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Lower tropospheric vertical distribution of aerosol particles over Syowa Station, Antarctica from spring to summer 2004

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Abstract: Vertical distributions of atmospheric aerosol particles were measured nine times up to 5200 m a.s.l. using an aircraft over Syowa Station, East Antarctica during September–December 2004. Measurements were made for number concentrations of condensation nuclei (CN, D$p > 10$ nm), number–size distribution larger than 0.3 μm diameter, air temperature, humidity, and GPS position. During spring, the vertical profile of CN concentration showed large variability (100–1000 cm^{-3}), but it was mostly constant in summer. Vertical profiles of number–size distribution larger than 0.3 μm showed a systematic shift at altitudes greater than 4000 m. Both concentrations of aerosol number for D$p > 0.3$ μm and integrated volume between 0.3 and 1.0 μm showed constantly lower values at altitudes greater than 4000 m. These data suggest that the shift of aerosol parameters results from depletion of coarse particles such as sea salts. Maximum concentrations of the volume and CN were obtained respectively from the profiles on 7 October and 29 November 2004. These events are interpreted as sources and transport processes based on synoptic meteorological data, an ocean color index produced by SeaWiFS, and backward air trajectory analysis.

key words: aerosol particles, vertical distribution, size distribution, condensation nuclei

1. Introduction

Aerosol particles play an important role in many respects in the atmosphere. They exert direct and indirect effects on the Earth's radiation budget. They also affect geochemical cycles through the atmosphere. Aerosol effects on the radiation budget include: scattering and absorbing solar radiation (the so-called direct effect), and modifying the optical properties and lifetime of clouds as indirect effects (see review in Charlson and Heintzenberg, 1995). Geographical and vertical distributions of aerosols according to their sizes are fundamental parameters to describe the atmospheric environment. Many studies have reported vertically resolved size distributions of aerosols in middle and lower latitudes (*e.g.*, Clarke *et al.*, 1998; Clarke and Kapustin, 2002; Kristament *et al.*, 1993; Lechner *et al.*, 1989). However, such data have rarely been reported for the Antarctic region (Hofmann, 1988).

On the other hand, analyses of ice cores obtained from inland Antarctica reveal changes in aerosols in atmospheres of the past (*e.g.*, Oeschger and Langway, 1989; Delmas, 1992; Legrand and Mayewski, 1997; Wolf *et al.*, 2003). Knowledge of source and transport factors of Antarctic aerosols under present meteorological conditions is necessary to properly interpret ice core data as atmospheric contexts in the past. Deep ice cores from the Antarctic ice sheet have been obtained from inland high elevation sites such as Dome C, Vostok, and Dome Fuji Stations. Therefore, information on vertical distribution of aerosol parameters and its variability might provide useful insight to deduce source-sink processes of aerosols relating to analyses of ice core data from Antarctica.

Several studies have examined the vertical distribution of aerosols in Antarctica (Hogan, 1986; Hofmann, 1988). At Syowa Station (69.0°S, 39.6°E, 29 m a.s.l.) in East Antarctica, balloon-borne (Ito *et al.*, 1986; Ito, 1989) and airplane-borne (Iwasaka *et al.*, 1985; Yamanouchi *et al.*, 1999; Wada *et al.*, 2001) observations have been made for studying aerosols around the station. Although a limited number of observations and parameters were observed at Syowa Station, those reports provided interesting findings. For instance, they presented the possibility of very long range transport of volcanic materials from El Chichon, Mexico (Yamazaki *et al.*, 1989). However, the vertical distribution of tropospheric aerosols in Antarctica is not well understood and must be related carefully with their source and transport processes to Antarctica. This paper presents results of nine vertical observations of aerosols up to 5200 m altitude during September–December, 2004. We use a simple parameterization to show major aspects of aerosols over Syowa Station. We also discuss factors controlling important events found in the vertical profiles.

2. Observations

The aircraft used was a PC-6 Turbo Porter, identical to the one used in Iwasaka *et al.* (1985). Outside air temperature and relative humidity were measured and stored on a data logger (MR-6661; CHINO Co. Ltd.). A handheld optical particle counter (KR-12A; Rion Co. Ltd.) and a condensation nucleus counter (CPC-3007; TSI Inc.), both having data storage systems, were used to measure the aerosol size distribution and number concentration. The KR-12 measures the number concentration of particles at a flow rate of 2.83 l min^{-1} using six diametric groups: $>0.3, >0.5, >0.7, >1.0, >2.0$ and >5.0 μm. The CPC-3007 quantifies condensation nuclei (CN) larger than 10 nm at a flow rate of 0.7 l min^{-1}. All data were obtained for every minute, except for the GPS position and height data, which were logged every 2 s.

Sample air was introduced into the cabin of the aircraft through an inlet at the upper end of the wing stay, similar to that illustrated in Iwasaka *et al.* (1985). The inlet's location was chosen to eliminate contamination from aircraft engines and to minimize the loss of particles in the tube. A Tygon tube of 9.6 m length, 6 mm i.d. and 13 mm o.d. was used to introduce aerosol particles. Figure 1 shows a comparison of data obtained simultaneously in the aircraft (using the KR-12) during movement on the taxiway and in the aerosol observatory (TD-100; YGK Co. Ltd.) at Syowa Station on 9 December 2004. Instrumental differences between the KR-12 and TD-100 and inlet losses at the aerosol observatory were corrected beforehand through simultaneous measurements performed inside and outside of the observatory (Osada *et al.*, 2006). As Fig. 1 shows, although the length of the inlet tube used

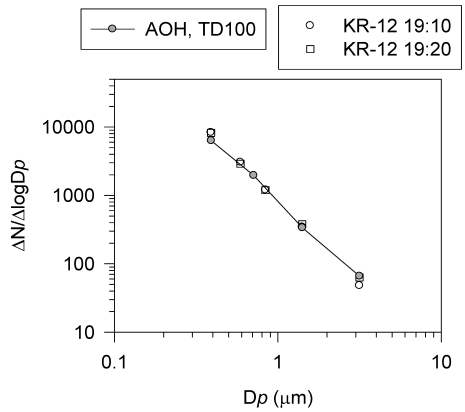

Fig. 1. Comparison between aerosol data obtained simultaneously in the aircraft (marked as KR-12) and in the aerosol observatory (AOH) at Syowa Station on 9 December 2004.

in this study is sufficiently long to affect the size distribution, especially for aerosols of super-micrometer sizes, the aerosol size distribution measured using the KR-12 on the aircraft during movement on the taxiway is almost identical to that of the TD-100 at the aerosol observatory. Based on this comparison, we assume that sampling artifacts of the size distribution obtained using the KR-12 in the aircraft are negligible. For this reason, the size distribution in this study will be presented without any correction for inlet losses.

The aerosol observation flight path was limited to an area over sea ice and a fringe of the continental ice sheet around Syowa Station, mostly within several tens of kilometers and 100 km at maximum from the station, to avoid large spatial differences. Because of severe Antarctic weather conditions, observation flights were made only under clear-sky conditions with calm to modest winds at the ground. Vertical profiles of aerosols up to the pressure altitude of 18000 ft (*ca*. 5.2 km in Antarctica) were obtained eight times in 2004: on 22 September, 7 October, 17 October, 31 October, 6 November, 29 November, 4 December, and 9 December. Aerosol data on 14 November were also included in this study, but the flight was limited to 12000 ft because of changes in weather conditions. Aerosol samples for analyses by scanning electron microscopy (SEM) were obtained during the ascending flight at pressure altitudes of 6000, 12000, and 18000 ft to analyze morphology and elemental compositions of individual particles. Results of SEM analysis will be reported elsewhere. A constant descending rate was applied for the return flight to the station.

3. Results and discussion

3.1. Vertical distribution during spring and summer

Figure 2 shows vertical profiles of aerosol parameters for all flights during descent: (a) CN concentration; (b) Junge's β exponent, as derived from the size distribution between 0.3 and 1 μm, fitted to a power-law function ($dN/d\log D = \text{Const}*D^{\beta}$, Junge, 1963); (c) volume concentration of accumulation mode between 0.3 and 1 μm; and (d) number concentration larger than 0.3 μm ($N > 0.3\ \mu$m).

Figure 2a shows that most CN data were 100–1000 cm^{-3}, with some variations. The

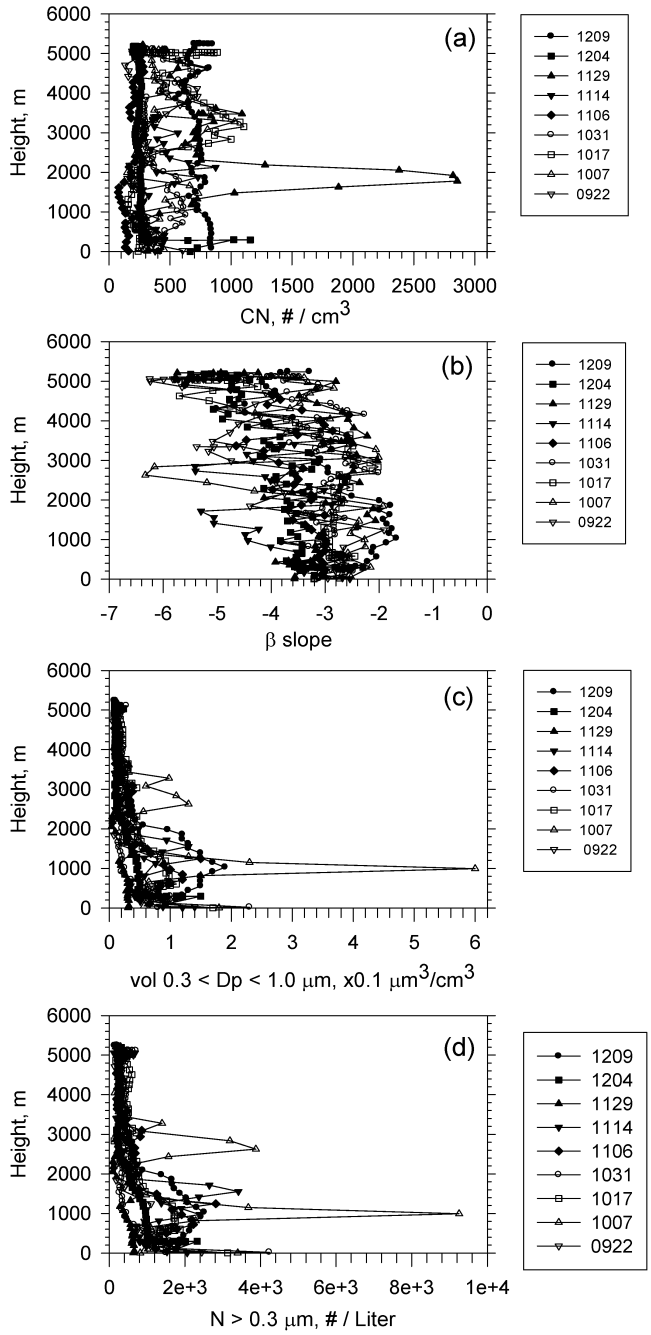

Fig. 2. Vertical distributions of (a) CN concentration (Dp > 10 nm), (b) Junge's β exponent (see text), (c) volume concentration of accumulation mode between 0.3 and 1 μm, and (d) number concentration of particles larger than 0.3 μm (N > 0.3 μm).

range of CN concentrations agrees well with ground data at Syowa Station (Ito, 1985; Osada *et al*., 2006) and for the troposphere (Ito, 1989). Nevertheless, two vertical profiles of the CN concentration on 4 and 9 December show almost constant vertical concentrations; other profiles indicate large variability through the atmosphere. The highest CN (*ca*. 3000 cm^{-3}) concentration was found at about 2000 m on 29 November. Its origin will be discussed later.

The β exponent in Fig. 2b is mostly between –2 and –6. The typical range of the β exponent might be represented as –3 to –5 over wide areas of oceans and continents (Pruppacher and Klett, 1997) and as -4.1 ± 0.5 for free tropospheric observations over the southwest Pacific (Kristament *et al*., 1993). Over Syowa Station, values of the β exponent in this study are *ca*. -3 ± 1 below 4000 m, but most of the β exponent values decreased to -4 ± 1 at altitudes greater than 4000 m. A lower value of the β exponent indicates a steep slope of size distribution, possibly resulting from either enrichment of fine sulfate particles or depletion of coarser sea salt particles, as deduced from size-segregated aerosol composition at the ground (Osada *et al*., 1998). As Fig. 2c shows, the volume concentration between 0.3 and 1 μm shows a decreasing trend with increasing height to 4000 m. The volume concentrations are invariably low above that altitude. Combining the low volume concentration above 4000 m with the fact that values of $N > 0.3$ μm are also invariably low above 4000 m (Fig. 2d), lower values of the β exponent above 4000 m are considered to result from depletion of coarse particles aloft. A similar tendency of depletion of coarse particles has been found over the southwest Pacific (Kristament *et al*., 1993) and in other places (Pruppacher and Klett, 1997).

Materials that are considered as coarse particles include sea salts, although fine sea salt particles might also exist (Cipriano *et al*., 1983; Osada *et al*., 1998; Murphy *et al*., 1998). Data recorded at Dome Fuji Station (39.62°E, 77.37°S, 3810 m a.s.l.) and Syowa Station (Hara *et al*., 2004) demonstrate that the Na^+ concentration of aerosols in summer at Dome Fuji (*ca*. 0.4 nmol m^{-3}) was about 1/10 that at Syowa (*ca*. 4 nmol m^{-3}), indicating a vertical gradient of sea salt concentration between Dome Fuji and Syowa. This observation agrees with the depletion of coarse particles at higher altitude observed in this study. On the other hand, the maximum volume concentration (0.6 μm^3 cm^{-3}) was obtained at about 1000 m on 7 October, which will also be discussed later in detail.

In the following sections, we will discuss factors relating to maxima of parameters in vertical profiles of aerosols, based on backward air trajectory analyses. Trajectories for 120 h from the flight time were calculated using the HYSPLIT 4 model (Draxler and Rolph, 2003). The starting height of the trajectories was set at six altitude levels: 900, 1800, 2700, 3500, 4300 and 5200 m a.s.l.

3.2. High volume concentration on 7 October 2004

Figure 3 shows vertical profiles of the following on 7 October 2004: *Ta*, air temperature; RH, relative humidity; and number concentrations of CN and aerosol particles in four size ranges. Open and filled symbols respectively represent measurements for ascending and descending flights. Large discrepancies are apparent between ascending and descending data for all parameters, suggesting rapid changes of atmospheric conditions during the flight or great spatial non-uniformity within several tens of kilometers to 100 km around Syowa Station. Number concentrations of accumulation particles are high for all size ranges at

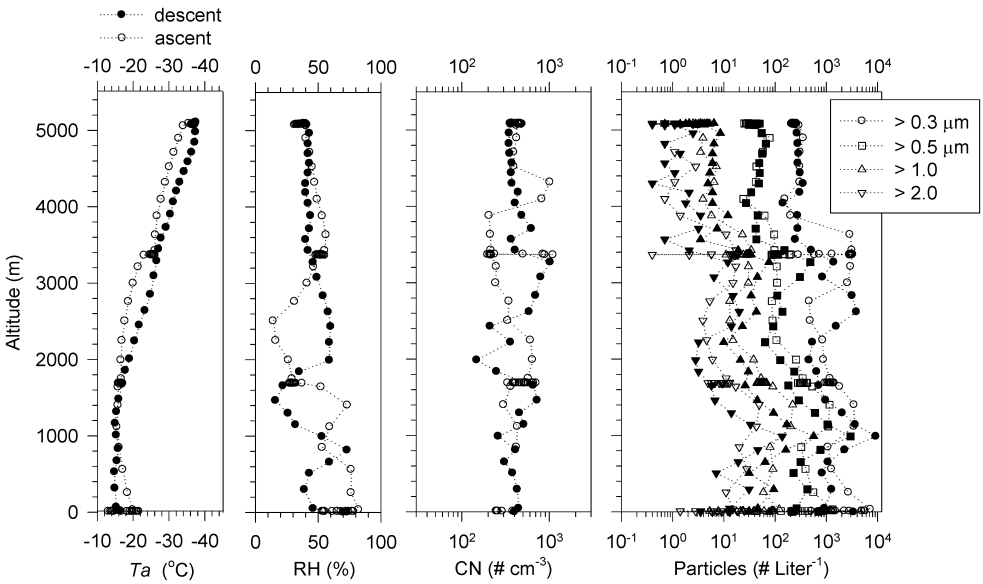

Fig. 3. Vertical profiles of Ta: air temperature, RH: relative humidity, number concentrations of CN and aerosol particles with four size ranges on 7 October 2004. Open and filled symbols respectively represent data for ascending and descending flights.

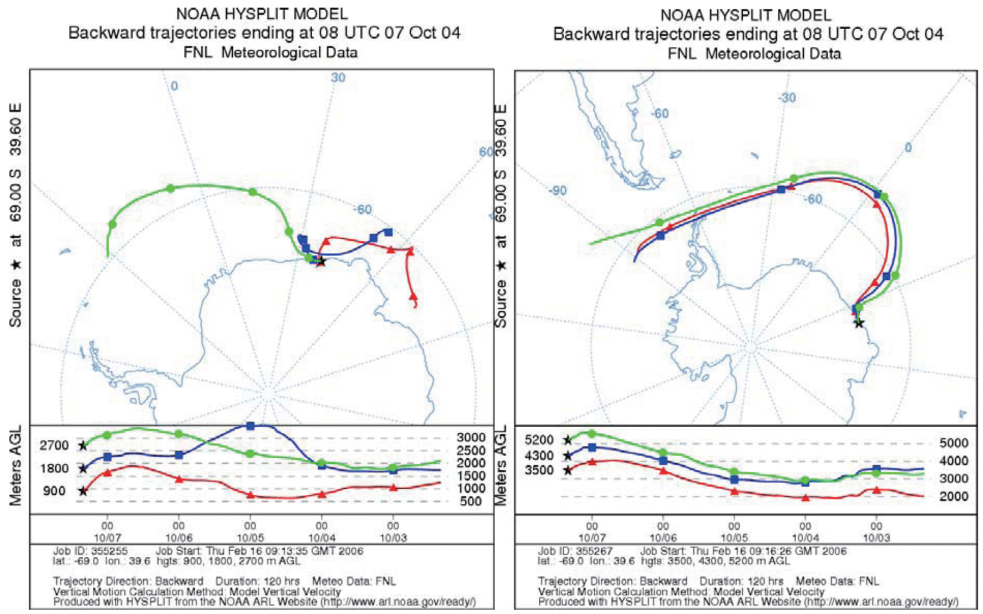

Fig. 4 Backward air trajectories starting from the flight time on 7 October for 120 h calculated using the HYSPLIT 4 model (Draxler and Rolph, 2003; http://www.arl.noaa.gov/ready/hysplit4.html). Left: trajectories from 900, 1800 and 2700, right: trajectories from 3500, 4300 and 5200 m a.s.l. over Syowa Station.

about 1000 m and at 2500–3500 m.

Figure 4 shows backward air trajectories starting from the flight time on 7 October. The trajectory lines indicate air mass movement of clockwise transport from the west, except for the trajectory starting at 900 m and 1800 m advected from the east. As indicated in vertical motion, an air parcel starting from 900 m was transported from the marine boundary layer (MBL) well below 1000 m at 2 days prior. The air mass from MBL might contain high sea salt concentrations if the abundance of sea salt aerosols were high in the MBL, and if the scavenging and dry deposition of aerosols were not substantial during transport. For the aerosol peak at about 1000 m, the air mass was transported from the MBL of the stormy area at around 60°S, where high sea salt concentrations would be expected (Erickson *et al*., 1986). Two days passed before it arrived at Syowa Station, so the effect of dry deposition might have been small. The amount of precipitation estimated by the HYSPLIT 4 model during transport from the MBL was also small (2.3 mm accumulated after leaving MBL) for the trajectory from 900 m. The proximity to a probable stormy source area and minor scavenging by precipitation might have caused high volume concentration at that altitude.

3.3. High CN concentration on 29 November 2004

Figure 5 shows the same parameters as those presented in Fig. 3, but for 29 November 2004. Differences of data between ascending and descending profiles are smaller than in the example of 17 October but some are noticeable at 1000–2500 m. As mentioned earlier, the highest CN concentration was observed at about 2000 m with a low number concentration of accumulation mode particles. For the ascending (open circles) and descending (filled circles) profiles, the CN peaks were observed at about 2400 m and 1800–2000 m with lower

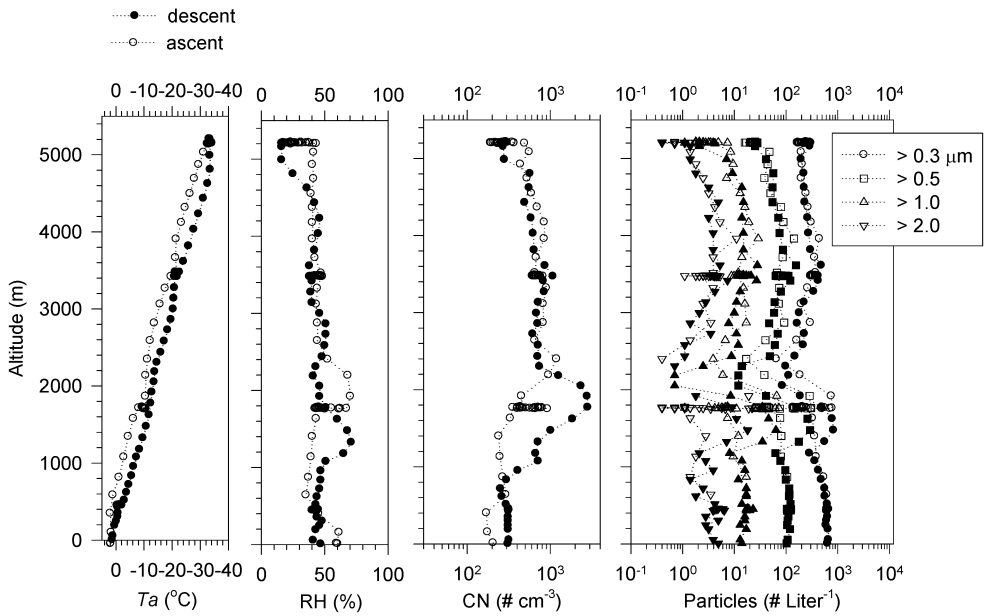

Fig. 5. As in Fig. 3, but for 29 November 2004.

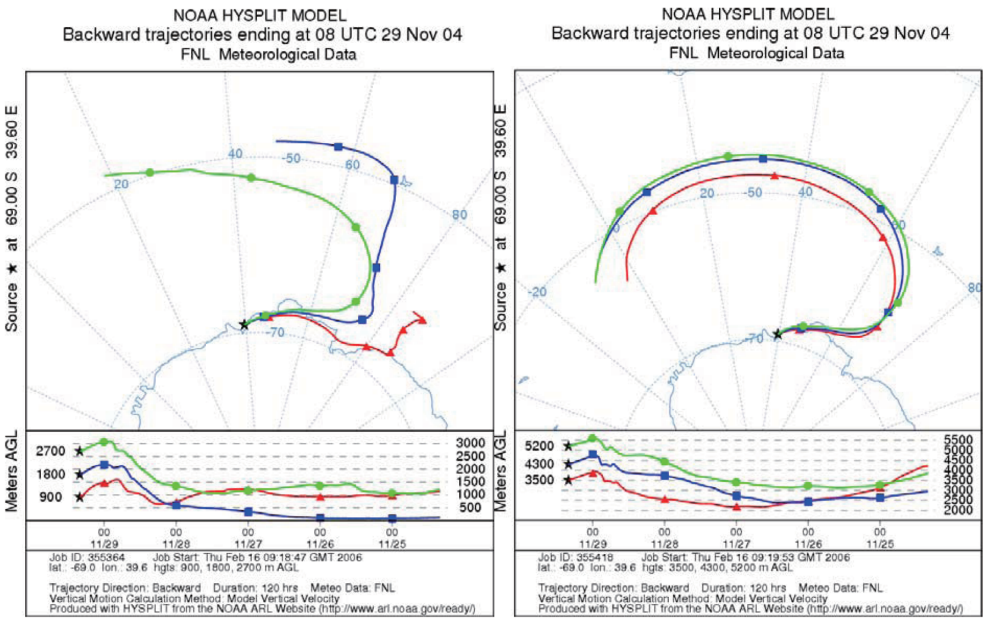

Fig. 6. As in Fig. 4, but for 29 November 2004.

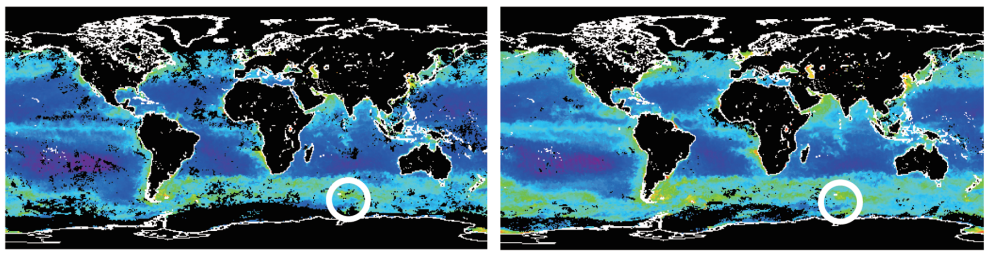

Fig. 7. Ocean color index, produced by the SeaWiFS Project (http://oceancolor.gsfc.nasa.gov/), representing chlorophyll a concentration. The warm color represents higher chlorophyll concentration. Left and right panels represent data for 24 November to 1 December and 2 December to 9 December 2004, respectively. Circles represent the location (origin) of the backward air trajectory (see text).

concentration of accumulation mode particles, respectively. Figure 6 shows trajectories from the flight on 29 November. About one day prior, the air mass at 1800 m that had arrived over Syowa Station was located vertically within the MBL and geographically the north of the Amery ice shelf and south of the Kerguelen Archipelago. Figure 7 shows eight-day composites of the ocean color index representing chlorophyll *a* concentration; the SeaWiFS Project (http://oceancolor.gsfc.nasa.gov/) produced the composites. Warm colors signify higher chlorophyll *a* concentration. For the air mass that had arrived at 1800 m over Syowa, white circles in Fig. 7 mark the location within the MBL, as deduced from the trajectory. During the week of 24 November 2004 (Fig. 7, left), the sea ice distribution (black area outside the thin white line showing Antarctic continental margin in Fig. 7) retained much of its northern extension, but it retreated rapidly southward during the week of 2

December 2004 (Fig. 7, right). Corresponding with the sea ice retreat, the area showing high chlorophyll *a* concentration in the marginal ice zone increased and advanced to the south. Similarly elevated chlorophyll concentration near the Kerguelen Plateau has also been reported from analyses of satellite ocean color data (Moore and Abbott, 2002). The location of the high chlorophyll *a* area coincides with the plausible source area deduced from the trajectory at 1800 m over Syowa Station. High chlorophyll *a* concentration in the ocean surface might engender high dimethyl sulfide (DMS) emission. Therefore, the air mass from this area might contain high DMS. Based on measurements on board the R/V *Shirase* in 1996, Inomata *et al*. (2006) reported the very high value of 755 pptv of atmospheric DMS concentration in the marginal ice zone near the area mentioned above, but those observations were made in a different year from this study. Subsequent oxidation of such a high DMS level engenders a remarkably increased number of nucleation mode particles (Pirjola *et al*., 2000). After formation of new particles, the nucleation mode particles grow during transportation to larger than 10 nm, which is sufficiently large to be detectable using our instruments because the growth rate of nucleation mode particles in Antarctica is assumed to be about 1 nm h^{-1} (Osada *et al*., in preparation; Kulmala *et al*., 2005). The CN peak at about 1800 m altitude over Syowa Station is considered to be derived from new particle formation after high DMS emission south of the Kerguelen Archipelago and in subsequent growth during transportation.

3.4. Vertically uniform distribution of CN on 9 December 2004

Figure 8 shows the same parameters as those in Fig. 3, but for 9 December 2004. Almost identical profiles are apparent for all parameters in ascending and descending flights, suggesting that temporal and spatial distributions of aerosols during the flights were

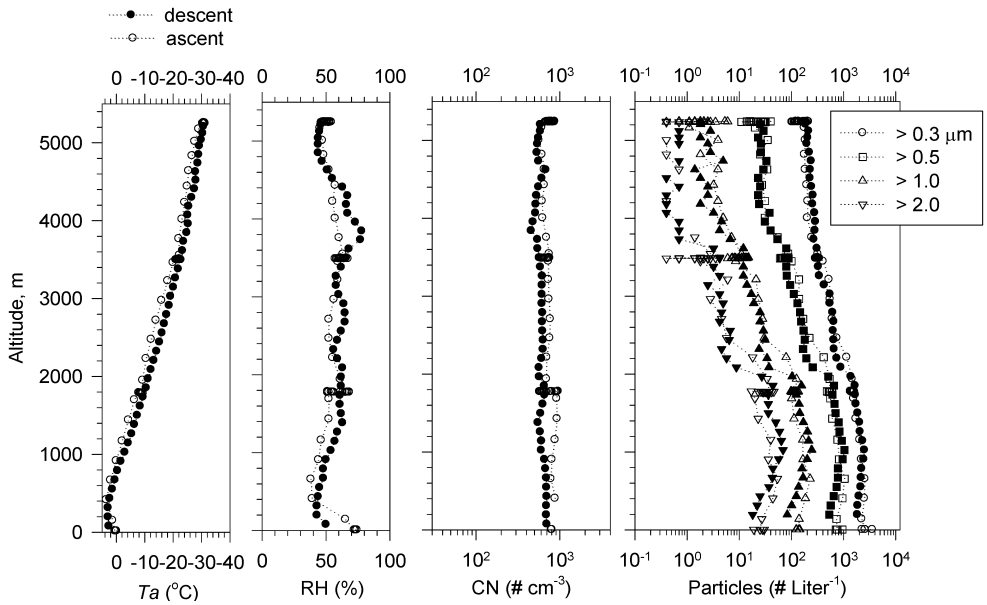

Fig. 8. As in Fig. 3, but for 9 December 2004.

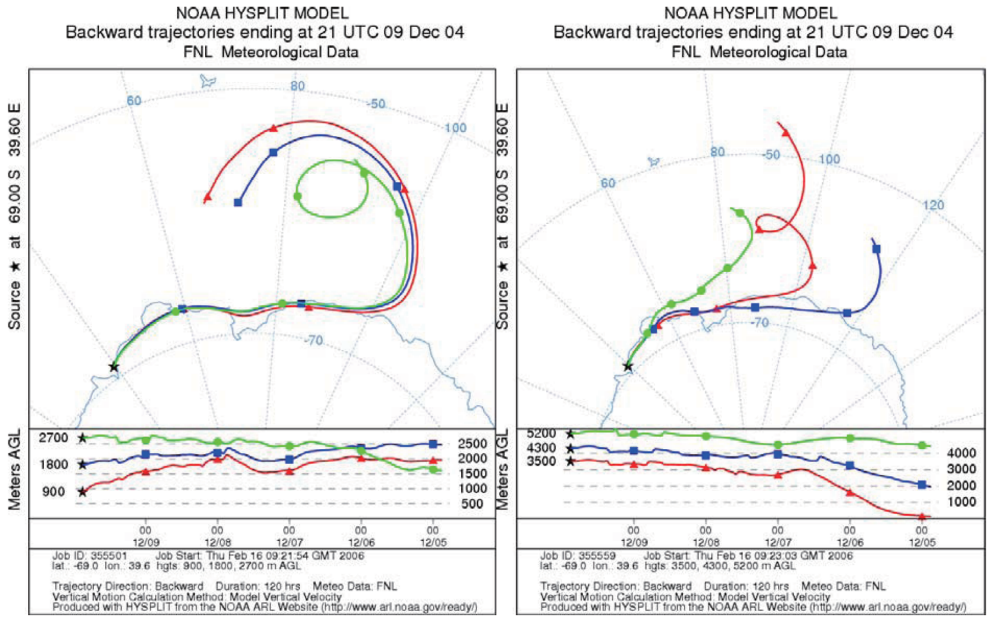

Fig. 9. As in Fig. 4, but for 9 December 2004.

mostly uniform. The uniformity that is shown in temporal and spatial distribution agrees with those of previous reports of the summer atmosphere at Syowa Station (Ito, 1989; Yamanouchi *et al*., 1999). Furthermore, the vertical distribution of CN concentration is surprisingly uniform up to the maximum altitude of 5200 m. As mentioned in an earlier section, a similar uniformity of CN concentration was also apparent in data of 4 December. 2004. The CN profile on 29 November showed nearly uniform concentrations above the pronounced peak at *ca*. 2000 m. These cases at Syowa Station imply that the CN concentrations in summer were uniformly distributed through the middle to lower troposphere, with sporadic peaks like that on 29 November. Other observations during summer (Hogan, 1986; Hofmann, 1988; Ito, 1989) reported variable CN profiles within the troposphere, but they might be partly attributable to the presence of sporadic peaks after nucleation events that are often observed during Antarctic summers (Ito and Iwai, 1981; Koponen *et al*., 2003).

Figure 9 shows trajectories from the flight on 9 December. The trajectories in Fig. 9 and those on 4 December (not shown) are characterized by westward transportation along the edge of the continent for a few days before arriving at Syowa Station. According to statistical analyses of air transport to Syowa on 1997 (Suzuki *et al*., 2004), the length of the backward air trajectory during the 5 days is shorter in summer than in other seasons for altitudes of 500 and 850 hPa. This dependence of transport distance on season implies that air mass transport in the Antarctic summer occurs through a rather slow and gradual mixing process. In summer, weather conditions at Syowa Station are favorable to develop aged Aitken-mode particles because of reduced precipitation scavenging (Konishi *et al*., 1992). Although we have no convincing explanation for the vertical uniformity of observed CN concentrations, seasonal changes of air transport and source-sink processes might explain the unique CN distribution in summer.

4. Summary and conclusions

Atmospheric aerosol particles over Syowa Station, Antarctica were measured using an aircraft during September–December 2004. During austral spring, concentrations of condensation nuclei (CN) showed large variability (100–1000 cm^{-3}) with altitude, but they remained mostly constant in summer up to the maximum altitude of 5200 m. The vertical profile of number–size distribution of particles larger than 0.3 μm showed a shift at about 4000 m: constantly low-volume concentrations pertained above that altitude and the coarse fraction of aerosols decreased with increasing altitude. The highest volume and CN concentrations in the vertical profiles in this study were correlated with air mass transportation, possibly containing numerous sea salt and high Aitken particles, both from the marginal sea ice area of the Southern Ocean.

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