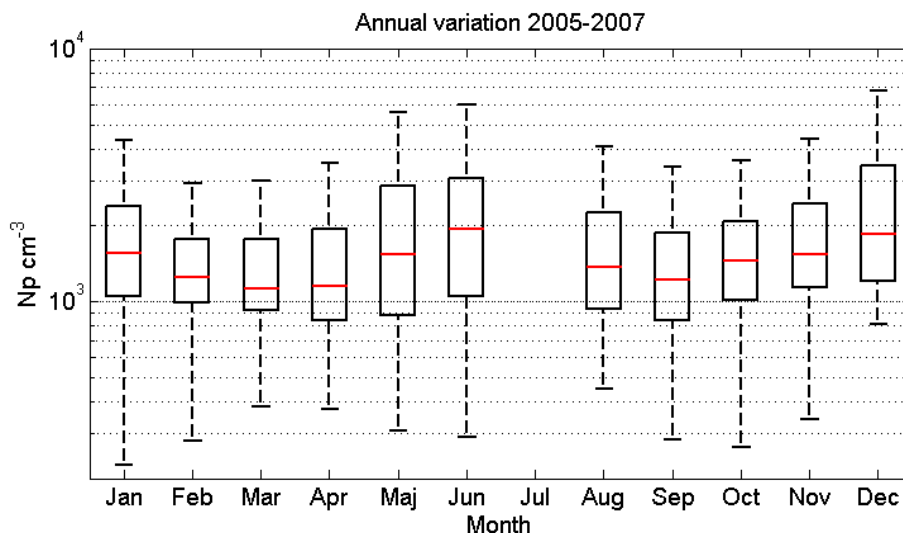


Fig. 4. The seasonal variation of particle number concentration at Mount Waliguan. The median values (red lines inside the boxes), 25 and 75 percentile values (box upper and lower limits) and 5 and 95 percentile values (error bars) of N_p for each month in the combined data. There were no data available for the month of July.

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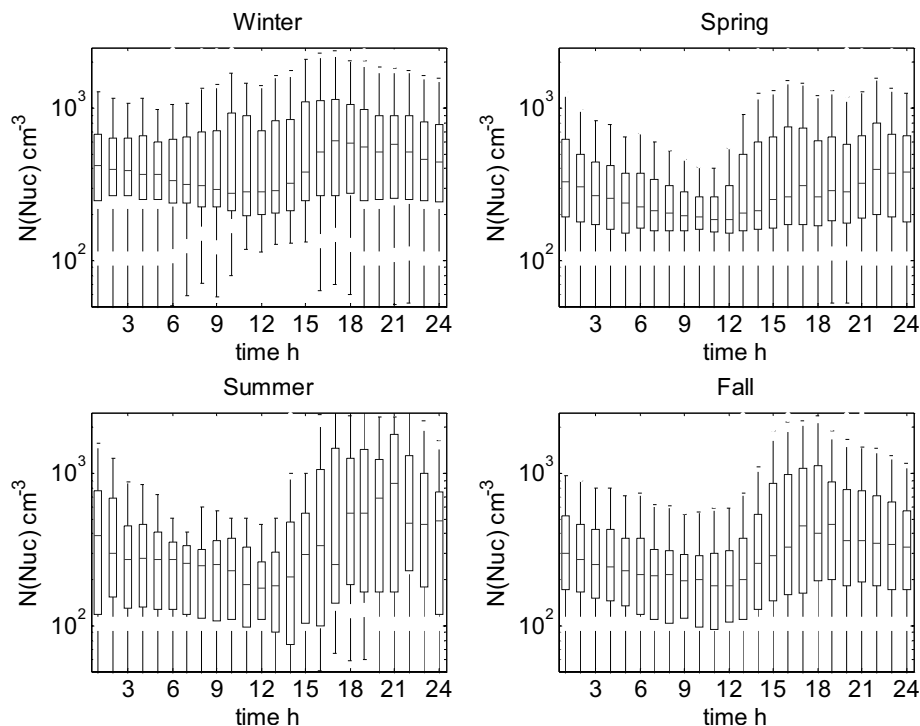


Fig. 5. The average diurnal pattern of nucleation mode particle number concentration variation in different seasons. The number concentration axis is logarithmic. The line in the middle of each box is the median value, the box extends from 25% to 75% values, and the error bars are the 5% and 95% limits.

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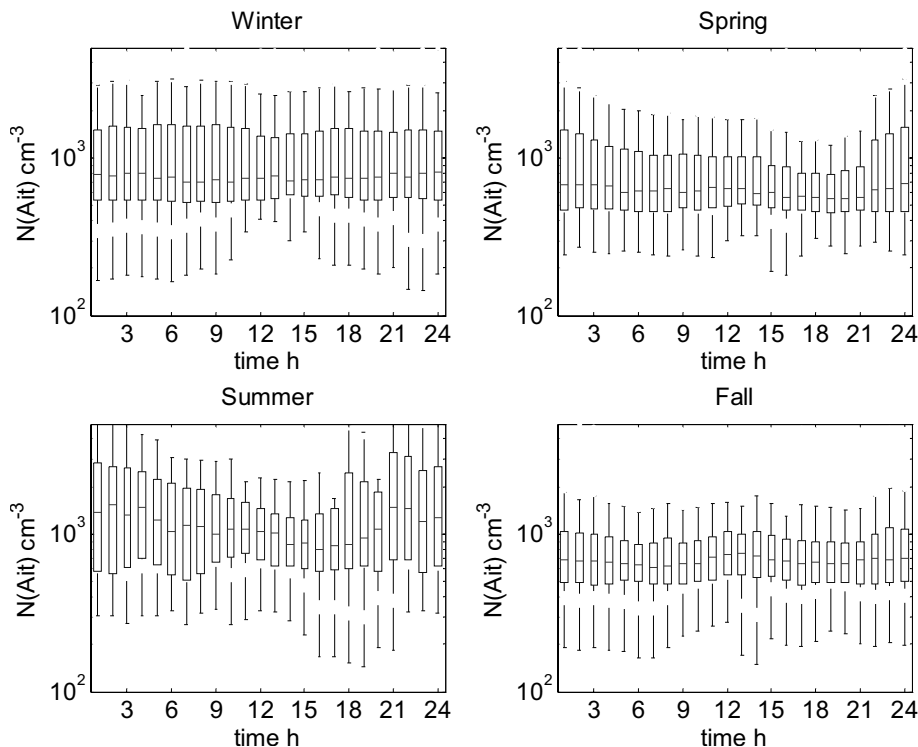


Fig. 6. The average diurnal pattern of Aitken mode particle number concentration variation in different seasons. The number concentration axis is logarithmic. The line in the middle of each box is the median value, the box extends from 25% to 75% values, and the error bars are the 5% and 95% limits.

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measurements at
Mount Waliguan,
inland China**

N. Kivekäs et al.

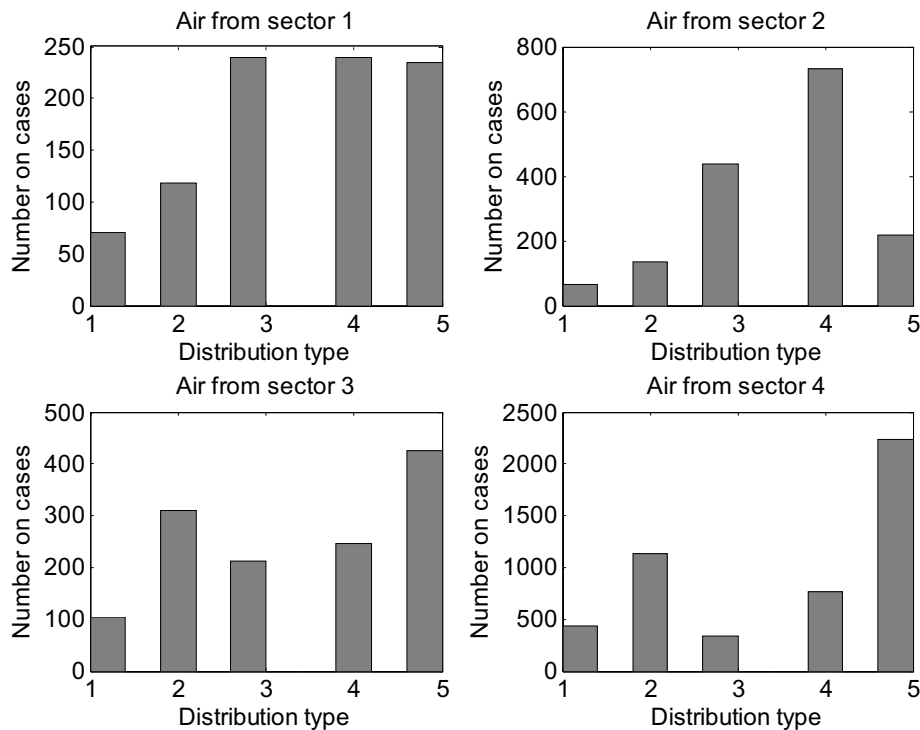


Fig. 7. Abundancy of the five distribution types in air masses coming from the four different sectors.

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