

# Study on Size Distributions of Airborne Particles by Aircraft Observation in Spring over Eastern Coastal Areas of China

WANG Wei\* (王 玮), LIU Hongjie (刘红杰), YUE Xin (岳 欣), LI Hong (李 红),  
CHEN Jianhua (陈建华), and TANG Dagang (汤大钢)

*Chinese Research Academy of Environmental Sciences, Beijing 100012*

(Received 16 August 2004; revised 3 February 2005)

## ABSTRACT

The authors studied the size distributions of particles at an altitude of 2000 m by aircraft observation over eastern coastal areas of China from Zhuhai, Guangdong to Dalian, Liaoning (0.47–30  $\mu\text{m}$ , 57 channels, including number concentration distribution, surface area concentration distribution and mass concentration distribution). In these cities, the average daily concentrations of PM<sub>10</sub> are very high. They are among the most heavily polluted cities in China. The main pollution sources are anthropogenic activities such as wood, coal and oil burning. The observed size distributions show a broad spectrum and unique multi-peak characteristics, indicating no significant impacts of individual sources from urban areas. These results are far different from the distribution type at ground level. It may reflect the comprehensive effect of the regional pollution characteristics. Monitoring results over big cities could to some extent reflect their pollution characteristics.

**Key words:** airborne particles, size distribution, aircraft observation, coastal areas, China

---

## 1. Introduction

From the 1980s on, as China's economy gained momentum, the emission amount of various air pollutants has been increasing rapidly, causing a series of air pollution problems (Wang et al., 2004). Although the Chinese government endeavors to reduce the emission of air pollutants, and in spite of the fact that the air quality in some cities and areas is becoming better, the overall air pollution level is still quite high, resulting in air pollution problems, such as acid rain, particulate matter pollution and photochemical pollution (Wang et al., 1988, 1997, 1998; Jiang et al., 1990). Of 113 main cities, 70% of them have air pollution problems. The primary air pollutant is mainly particulate matter, and the daily average concentration of PM<sub>10</sub> is over than 0.2 mg m<sup>-3</sup> (SEPA, 1996, 2003).

Atmospheric aerosol particles affect the Earth's radiative balance directly by scattering (Chylek and Coakley, 1974; Charlson et al., 1992; IPCC, 2001) or absorbing (Twoney, 1991; Chylek et al., 1996) light, and indirectly by acting as cloud condensation nuclei (CCN) (Hobbs, 1993; Meson et al., 2002) thereby influencing the albedo, lifetime, extent and precipitation

of clouds.

The Aerosol Characterization Experiments (ACE), organized by the International Global Atmospheric Chemistry (IGAC) Program, were designed to increase the understanding of how aerosols affect the global climate. The first Aerosol Characterization Experiment, ACE-1, was conducted over the South Pacific Ocean, south of Australia, in an attempt to quantify the chemical and physical processes that control properties of marine aerosols important for radiative forcing or climate considerations (Bates et al., 1998). The second Aerosol Characterization Experiment, ACE-2, took place over the Northeast Atlantic in order to study the properties and effects of background and anthropogenic pollution aerosols as well as dust in the marine boundary layer and free troposphere (Raes et al., 2000). The Asia Pacific Regional Aerosol Characterization Experiment, ACE-Asia, made simultaneous measurements of aerosol chemical, physical, and optical properties and their radiative impacts in a variety of air masses (Huebert et al., 2003). These aircraft measurements are important ways to study and master the laws of pollutant vertical distribution, and they

---

\*E-mail: wangwei@craes.org.cn

can contribute to further recognition of the impacts of airborne particles on climates.

At present, China has done much work on major air pollutants at ground-based sites and has obtained much valuable information (Qiu, 1995; Qian et al., 1998). However, due to the limit of facilities and budget, very few monitorings and studies based on aircraft have been done concerning vertical variation of air pollution. This to some degree restrains the understanding of air pollution on regional scales (Wang et al., 2000a, 2000b).

We carried out aircraft observation in the spring of 2002 to monitor air pollutants over almost the whole of China's eastern coastal areas. The monitored items include gaseous air pollutants ( $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{O}_3$  and VOCs) and particulate matter (filter samples of particulate matter with different diameters, chemical components, size distributions, CNC, etc). In this paper, we will focus on the aerodynamic size distributions of particulate matter and discuss the pollution status and pollution characteristics of airborne particulate matter.

## 2. Flight information and equipment

### 2.1 Flight information

In the spring of 2002, we launched 6 flights, using the Chinese aircraft Yun-5B single propeller light-duty cargo plane. The flights covered coastal areas from Zhuhai, Guangdong to Dalian, Liaoning. The total flight time is around 12 hours, and flight altitude is from the ground to 3000 m, with a mean altitude of about 2000 m. The airspeed is  $180 \text{ km h}^{-1}$ . Flight information and flight routes are shown in Table 1 and Fig. 1.

### 2.2 Equipment and installation

We used the TSI APS-3310A Aerodynamic Parti-

cle Sizer to sample particle size distributions. It consists of the Model 3310A Aerodynamic Particle Sizer Spectrometer (APS) and data analysis center. It uses 57 channels to analyze particles with diameters in the range  $0.43\text{--}30 \mu\text{m}$  and gives number size distribution automatically. The number size distribution, surface size distribution and mass size distribution are calculated on the assumption that sampled particles have the same sedimentation rate as global particles of the density of  $1 \text{ g cm}^{-3}$  with the same diameters.

Ambient air was introduced into the aircraft cabin through an inlet as shown in Fig. 2, and the structure of the TSI APS-3310A sampler can be seen in Fig. 3. Air was introduced into the instrument through a Tdrlon tube and stainless steel tube at a rate of  $5 \text{ L min}^{-1}$ . The inside of the tube was treated with glycerin in order to reduce the static charge and to further reduce the loss of particles.

However, the loss of particles is inevitable in the sampling process, especially for large particles. To examine the extent of loss caused by the bend, we modeled the transmission characteristics using methods from Pui et al. (1987) and Matsuki et al. (2003). And calculations from Muysshondt et al. (1996) were used to estimate the transmission efficiencies of the straight section. Calculations were made by assuming a flow velocity of  $1.04 \text{ m s}^{-1}$ , particle densities of 1.0, 2.0, 3.0  $\text{g cm}^{-3}$ , and particle diameters of 3.0, 5.0, 7.0, 10.0  $\mu\text{m}$ . Transmission efficiencies calculated for particles are shown in Tables 2 and 3. From these results, it can be seen that  $7\text{-}\mu\text{m}$  particles with a  $3.0\text{-g cm}^{-3}$  density would pass through the bent section with 77% efficiency, while they would be transmitted in the straight section with a 64% efficiency. This means that particle loss caused by deposition would be only slight.

**Table 1.** Flight information.

No.	Date (Mar 2002)	Time (LST)	Route	Start coordinates	End coordinates	Altitude (m)
1	1	0930–1103	Zhuhai–Shantou	22°23'61"N 113°39'65"E	23°01'45"N 116°23'39"E	2213
2	1	1316–1509	Shantou–Fuzhou	23°38'81"N 117°00'37"E	25°59'48"N 119°14'11"E	2119
3	6	1330–1512	Fuzhou–Huangyan	26°09'18"N 119°29'18"E	28°20'25"N 121°10'06"E	2090
4	7	0812–1042	Huangyan–Rugao	28°42'02"N 121°17'28"E	32°03'37"N 120°48'91"E	2370
5	7	1621–1814	Rugao–Qingdao	32°46'01"N 120°22'90"E	35°41'37"N 119°58'17"E	2300
6	8	1547–1728	Qingdao–Dalian	36°01'49"N 120°15'22"E	39°25'89"N 121°40'50"E	2098

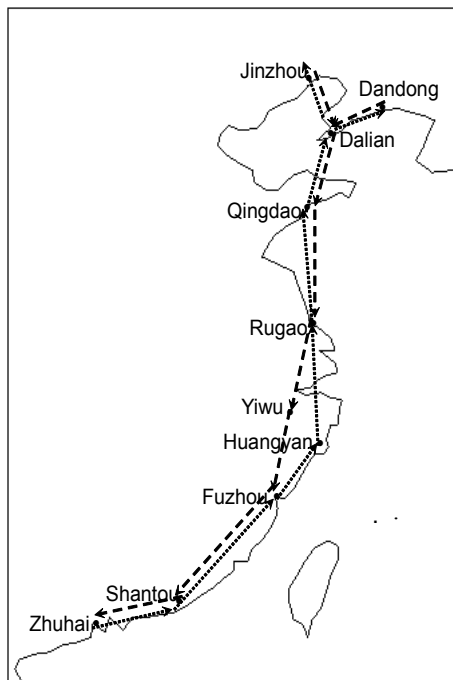


Fig. 1. Flight route.

Table 2. Estimated transmission efficiencies for the bent section of the inlet (at 20°C, 1013 hPa).

Particle diameter ( $\mu\text{m}$ )	Transmission efficiency at various densities (%)		
	1.0 g cm <sup>-3</sup>	2.0 g cm <sup>-3</sup>	3.0 g cm <sup>-3</sup>
3.0	98	97	95
5.0	96	91	87
7.0	92	84	77
10.0	83	70	58

Table 3. Estimated transmission efficiencies for the straight sections of the inlet (at 20°C, 1013 hPa).

Particle diameter ( $\mu\text{m}$ )	Transmission efficiency at various densities (%)		
	1.0 g cm <sup>-3</sup>	2.0 g cm <sup>-3</sup>	3.0 g cm <sup>-3</sup>
3.0	97	95	93
5.0	93	87	81
7.0	87	75	64
10.0	75	54	36

The two main parts of the sampler are the accelerating orifice and the laser velocimeter. Aerosol enters the outer inlet tube of the APS at a flow rate of 5 liters per minute. The flow is split to provide 1 liter per minute to the inner nozzle and 4 liters per minute of filtered sheath air to the outer nozzle. To measure the particle velocity, a laser beam is first split and focused into two rectangular beams in front of the orifice. The light scattered by a particle is focused through a series of lenses onto a photomultiplier tube. The tube emits two pulses separated by the time the particle crosses the distance between the two beams. This electronically-measured time interval is then used to calculate the aerodynamic diameter of the particle. The flow is maintained by a vacuum pump that draws aerosol through the system. The flow in this feedback loop is controlled by the total flow potentiometer on the front panel of the APS. The sheath-air valve manually controls the ratio of sheath air to aerosol flow (inner nozzle). These two flows must be maintained to keep the APS calibrated.

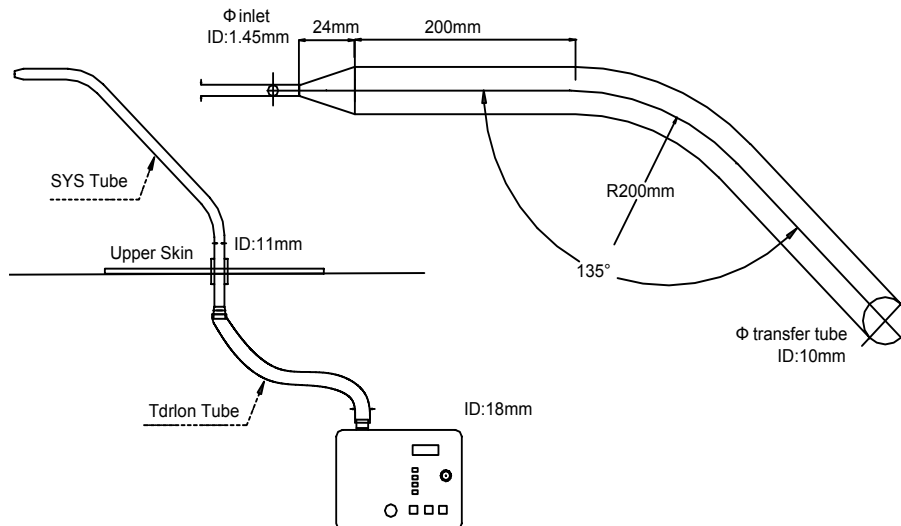


Fig. 2. Schematic diagram of the inlet used in the aircraft-borne measurements.

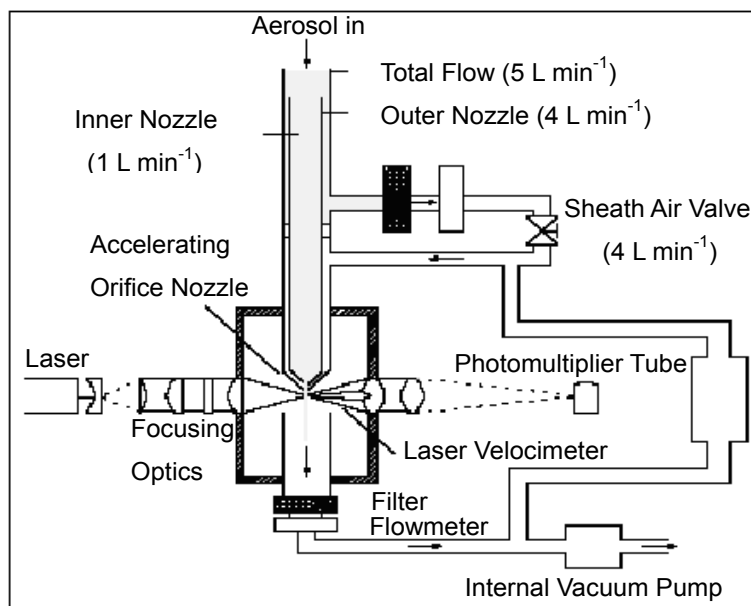


Fig. 3. The structure of the TSI APS-3310A sampler.

In order to avoid the influence of flight speed on sampling, we use a cone-shaped isokinetic inlet. Furthermore, the inlet is placed 0.5 m higher than the propeller rotation circle to avoid the impact of turbulence on particle sampling.

### 3. Results and discussions

#### 3.1 Aerodynamic particle size distributions

Figure 4 to 9 give the representative aerodynamic particle size distributions during the 6 flights (number size distribution, surface area size distribution and mass size distribution, where size is given by diameter  $D$ , number concentration by  $N$ , surface area concentration by  $S$  and mass concentration by  $M$ ).

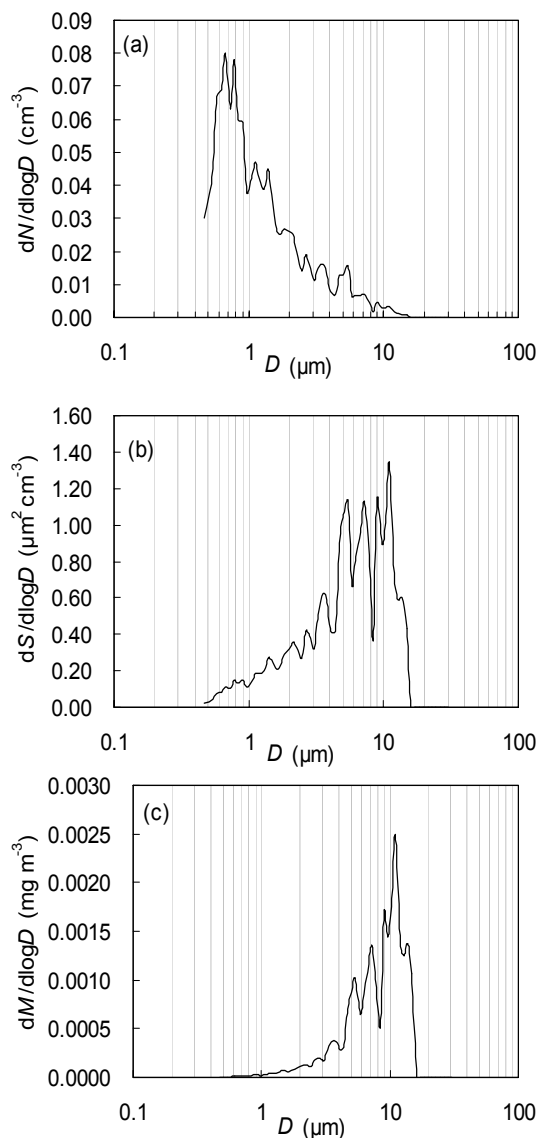
From Figs. 4, 5, 6, and 9, we can see that during the flights, the number size distributions at 2000 m appear to be multi-peak distributions; the diameters range between 0.6 and 0.9  $\mu\text{m}$ ; the main peak usually stays around 0.7–0.8  $\mu\text{m}$ . Such distributions are different from those obtained in an urban environment, where there are usually several small peaks at sizes over 1  $\mu\text{m}$ . The surface area size distributions appear to be irregular partial lognormal distributions with a single peak or multiple peaks; the single peak also contains several small peaks while the multi-peak spread sometimes has 7–8 peaks. The diameters corresponding to peaks vary greatly, usually appearing at 1.1–1.4  $\mu\text{m}$  and at around 10  $\mu\text{m}$ . The mass size distributions appear to be partial lognormal multi-peak distributions as well. They have peaks at around 1.5–10.5  $\mu\text{m}$  with the main peak often occurring at 10  $\mu\text{m}$ .

We can also notice that the number concentration, surface area concentration and mass concentration are far lower than those at ground level.

Figures 7 and 8 show the results obtained close to Shanghai and Qingdao, which are different from the above results. Over Shanghai, the number size distributions and surface area size distributions appear to be ordinary single peak distributions while the mass size distributions appear to be partial lognormal 2-peak distributions (Lestari et al., 2003). Over Qingdao, the number size distributions are single peak distributions with 3 small embedded peaks; the surface area size distributions are partial lognormal 4-peak distributions and the mass size distributions are 3-peak partial lognormal distributions with several small peaks. We can notice that these 2 flights were performed close to urban areas, with single peak distributions for number size distributions and surface area size distributions and higher concentrations than the others (Chen et al., 2003).

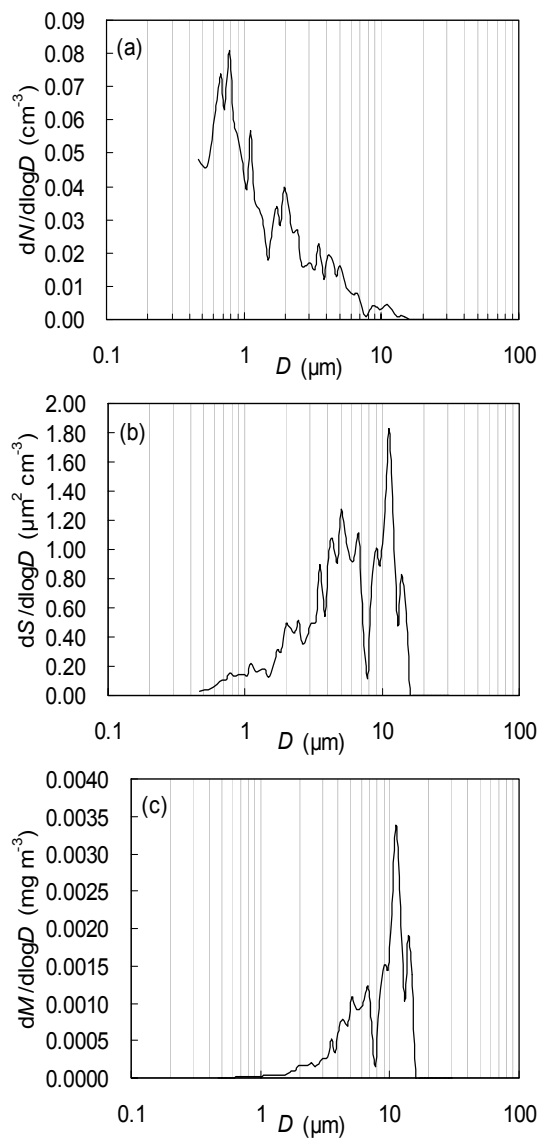
#### 3.2 Median diameters and sum of concentrations

Table 4 shows the calculated sums of number concentrations, surface area concentrations and mass concentrations and their median diameters. From the table, we can see that the median diameters of number concentrations are similar at around 1  $\mu\text{m}$ . These diameters are aerodynamic diameters, and if we consider that the relative density of these particles is more than 1  $\text{g cm}^{-3}$ , then the geometric diameters of these particles should be less than 1  $\mu\text{m}$ , at around 0.3–0.5  $\mu\text{m}$ .



**Fig. 4.** Size distributions at 2000 m during the flight from Zhuhai to Shantou on a March 2002 (a) number concentration; (b) surface area concentration; (c) mass concentration (the average of 60 measured concentrations, each sampled for one minute).

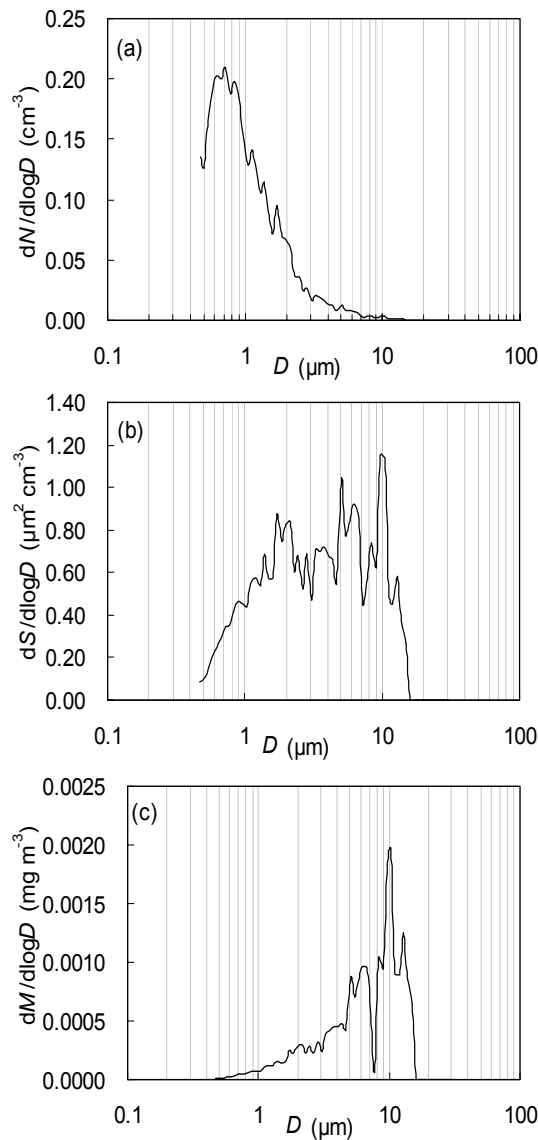
These particles are small particles that may be composed of carbon-laden particles with diameters less than  $0.3 \mu\text{m}$  and sulfates and nitrates with diameters around  $0.7\text{--}0.8 \mu\text{m}$  (Morawska et al., 1998, 1999). We can believe that large particles have no significant contribution to number concentrations during the flights. The median diameters of surface area concentrations vary greatly between flights, from  $5.77 \mu\text{m}$  to  $1.75 \mu\text{m}$ . We think the surface area concentrations and their median diameters represent the comprehensive properties of small particles and large particles. When the median diameters are large (small), they represent large



**Fig. 5.** Size distributions at 2000 m during the flight from Shantou to Fuzhou on 1 March, 2002 (a) number concentration; (b) surface area concentration; (c) mass concentration (the average of 60 measured concentrations, each sampled for one minute).

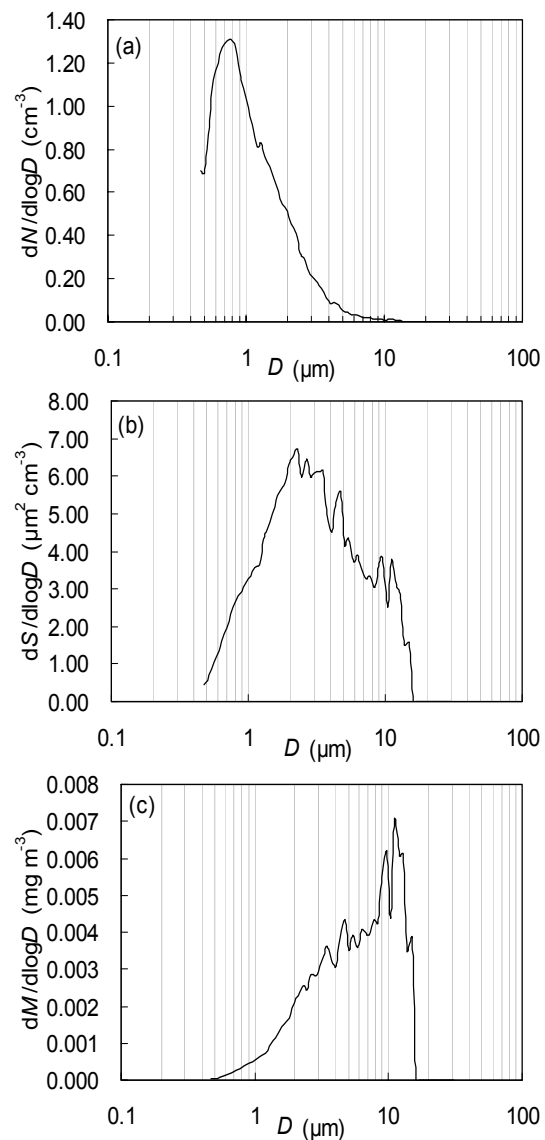
(small) particles. The median diameters of mass concentrations are larger than those of number concentrations and surface area concentrations, varying from  $9.40 \mu\text{m}$  to  $3.19 \mu\text{m}$ . The reason the median diameters of mass concentrations are larger than those two is that larger particles usually have larger mass. Even though there are many small particles, they cannot compete with large particles in mass. The mass concentrations and their median diameters represent the properties of large particles.

We can see that the pollution status of the first 3 flights is similar while the pollution characteristics are



**Fig. 6.** Size distributions at 2000 m during the flight from Fuzhou to Huangyan on 6 March 2002 (a) number concentration; (b) surface area concentration; (c) mass concentration (average of 60 measured concentrations, each sampled for one minute).

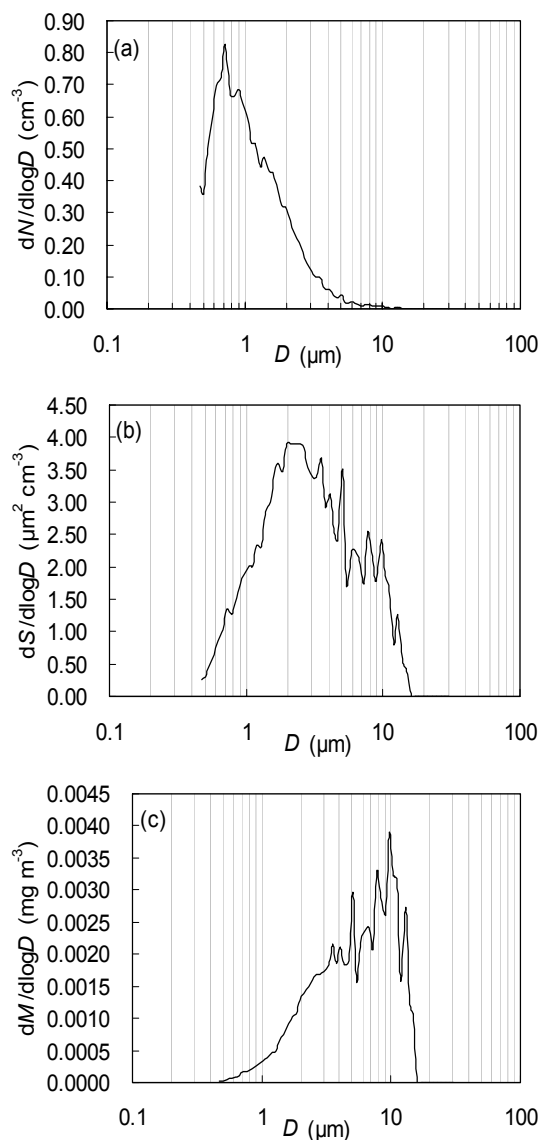
slightly different. The median diameters are small when number concentrations are high. This indicates that there are many small particles constituting the number concentrations. During the flight on 6 March 2002, the number concentrations and surface area concentrations are high, yet due to their smaller median diameters, the mass concentrations are lower than the other 2 flights. All concentrations are low during the flight from Qingdao to Dalian on 3 March 2002, showing good dispersion and dilution conditions.



**Fig. 7.** Size distributions at 2000 m during the flight from Huangyan to Rugao on 7 March 2002 (a) number concentration; (b) surface area concentration; (c) mass concentration (average of 15 measured concentrations, each sampled for one minute)

### 3.3 Comprehensive discussion

Although all the flights are performed at around 2000 m, the size distributions are quite different (Zaizen et al., 1996; Cztrowszky et al., 1996; Lü et al., 2004). In most cases, when the aircraft flew over urban areas or seriously polluted areas, there were fewer peaks in the distributions and the concentrations were higher. This on the one hand implies the relatively simple sources of the particles; on the other hand it reflects the pollution characteristics and their impacts on particles at higher altitudes (Pakkanen et al., 2001).

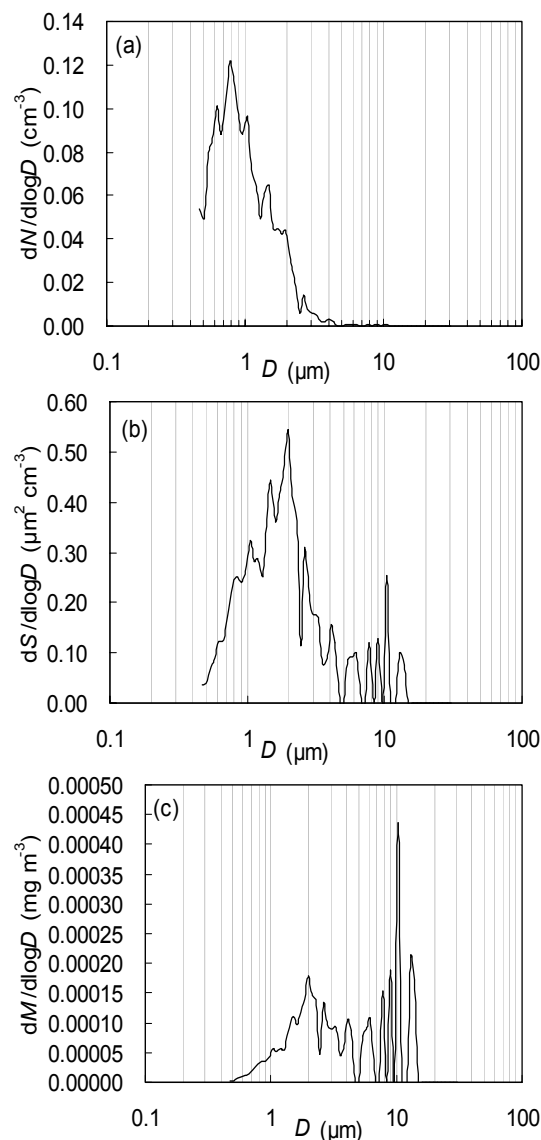


**Fig. 8** Size distributions at 2000 m during the flight from Rugao to Qingdao on 7 March 2002 (a) number concentration; (b) surface area concentration; (c) mass concentration (average of 15 measured concentrations, each sampled for one minute).

Because there are many particles concentrated in a certain diameter range, the contribution of particles in other diameter ranges is somehow masked. But over clean areas the distributions often appear to be multi-peak distributions, showing from 5 to 10 peaks. Such distributions are rarely seen at ground sites, showing the important pollution characteristics at higher altitudes.

#### 4. Conclusions

(1) The aircraft observation carried out in the



**Fig. 9** Size distributions at 2000 m during the flight Qingdao to Dalian on 8 March 2002 (a) number concentration; (b) surface area concentration; (c) mass concentration (average of 60 measured concentrations, each sampled for one minute).

spring of 2002 obtains very different results compared to ground observations, showing specific pollution characteristics.

(2) The size distributions over relatively clean areas show multi-peak distributions, which indicates the complex sources and complicated influence factors. These represent the regional pollution characteristics. Furthermore, there are no impacts like in urban areas of simple sources on the distributions; the distributions appear to have a broad spectrum. Such kind of distribution is rarely seen at ground sites.

(3) When close to urban areas, the distributions

**Table 4.** Sums of number concentrations, surface area concentrations and mass concentrations and their median diameters.

Date (Mar 2002)	Route	Number		Surface area		Mass	
		Concentration ( $\text{cm}^{-3}$ )	Median diameter ( $\mu\text{m}$ )	Concentration ( $\mu\text{m}^2 \text{cm}^{-3}$ )	Median diameter ( $\mu\text{m}$ )	Concentration ( $\mu\text{g m}^{-3}$ )	Median diameter ( $\mu\text{m}$ )
1	Zhuhai–Shantou	1.24	1.01	22.5	5.77	23.8	8.87
1	Shantou–Fuzhou	1.30	1.06	25.8	5.52	27.7	9.40
6	Fuzhou–Huangyan	3.31	0.90	28.7	3.52	22.3	7.51
7	Huangyan–Rugao	22.30	0.97	190.0	2.81	126.0	6.05
7	Rugao–Qingdao	12.90	0.99	110.0	2.71	69.3	5.64
8	Qingdao–Dalian	1.67	0.91	8.5	1.75	3.6	3.19

are similar to those at ground level. The concentrations are also higher than those over clean areas. This is the influence and contribution of urban emission.

**Acknowledgments.** This work was supported by the national 973 projects (2002CB211600, 2002CB410800) and Social Public Welfare Project from MOST (2002DIA20012, ABC project).

## REFERENCES

- Bates, T., B. J. Huebert, and J. L. Gras, 1998: International Global Atmospheric Chemistry (IGAC) project's first Aerosol Characterization Experiment (ACE 1): Overview. *J. Geophys. Res.*, **103(D13)**, 16 297–16 318.
- Charlson, R. J., S. E. Schwartz, and J. M. Hales, 1992: Climate forcing by anthropogenic aerosols. *Science*, **255**, 423–430.
- Chen Longxun, Zhu Wenqin, Zhou Xiuji, and Zhou Zijiang, 2003: Characteristics of the Heat Island Effect in Shanghai and Its Possible Mechanism. *Adv. Atmos. Sci.*, **20**, 991–1001.
- Chylek, P., and J. A. Coakley, 1974: Aerosols and climate. *Science*, **183**, 75–77.
- Chylek, P., G. B. Lesins, and G. Videen, 1996: Black Carbon and absorption of solar radiation by clouds. *J. Geophys. Res.*, **101(23)**, 265–271.
- Cztrvoszky, A., P. L. Czonka, P. Jani, A. Ringelhann, and J. Bobvos, 1996: Experimental Investigation of Altitude Dependence of Size Distribution and Concentration of Dust Particles within the City of Budapest. *J. Aerosol Sci.*, **27**, 117–118.
- Hobbs, P. V., 1993: *Aerosol–cloud–climate interactions*, Academic press, San Diego, California.
- Huebert, B. J., T. Bates, B. R. Philip, G. Y. Shi, Y. J. Kim, K. Kawamura, G. Carmichael, and T. Nakajima, 2003: An overview of ACE-Asia: Strategies for quantifying the relationships between Asian aerosols and their climatic impacts. *J. Geophys. Res.*, **108(D23)**, 8633, doi: 1029/2003JD003500.
- IPCC, 2001: *Climate Change 2001: The Scientific Basis, Contribution of Working Group 1 to the Third Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge Univ. Press, New York, 892pp.
- Jiang Zhenyuan, Wang Wei, and Tang Dagang, 1990: Study on Size Distribution of China's Aerosols. *Academic Proc. Chinese Research Academy of Environmental Sciences*, 70–79. (in Chinese)
- Lestari, P. J., A. K. Oskouie, and K. E. Noll, 2003: Size distribution and dry deposition of particulate mass, sulfate and nitrate in an urban area. *Atmos. Environ.*, **37**, 2507–2516.
- Lü Daren, Yi Fan, and Xu Jiyao, 2004: Advances in studies of the middle and upper atmosphere and their coupling with the lower atmosphere. *Adv. Atmos. Sci.*, **21**, 361–368.
- Matsuki, A., and Coauthors, 2003: Seasonal dependence of the long-range transport and vertical distribution of free tropospheric aerosols over East Asia: On the basis of aircraft and lidar measurements and isentropic trajectory analysis. *J. Geophys. Res.*, **108(D23)**, 8663, doi: 10.1029/2002JD003266.
- Menon, S., J. Hansen, and L. Nazarenko, 2002: Climate effects of black carbon aerosols in China and India. *Science*, **297**, 2250–2253.
- Muysshondt, A., N. K. Anand, and A. R. McFarland, 1996: Turbulent deposition of aerosol particles in large transport tubes. *Aerosol Sci. Technol.*, **24**, 107–116.
- Morawska, L. D., S. Thomas, N. Bofinger, D. Wainwright, and D. Neale, 1998: Comprehensive characterization of aerosols in a subtropical urban atmosphere: particle size distribution and correlation with gaseous pollutants. *Atmos. Environ.*, **32**, 2467–2478.
- Morawska, L. D., S. Thomas, M. Jamriska, and G. Johnson, 1999: The modality of particle size distributions of environmental aerosols. *Atmos. Environ.*, **33**, 4401–4411.
- Pakkanen, T. A., V. M. Kerminen, C. H. Korhonen, R. E. Hillamo, P. A. Aarnio, T. Koskentalo, and W. Maenhaut, 2001: Use of atmospheric elemental size distributions in estimating aerosol sources in the Helsinki area. *Atmos. Environ.*, **35**, 5537–5551.
- Pui, D. Y. H., R. N. Francisco, and B. Y. H. Liu, 1987: Experimental study of particles deposition in bends of circular cross section. *Aerosol Sci. Technol.*, **7**, 301–315.
- Qian Yun, Wang Hongqi, and Fu Congbin, 1998: Seasonal and spatial variation of radiative effects of Anthropogenic sulfate aerosol. *Adv. Atmos. Sci.*, **15**, 380–392.



- Qiu Jinhuan, 1995: Two-wave length lidar measurement of cloud-aerosol optical properties. *Adv. Atmos. Sci.*, **12**, 177–186.
- Raes, F., T. Bates, and F. McGovern, 2000: The 2nd Aerosol Characterization Experiment (ACE 2), General overview and main results. *Tellus (B)*, **52**, 111–125.
- SEPA (The China State Environmental Protection Administration), 1996: National Ambient Air Quality Standards (GB 3095–1996). [Available from: URL: <http://www.es.org.cn/download/571-1.pdf>]. (in Chinese)
- SEPA (The China State Environmental Protection Administration), 2003: Report of China's Environmental Quality. [Available from: URL: <http://www.gyce.cn/ReadNews.asp?NewsID=1437>]. (in Chinese)
- Twoney, S., 1991: Aerosols, clouds and radiation. *Atmos. Environ., Part (A)*, **25**, 2435–2442.
- Wang MingXing, Liu Qiang, and Yang Xin, 2004: A review of research on human activity induced climate change I. Greenhouse gases and aerosols. *Adv. Atmos. Sci.*, **21**, 314–321.
- Wang Wei, Liu Hongjie, and Zhang Yutian, 1998: Study on the aerosol pollution characteristics in the desert areas-Mass. Proc. 6th International Symposium on Atmospheric Sciences and Applications to Air Quality. Beijing, 507–521.
- Wang Wei, Tang Dagang, and Liu Hongjie, 2000a: Aircraft measurement of atmospheric pollutants in Winter in North China I: Research on pollution properties of gas pollutants. *Research of Environmental Sciences*, **13**, 6–9. (in Chinese)
- Wang Wei, Tang Dagang, and Liu Hongjie, 2000b: Aircraft measurement of atmospheric pollutants in Winter in North China II: Research on pollution properties of atmospheric aerosol. *Research of Environmental Sciences*, **13**, 10–13. (in Chinese)
- Wang, W., K. Sakamoto, and W. M. Wang, 1997: Pollution characteristic of atmospheric aerosol and its relations to acid rain at the southern area of Fujian province in China. *Journal of Japan Society for Atmospheric Environment.*, **32(4)**, 204–215.
- Wang Wei, Wang Wenxing, and Zhao Deshan, 1988: Study on impacts of aerosols on precipitation. *Research of Environmental Sciences*, **1**, 38–44. (in Chinese)
- Zaizen Y., M. Ikegami, Y. Tsutsumi, Y. Makino, K. Okada, J. Jensen, and L. G. John, 1996: Number concentration and size distribution of aerosol particles in the middle troposphere over the western Pacific ocean. *Atmos. Environ.*, **30**, 1755–1762.