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SIZE DISTRIBUTION OF AEROSOL PARTICLES OVER THE WESTERN PACIFIC OCEAN AND THE SOUTHERN OCEAN

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Abstract: On cruise KH-94-4 of the research vessel HAKUHO MARU, we measured aerosol concentrations over the western Pacific Ocean and the Southern Ocean. Aitken particle concentration was measured with a Pollak condensation nuclei counter. Size distribution for large and giant particles was measured with an optical particle counter. On this expedition, we observed at a nearly fixed station (64–65.5°S, 140°E) twice, in December 1994 and January 1995, for a total of sixteen days. The concentrations of Aitken particles at the fixed station were somewhat higher than those measured around 10°N latitude during this expedition and previous expeditions. These values were nearly equal to or lower than values obtained by other workers. Moreover, Aitken particle concentration at the fixed station sometimes showed a clear diurnal variation. This implies that they were produced by gas-to-particle conversion of organosulfur gases. For large and giant particles, we first confirmed the effect of wind force on the size distribution over the Southern Ocean. These results suggest that the principal constituents of particles over the Southern Ocean in summer are sulfur-dominant Aitken particles and sea salt larger particles.

1. Introduction

Background aerosol particle concentration is of great interest in the fields of atmospheric radiation and cloud physics. The concentration has been measured on board the research vessel HAKUHO MARU, of the Ocean Research Institute, the University of Tokyo, on eight cruises (KH-89-T3, KH-89-2, KH-91-5, KH-92-5, KH-93-3, KH-94-1, KH-94-3 and KH-94-4). We have reported the global distribution of Aitken particles over the oceans drawn with data obtained on our three cruises (KH-89-T3, KH-89-2 and KH-91-5) in addition to data by other workers (MIURA *et al.*, 1993). But there are few observations over the Southern Ocean (KOJIMA and SEKIKAWA, 1974; IWAI *et al.*, 1979; ITO *et al.*, 1980; HOGAN, 1981; BIGG, 1990), so that the Southern Ocean part of the map was only roughly drawn.

On cruise KH-94-4, we measured particle concentration over the Southern Ocean, and collected aerosols on filters and on a carbon-covered nitrocellulose film supported on an electron microscopic grid. Moreover, we observed solar radiation and measured atmospheric electrical conductivity and several gases. In this paper, the number concentrations of aerosol particles over the western Pacific Ocean and the Southern Ocean are

presented.

2. Methods

Cruise KH-94-4 of R/V HAKUHO MARU consisted of four legs. Track charts on these legs are shown in Fig. 1. On the first leg the ship departed from Tokyo on 22 November 1994 and arrived in Port Lyttelton, New Zealand on 9 December 1994. On the second leg the ship departed from Lyttelton on 13 December 1994 and went to the Southern Ocean and arrived in Hobart, Australia on 4 January 1995. On the leg 3 the ship departed from Hobart on 9 January 1995 and crossed the Southern Ocean, arriving in Sydney on 28 January 1995. On the last leg the ship departed from Sydney on 1 February 1995 and came back to Tokyo on 14 February 1995.

Counting of aerosol particles was continuously carried out with two counters on an upper deck, being about 15 m above the sea surface and in the direction of 30 degrees left from the front of a funnel. Air was drawn with a cleaner, whose flow rate was 180 *l*/min, through a sampling tube made of vinyl chloride 18 mm in diameter and about 5 m long, to the No. 1 laboratory room behind the wheel house. Sample air was bypassed from the sampling tube into two counters. In this process air temperature and humidity were not controlled.

Aitken particle concentrations ($r<0.1$ μm) were measured with a Pollak condensation nuclei counter with a diffusion battery. Larger particle concentrations ($r\geqq 0.15$, 0.25, 0.5, 1 and 2.5 μm) were measured with an optical particle counter (KC01, Rion Co. Ltd.). Size distribution for large and giant particles was calculated with the five concentrations. We also treated concentrations larger than 0.15 and 1 μm in radius as those of large and giant particles, respectively.

High concentrations of aerosol particles were sometimes caused by the ship exhaust. To avoid the influence of the ship exhaust, we omitted the data measured during a calm (relative wind speed less than 1 m/s) period and periods in which relative wind direction was from −135 to +45 degrees.

3. Results and Discussion

3.1. Latitudinal distributions of particle concentrations

Latitudinal distributions of Aitken particle, large particle and giant particle concentrations measured on legs 1 and 2, and legs 3 and 4, are shown in Figs. 2 and 3, respectively. Figure 2 shows that the concentration of Aitken particles decreased leaving Tokyo and recorded a minimum value around 10°N (5°–15°N) latitude. The concentration measured at 0°–10°S was somewhat high because the ship passed near islands. High concentrations at 43°S were measured at Port Lyttelton and Hobart. The concentration at a nearly fixed station (64°–65.5°S, 140°E) over the Southern Ocean was lower than that in the storm zone (around 50°S) but somewhat higher than that around 10°N.

Figure 3 shows that the concentration in the storm zone on leg 3 was lower than that measured in the same area on leg 2. In the storm zone, high concentrations greater than 1000 particles/cm^3 have been measured by some workers (KOJIMA and SEKIKAWA, 1974; IWAI *et al.*, 1979; ITO, 1983). The concentration in the storm zone strongly depends on

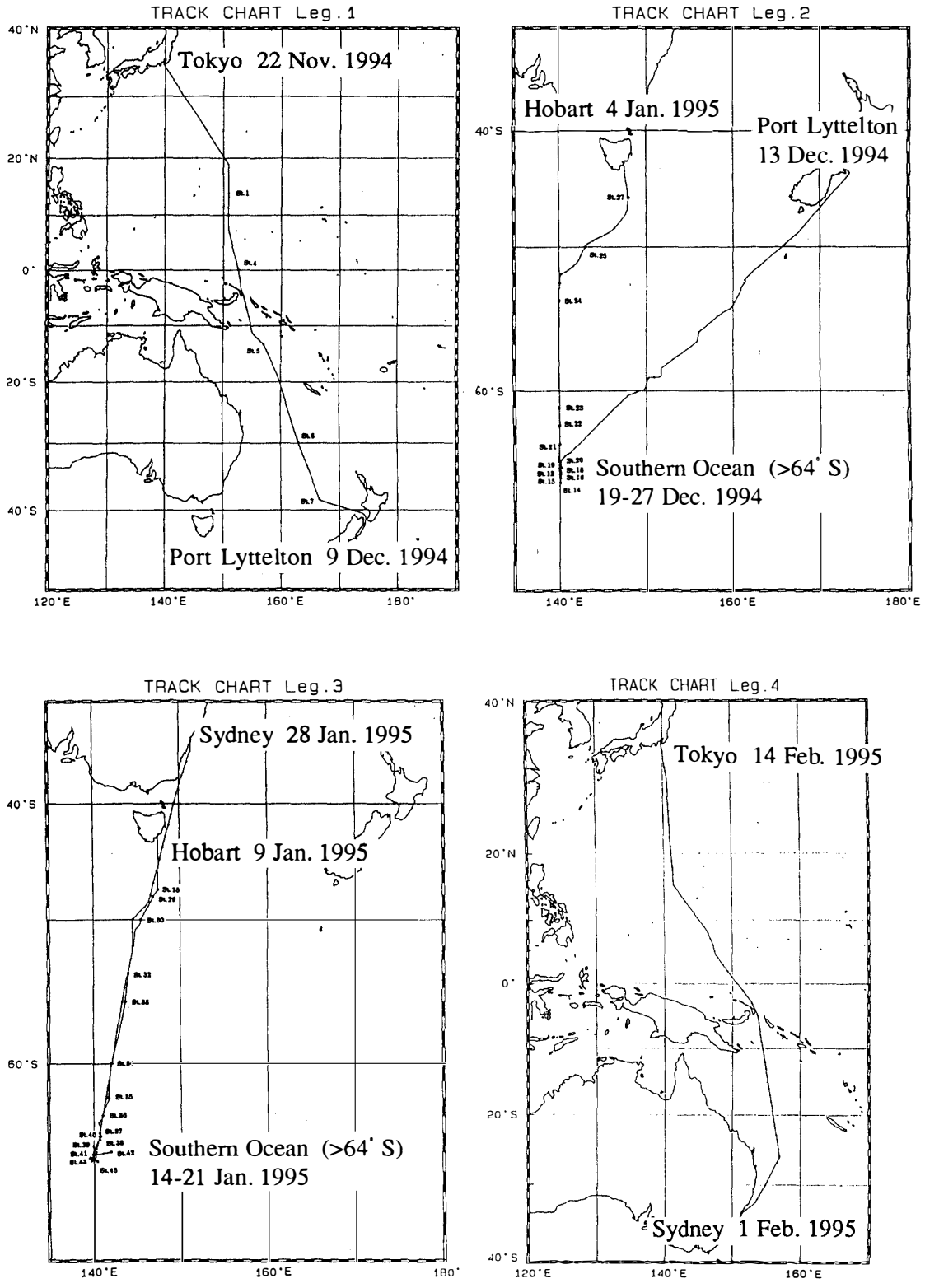

Fig. 1. Track charts on cruise KH-94-4. This expedition consisted of four legs. Departure and arrival dates, and observation period at the fixed station are listed in charts.

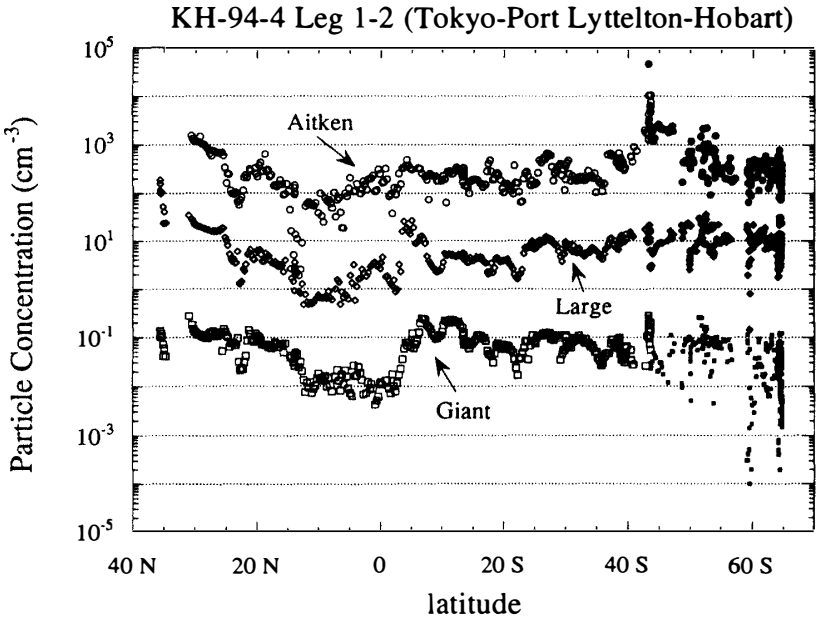

Fig. 2. The variations of concentrations of Aitken particles, large particles, and giant particles measured on legs 1 and 2 from Tokyo to Hobart through Port Lyttelton and the Southern Ocean. Open and closed marks show the data measured on legs 1 and 2, respectively.

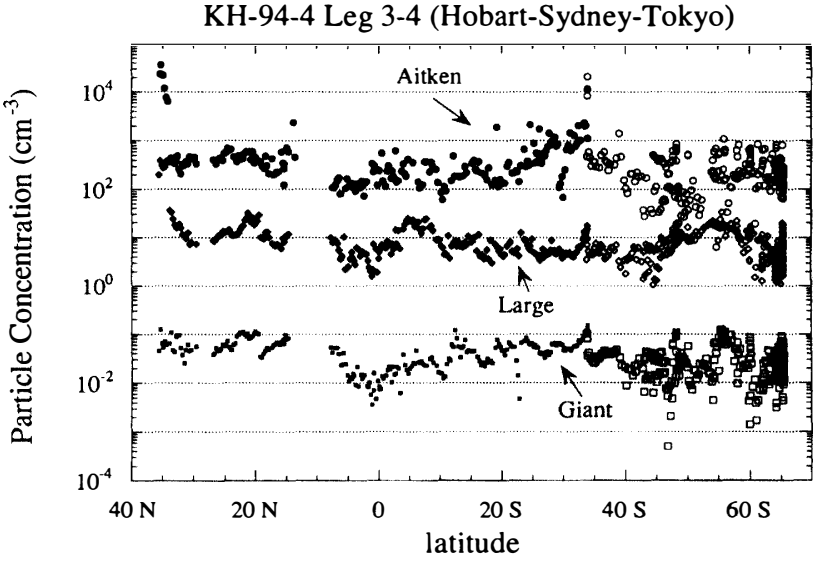

Fig. 3. The same as Fig. 2, but for legs 3 and 4 from Hobart to Tokyo through the Southern Ocean and Sydney. Open and closed marks show the data on legs 3 and 4, respectively.

the weather in the zone. High concentrations at 33°S were measured in Sydney. The concentrations measured on leg 4 north of 10°S were similar to those on leg 1.

We have measured latitudinal distributions of Aitken particle concentrations over the western Pacific five times. On these five legs (legs 1 and 2 of KH-91-5, leg 2 of KH-92-5 and legs 1 and 4 of KH-94-4), we usually recorded a minimum value (60–240

particles/cm^3) around 10°N (MIURA *et al.*, 1995b). The nearest island, New Guinea, is 1000 km from the sampling stations (5°–15°N, 144°–165°E). Moreover, strong showers sometimes scavenge aerosols. Therefore, low concentrations of Aitken particles are usually observed over this area.

Concentrations of large and giant particles had nearly similar variations to those of Aitken particles.

3.2. Particle concentrations over the Southern Ocean

Observations at the fixed station (64°–65.5°S, 140°E) over the Southern Ocean were carried out twice from 19 to 27 December 1994 on leg 2 and from 14 to 21 January 1995 on leg 3. Usually the weather in this area is stable, but it was not stable on leg 3.

The average, standard deviation and number of data are listed in Table 1. The average concentrations of Aitken particles were 280 and 196 particles/cm^3 on legs 2 and 3, respectively. They were lower than the value, 320 particles/cm^3, measured over the western equatorial Pacific Ocean during KH-92-5 (MIURA *et al.*, 1995a) but were somewhat higher than those measured around 10°N (60–240 particles/cm^3).

Table 1. Aerosol concentration (cm^{-3}) over the Southern Ocean (>64°S).

	Aitken		Large*		Giant**	
	leg 2	leg 3	leg 2	leg 3	leg 2	leg 3
Average value	280	196	7.535	6.119	0.021	0.035
Standard deviation	133	79	5.199	4.444	0.023	0.020
Number of data	97	78	103	84	103	84

* Particles with radii $\geqq$ 0.15 μm.
** Particles with radii $\geqq$ 1 μm.

Most Aitken particles over land are anthropogenic. But it is difficult to accept that the air over this area was transported from Australia. In the marine boundary layer over remote oceans, the composition of Aitken particles is sulfur-dominated (*e.g.*, MESZAROS and VISSY, 1974). These sulfur-dominant particles are produced by gas-to-particle conversion of organosulfur gases. They have been emitted from the ocean as dimethyl sulfide (DMS, CH_3SCH_3), and then converted into sulfur dioxide (SO_2), sulfuric acid (H_2SO_4), or methanesulfonic acid (CH_3SO_3H) (LOVELOCK *et al.*, 1972; HATAKEYAMA *et al.*, 1982). KIRST *et al.* (1993) reported that the correlation between chlorophyll, DMSP, DMS and concentration of Aitken particles was clearly evident.

In summer, the activity of the phytoplankton is high over the Southern Ocean, so that DMS concentration in air becomes high. ERICKSON *et al.* (1991) simulated concentration of Aitken particles from DMS emissions. Results of the simulation showed that the concentrations in January were 200–300 particles/cm^3 while the concentrations in July were 20–50 particles/cm^3 over the southern ocean.

On a clear day, photo-chemical reactions become active in the daytime so there is a regular diurnal variation of Aitken particle concentration. But on a cloudy day there is not a clear diurnal variation. Our results showed that the concentration on leg 2 was higher than that on leg 3. Moreover, the diurnal variation only appeared on leg 2 as shown in Fig. 4. This implies that the principal constituent of Aitken particles over the

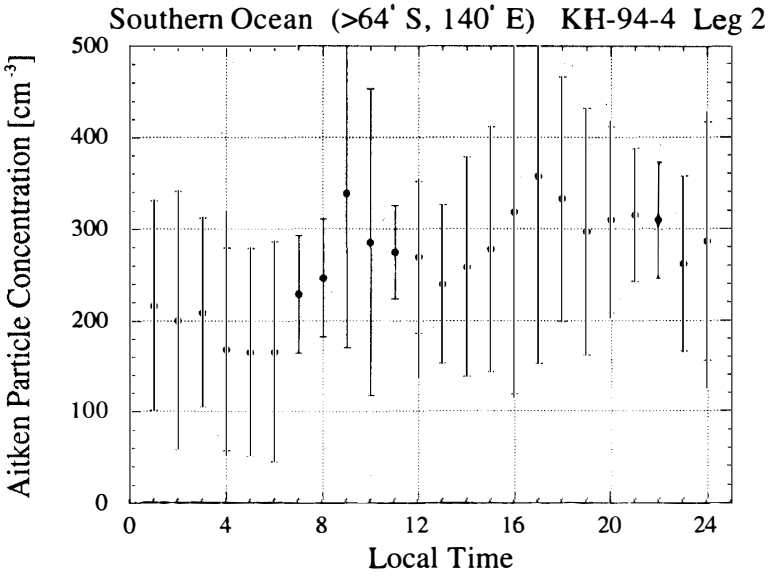

Fig. 4. Diurnal variation of Aitken particle concentration measured over the Southern Ocean (>64°S). Error bars show the standard deviations.

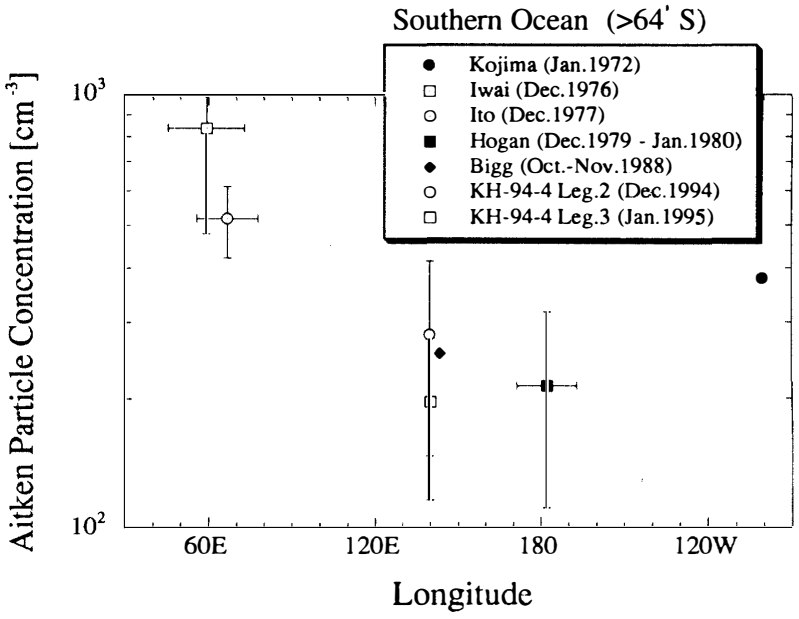

Fig. 5. Aitken particle concentration over the Southern Ocean (>64°S). Error bars show the standard deviations.

Southern Ocean in summer is sulfur-dominant particles produced by gas-to-particle conversion of organosulfur gases.

CIPRIANO *et al.* (1983) claimed from laboratory studies that even salt particles with radii less than 0.01 μm were produced. BIGG *et al.* (1995) found in the south central Indian Ocean that there is a mean relationship between wind speed and the concentrations of condensation nuclei. They showed that the emperical formula: $\log C = 1.25 + 0.037\,U$, where C is the particle concentration (particles/cm^3) and U is the wind velocity

in m/s, holds. When U is 10 and 20 m/s, C is 42 and 98 particles/cm^3, respectively. At the fixed station in our observation, the concentrations of Aitken particles were 200–300 cm^{-3}. And wind speed was weaker than 14 and 22 m/s on legs 2 and 3, respectively. This means that the wind-generated Aitken particles were not dominant. In fact, there is no correlation between the concentration of Aitken particles and wind force even on leg 3.

Figure 5 shows the concentration of Aitken particles measured south of 64°S on cruise KH-94-4 compared with other workers' measurements (KOJIMA and SEKIKAWA, 1974; IWAI *et al.*, 1979; ITO *et al.*, 1980; HOGAN, 1981; BIGG, 1990). These data were obtained from late spring to summer. Our values are in good agreement with the values measured by HOGAN (1981) and BIGG (1990) but lower than the values measured by

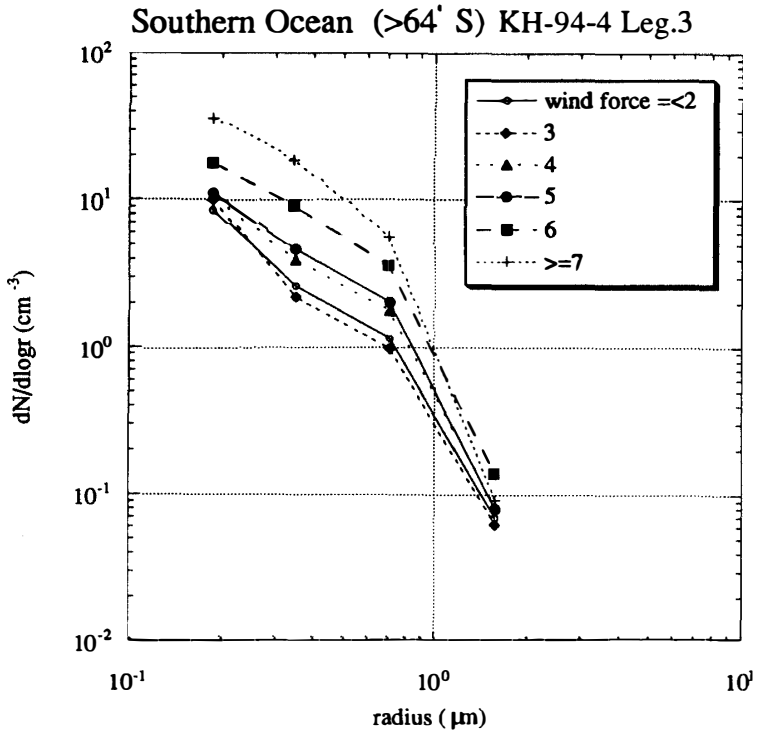

Fig. 6. The relation between size distribution for large and giant particles and wind force. The standard deviations are listed in Table 2.

Table 2. The average and standard deviation (in parentheses) of size distributions (dN/dlogr) classified by wind force measured at the station >64°S in the leg 3.

Wind force	Number of data	Radius (μm)			
		0.194	0.354	0.707	1.581
1,2	10	8.630 (2.445)	2.625 (1.249)	1.147 (0.600)	0.069 (0.038)
3	12	10.164 (3.023)	2.192 (0.862)	0.975 (0.483)	0.062 (0.037)
4	23	10.708 (4.604)	3.949 (2.443)	1.784 (1.084)	0.083 (0.046)
5	12	11.205 (5.109)	4.670 (2.831)	2.031 (1.147)	0.080 (0.037)
6	15	18.071 (6.073)	9.162 (2.981)	3.616 (0.714)	0.138 (0.065)
7≦	11	36.646 (6.775)	18.764 (3.894)	5.661 (1.785)	0.092 (0.044)

KOJIMA and SEKIKAWA (1974), IWAI *et al.* (1979) and ITO *et al.* (1980). The difference is not due to the longitude dependence but to the difference of weather conditions.

Figure 6 and Table 2 show size distributions for large and giant particles measured over this area on leg 3. They increased as wind force became strong. This relation has been previously reported (*e.g.*, KOJIMA and SEKIKAWA, 1974; MIURA *et al.*, 1995a) but it was first confirmed over the Southern Ocean. It suggests that the main constituent of large and giant particles was sea salt over the Southern Ocean.

4. Conclusion

It was found that the concentration of Aitken particles over the Southern Ocean was somewhat higher than that around 10°N. It is concluded that these aerosols were not transported from Australia but were produced by gas-to-particle conversion of organosulfur gases.

Moreover, we confirmed that wind force affects the size distribution for large and giant particles over the Southern Ocean. This suggests that large and giant particles over this area were mainly sea salt particles.

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