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Number concentration and size distributions of submicron particles in Jinan urban area: Characteristics in summer and winter

GAO Jian[1], WANG Jin[1,*], CHENG Shu-hui[1], XUE Li-kun[1], YAN Huai-zhong[2], HOU Lu-jian[2], JIANG Yu-quan[3], WANG Wen-xing[1]

1. Environment Research Institute, Shandong University, Jinan 250100, China. E-mail: duncan_gao@yahoo.com
2. Environment Monitoring Station of Jinan, Shandong Province, Jinan 250014, China
3. Shandong Technical College of Labor and Vocation, Shandong Province, Jinan 250022, China

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Abstract

The aerosol number concentration and size distribution were measured with the newly developed Wide-range Particle Spectrometer in summer and winter of 2006 at the urban site of Jinan City. Here reported the characteristics of fine particles of the different observation seasons. Relative high number concentrations for the particles in the diameter range of 10–500 nm were observed in both seasons. It was found that the dominant number distributed in particle diameter smaller than 100 nm and the percentage over the number concentration of all air particles is much higher than what has been measured in other urban sites over the world. The number mean diameter in summer was much smaller than in winter, strongly suggesting the different origin of ultrafine particles in different seasons. That is, particles in ultrafine mode mainly came from nucleation and new particle formation in summer while from traffic emission in winter. The diurnal variation also supported this point. Number concentration in the diameter range of 10–200 nm got their peak values at noontime, well correlated with the mixing ratio of SO_2 and the intensity of solar radiation in summer. While in winter, those in the same diameter range showed the main peaks during the traffic hours happened in the morning and evening.

Key words: number concentration; size distribution; seasonal variation; diurnal variation

Introduction

Ambient fine particulate matter has attracted major public health and atmospheric science concern in the recent years. Especially, the size distribution of atmospheric aerosols, together with their composition, sources, and sinks, is a key element in understanding and managing aerosol effects on health (Harrison *et al*., 1999; Samet *et al*., 2000; Wichmann and Peters, 2000; Lippmann *et al*., 2000), visibility (Stott *et al*., 2000; Ramanathan *et al*., 2001; Yu and Saxena, 2001; Menon *et al*., 2002), and climate (Seinfeld and Pandis, 1998; Oberdorster *et al*., 1996).

There have been many sampling efforts to measure aerosol number concentrations and size distributions in urban, rural and remote sites around the globe and more than 100 publications have been reported on observations of ultrafine particles in the atmosphere (Kumala *et al*., 2004). Some recent continental sampling campaigns that measured size distributions include the sampling campaigns in Europe (Ruuskanen *et al*., 2001; Kikas *et al*., 1996; Harrison *et al*., 1999; Birmili *et al*., 2001), North America (Kim *et al*., 2002; Cheng and Tanner, 2002; Woo *et al*., 2001) and Australia (Morawska *et al*., 2002). Most of the studies reported 24 h average number concentrations (10–500 nm) at continental sites ranging from around 5000–25000 cm^{-3}. Sites that have monitored 3–10 nm particles find comparable numbers of particles in that size range relative to the 10–500 nm size range (Woo *et al*., 2001).

Rapid urbanization and industrial developments in China in the past two decades have led to a significant increase of emission in both particle mass concentrations and its relative gas precursors (Streets and Waldhoff, 2000). Experienced a rapid increase in the use of vehicles, concurrent with large increases in energy consumption and high concentration of particle precursors give rise to new particle formation and nucleation and this will likely foster severe aerosol pollution in the major urban centers and the surrounding regions. Researchers have made relative studies on particle number size distribution in urban area of mega-city in China in the past decade. Shandong Province has been recorded to be the largest contributor for SO_2 in China and SO_2 has been thought to be the major precursor for newly formed particles (Kulmala *et al*., 2004). However, there is no observation

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E-mail: wangjin@sdu.edu.cn.

focuses on study of size distribution of ultrafine particle and particle formation and growth conducted over this region. Jinan is the capital of Shandong Province, with approximately 6 million inhabitants, facing serious air pollution problems which have been characterized by high concentrations of particulate matter and sulfur dioxide. In May and November of 2006 we conducted measurements of particle size distribution in the diameter scale from 10 to 10000 nm together with real time measurement of trace gases (SO_2, NO_x, CO) and mass concentration of PM_{10}. This site supplies us the good opportunity to sample air masses of urban area and make us have a better understanding about the characteristics of ultrafine particle in the influence of urban atmosphere. Another purpose of the study was to collect size distribution data in urban area of China for comparison with measurements concurrently made at the similar sites in global scale.

1 Methodology

1.1 Site description

The main sample location is on the top floor of Jinan Environment Monitoring Station, about 20 m high above ground level, 5 km east of downtown area. The site represents an urban site but with few significant local sources of emissions comparing with some industry areas. However, the concentrations of pollutant compounds in gaseous and condensed phase can be assumed to be strongly affected by the traffic due to the location very near from the main road of the district. And located near the inhabitation area, the pollutants from cooking emission will not be escapable.

1.2 Instrumentation

The Wide-range Particle Spectrometer (WPS^{TM}-model 1000XP) (MSP Corporation, USA) was used to measure size distribution data in the field study of this work. The instrument is a high-resolution aerosol spectrometer which combines the principles of differential mobility analysis (DMA), condensation particle counting (CPC) and laser light scattering (LPS). The DMA in the WPS^{TM} has a cylindrical geometry with an annular space for the laminar aerosol and sheath air flows. These critical dimensions were optimized to obtain size classification of particles between 10 and 500 nm in up to 96 channels with a maximum voltage smaller than 10000 volts, a sheath flow rate of 3 L/min. The CPC is of the thermal diffusion type, with a saturator maintained at 35°C. The LPS is a single-particle, wide-angle optical sensor used for measuring particle size from 0.35 to 10 μm in 24 additional channels. Particles are drawn into the aerosol inlet at a flow rate of 0.70 L/min and focused with a 3 L/min flow of sheath air towards the center of a laser beam generated by a laser diode. Before and after the observation campaign, the DMA was calibrated with NIST SRM 1691 and SRM 1963 PSL spheres (0.269 and 0.1007 μm mean diameter) to verify proper DMA transfer function and accurate particle sizing traceable to NIST. The LPS is calibrated with four NIST traceable sizes of PSL (0.701, 1.36, 1.6, and 4.0 μm mean diameter).

The mass concentrations of PM_{10} were monitored continuously by a real time ambient particulate monitor (Met One Instrument Inc., BAM-1020). CO, SO_2 and NO_x used in this study were supplied by the Environmental Monitoring Station of Jinan, using nondispersive infrared analyzer (Advanced Pollution Instrumentation, model 300A), pulsed UV fluorescence analyzer (API model 200A) and Chemiluminescence analyzer (API Model 100A) separately.

2 Results and discussion

2.1 Number concentration and size distribution

The results discussed in this study cover the data gained from 10 May to 21 May 2006 (summer) and 13 November to 30 November 2006 (winter). Considering that the number concentration was dominated by ultrafine particles (Seinfeld and Pandis, 1998), here we put our attention on the particles of accumulation mode and those smaller. The particles in the diameter from 10–1000 nm were divided into 6 sub size ranges: 10–20 nm (nuclei mode), 20–50 nm and 50–100 nm (Atkien mode), 100–200 nm and 200–500 nm and 500–1000 nm (accumulation mode).

Various descriptive statistics regarding the measured half hour average number concentrations in the series of sub size bins are included in Table 1. It can be seen that the number concentration of 10–20 nm mode particles is much higher in summer than that in winter (as high as 7374 cm^{-3} in summer and 3839 cm^{-3} in winter). As above

Table 1 Descriptive statistics of the measured number concentrations in the series of sub size bins in summer and winter

Diameter (nm)	10–20	20–50	50–100	100–200	200–500	500–1000
Summer						
Mean (cm^{-3})	7374	2288	639	217	168	96
Median (cm^{-3})	3218	984	278	100	143	59
Max. (cm^{-3})	58350	17233	5355	2144	659	432
Min. (cm^{-3})	425	131	34	10	17	7
SD	10000	3093	863	298	112	93
Winter						
Mean (cm^{-3})	3839	8252	3499	1268	528	21
Median (cm^{-3})	3151	7278	2805	878	354	12
Max. (cm^{-3})	15378	35067	17333	6183	2410	134
Min. (cm^{-3})	450	1125	274	152	54	1
SD	2584	4963	2477	1004	482	25

mentioned, most of observation studies at continental sites reported 24 h average number concentrations (10–500 nm) ranging from 5000–25000 cm^{-3}, and Table 2 supplies the comparison with the number concentration in the continental sites. With the consideration of the number concentration over the size range, that is 10685 cm^{-3} in summer and 17387 cm^{-3} in winter, the Jinan City has experienced relative serious pollution of ultrafine particles.

What should be noticed is the number size distribution in different seasons. Fig.1 shows the number size distribution during the summer and winter observation. It is evident that number concentration was dominated by particles smaller than 20 nm in summer while during winter observation the peak of number size distribution is located around 35 nm. Stanier *et al.* (2004) reported the mode of 40 nm observed in Pittsburg, suggesting that more particles in ultrafine mode in the Jinan City than in Pittsburg. With the consideration of the detection limit of the instrument in this study, we only measured the particle counts of those larger than 10 nm, it can be inferred that the particles might have dominate percentage in the size of 10 nm or smaller during summer season. Note that the high temperature and strong solar radiation intensity during summer month, the favorite meteorological condition may strongly promote the photochemical production and homogeneous nucleation of particle. Most of these newly formatted particles are in the size range of 3–10 nm. While in winter, the major peak around 35 nm strongly suggested a different source for the ultrafine particles. Without enough photochemical promotion under winter condition, less convection and stronger emission of primary particles in heating period may yield the formation of nano-particles. Previous studies also show a pronounced mode of particles in the 10–40 nm range from modern diesel engine exhaust (Kittelson *et al.*, 2000). Here the strong association between particle concentration and particles around 35 nm may support the hypothesis that vehicular or burning emissions are the major source of ultrafine particles.

Table 2 Comparison with the number concentration results in other continental sites

Observation site	Number counts (cm^{-3}, 12 h average)		Reference
	10–100 nm	100–500 nm	
Alkmaar, Netherlands	18300	2120	Ruuskanen *et al.*, 2001
Erfurt, Germany	17700	2270	Wichmann and Peters, 2000
Helsinki, Finland	16200	973	Ruuskanen *et al.*, 2001
Pittsburgh, Urban, USA	14300	2170	Stanier *et al.*, 2004
Pittsburgh, Rural, USA	6500	1900	Stanier *et al.*, 2004
Jinan, China (summer)	10300	385	This work
Jinan, China (winter)	15591	1796	This work

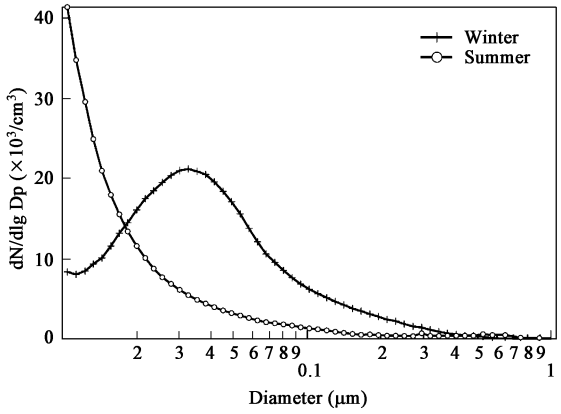

Fig. 1 Number size distribution of submicron particles in winter and summer.

Figure 2 show the average fractions of the particle number, surface and mass concentrations in the series of sub size bins from 10 nm to 2.5 μm in the two seasons. The surface and mass concentration were calculated with the assumption that the particles were spherical. It is evident that the mass concentrations are dominated by particles larger than 100 nm, which contribute 99.3% of the total volume in summer and 93% in winter. While the characteristic of particles smaller than 100 nm is not only their high number per unit of air volume but they also account for preponderance of the number of all urban air particles (up to 95.4% of the number concentrations in summer and

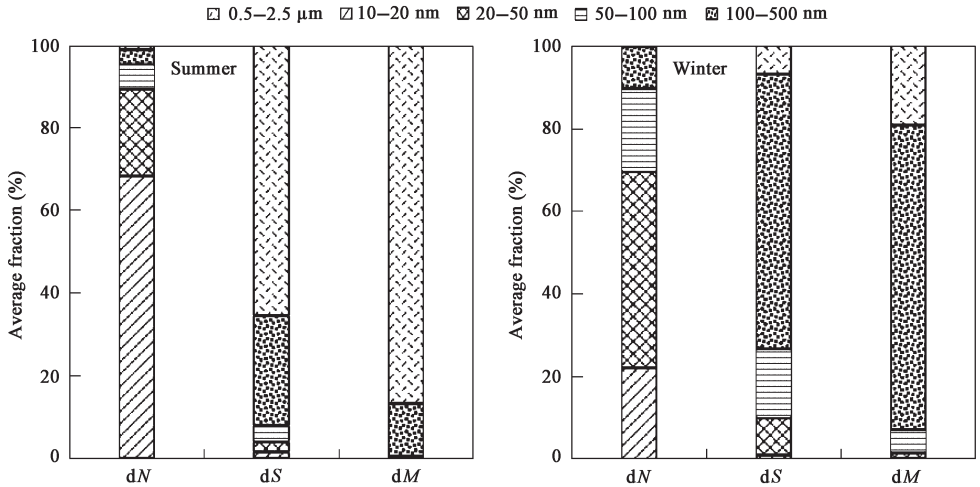

Fig. 2 Average fractions of the particle number, surface and mass concentrations in each of these sub size ranges in summer and winter. d*N*: diurnal number concentration; d*S*: diurnal surface concentration; d*M*: diurnal mass concentration.

89.4% in winter). It has been well known that most of the ambient particles in urban atmospheres are in the ultrafine size range (< 0.1 μm) (Peters *et al*., 1997; Woo *et al*., 2001). While, for the percentage associated with particles between 10 and 100 nm to total particle count, the values in this study are much higher than those currently reported, such as by Tuch *et al*., (1997) for Eastern Germany (72%) and Woo (2003) for Atlanta (61%). Note that the high fractional value of particles <100 nm in this study with the consideration of the fact that the lowest detection limit was 10 nm, supplying high opportunity for nucleation and new particle formation and this will likely promote severe aerosol pollution in the surrounding areas. Additionally, the number mean diameters (NMD) of the particles in the size range of 10–500 nm were calculated to be 21 and 36.3 nm, inferring that the size range were dominated by the particles of nucleation mode. Recently, there has been an increased interest in the relative health effects of particles of smaller sizes (Oberdorster *et al*., 1992, 1995; Donaldson and MacNee, 1998). Some laboratory studies have also shown that for a given mass concentration, health effects are larger for smaller particle sizes (Wichmann and Peters, 2000). Known that the high number concentrations and percentage of ambient ultrafine particles in Jinan atmosphere, potential jeopardy of human health caused by ultrafine particles should be given more concern. Note that the high fractional value of particles <100 nm in this study, suggesting the evidence of new particle formation and strong nucleation over the region.

2.2 Temporal variation

Figures 3a and 3b give a time series plot of the number concentration of submicron particles, together with meteorological factors (30-min average). Number concentration exhibits large temporal variability, in both of the two seasons. Especially, during the summer observation, the 30-min averaged number concentration in the particle size range of 10–200 nm varied a lot with occasional peaks as high as 10500 cm^{-3} that exceeded the mean by a factor of three to seven, implying that the site may experience very serious aerosol pollution. High particle counts were observed during 13–18 May, with the high temperature, strong solar radiation and low relative humidity. On the day 12, 20, 21 May, particle counts in 10–200 nm kept low values while those in 200–2500 nm were high. What should be noticed are the lower mixing height and the higher mixing ratio of trace gases (NO_x and CO) and mass concentration of PM_{10} (not shown), which inferred that the emission of primary pollutants under certain meteorological conditions may be the major cause of the high concentration of larger particles. While for the observation in winter, particle counts in both 10–200 nm and 200–2500 nm showed strong consistency with the mixing ratio of trace gases. And the particle number concentration in the series of modes kept high values when the site experienced poor visibility.

The relationships of the total sub-2.5 μm number concentration (N) and the surface concentration (S) were also calculated but there is no apparent correlation between them (shown in Figs.4a and 4c), neither was the relationship between the number concentration and the $PM_{2.5}$ mass concentration (M) (Figs. 4b and 4d). It accords with the results observed during Pittsburg Air Quality Study (PAQS) (Stanier *et al*., 2004). Comparatively, the two set of correlation factors in winter ($R_{dC_N \text{ vs } dC_S} = 0.68$, $R_{dC_N \text{ vs } dC_M} = 0.54$) is larger than those in summer ($R_{C_N \text{ vs } C_S} = 0.17$, $R_{C_N \text{ vs } C_M} = -0.14$). And this is thought to be caused by the number, surface and mass distribution in the different seasons, that is, the pronounced mode of particles is larger in winter than summer. Although the relationship is rather poor in summer time as above mentioned, data in certain hours show relative strong correlation with low value of particle number concentration. Here we divided the sampling hours when particle counts in the mode of 1.0–2.5 μm mode took higher percentage of the number of all urban air particles (> 0.35%). Interestingly, the data set showed better correlation which were shown in Figs.4a and 4b in black dots ($R_{dC_N \text{ vs } dC_S} = 0.65$, $R_{dC_N \text{ vs } dC_M} = 0.57$). The results reveal that the relationship of C_N vs C_S or C_N vs C_M are strongly influenced by percentages of particles in different size, that is PM mass or surface concentration area is a poor surrogate for number concentration in health effects studies or ambient visibility. Knowing that the ultrafine particle mainly responsible for ambient visibility and health effects, the strategies designed to meet national ambient air need to be improved. Quality standard based on mass concentration of PM_{10} or $PM_{2.5}$ might.

2.3 Diurnal variation

Previous studies suggested that diurnal patterns of particle number vary between two extremes: (1) influenced predominantly by meteorology for sites without local particle sources or nucleation; and (2) strongly influenced by local sources such as traffic and nucleation (Kim *et al*., 2002; Morawska *et al*., 2002). In this section we divided the particles into 4 modes based on the particle diameter, 10–20 nm (mode A), 20–50 nm (mode B) and 50–100 nm (mode C), 100–1000 nm (mode D), representing the so called nucleation mode, Aitken nuclei and accumulation mode, to study the diurnal profiles of fine particles. The concentration of SO_2, CO, NO_x, PM_{10} mass concentration are also studied to get further insights into mechanisms.

2.3.1 Summer

Figure 5a displays the diurnal pattern of the sub size bins from 10–1000 nm during summer observation. Number concentration of particles smaller than 200 nm showed the similar profile. Number concentration began to rise after the sunrise (06:00 local time (LT)) and kept climbing and got the peak values of the day at noontime (around 11:00 LT) (24612 cm^3 in 10–20 nm, 7638 cm^3 in 20–50 nm, 2160 cm^3 in 50–100 nm, 736 cm^3 in 100–200 nm). Temperature and intensity of solar radiation also started to increase and the relative humidity showed reverse pattern from the diurnal variation of meteorological factors (Fig.5b). Additionally, the mixing ratio of SO_2 has a consistent profile with particle counts, which got the major peak at 11:00 LT. As neither OH nor H_2SO_4 (g) was measured

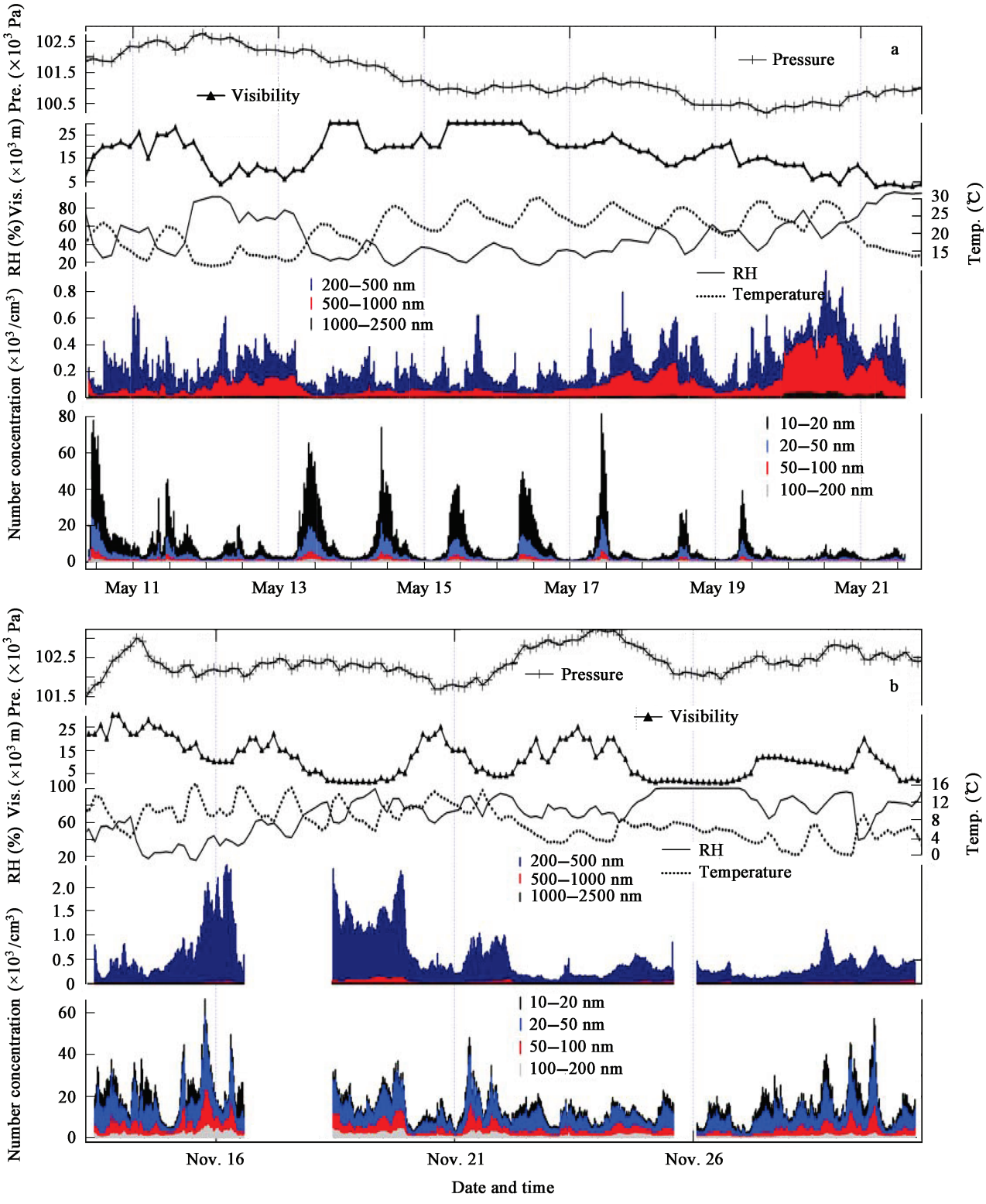

Fig. 3 Time series of number concentrations in different sub size bins together with meteorological factors. (a) summer; (b) winter.

during the period, the product of ultraviolet light and SO_2 (UV*SO_2) was used as a surrogate parameter for H_2SO_4 production (not shown). The UV*SO_2 showed the same pattern with particles of the two modes. The above information may suggest the homogeneous nucleation of sulfuric acid-water under the strong photochemical process and then the nano-particles grew to detectable new particles (>10 nm) by condensation of sulfuric acid or by self coagulation. Therefore, the high ambient temperature, low relative humidity and strong photochemical activity were supposed to supply a favorable environment of secondary particle formation and growth and it may be the most important contributor for the noontime peaks of the modes.

Interestingly, seen from Fig.5a, the diurnal variations of the particles in the size range of 200–500 nm were found to be a triple-peak pattern during a day. The number

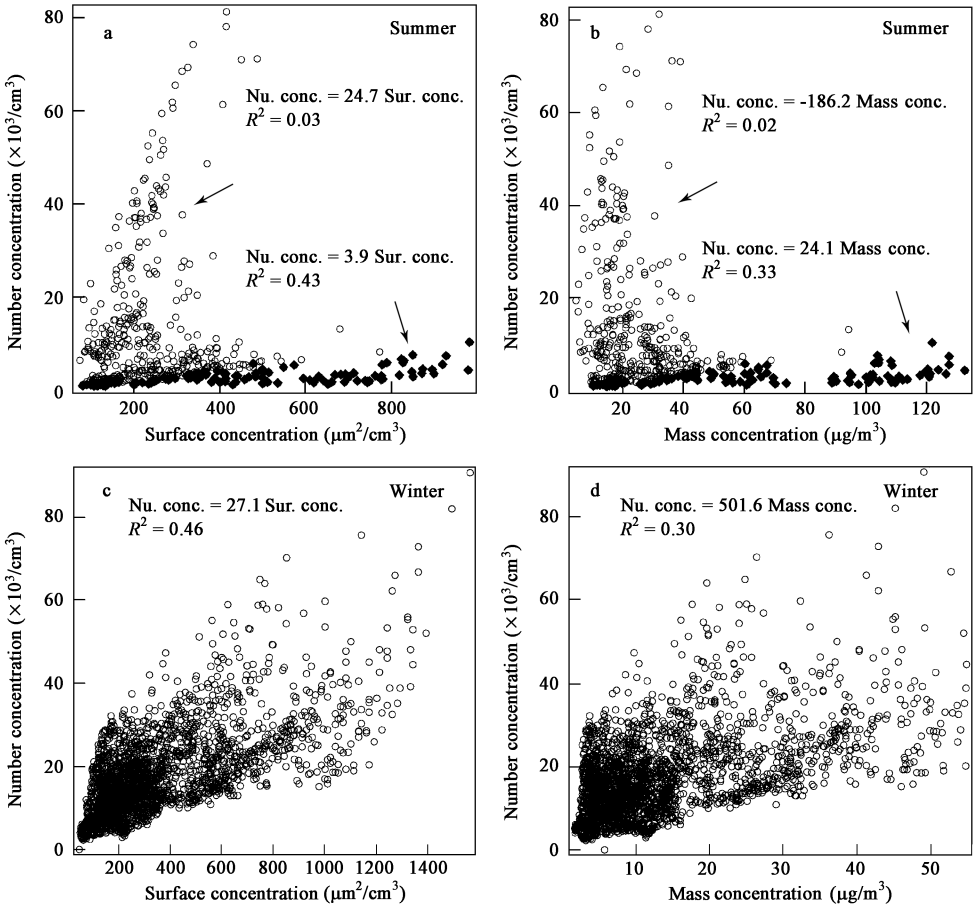

Fig. 4 Relationships of the total sub-2.5 μm particle number concentration (N) with the $PM_{2.5}$ (calculated with WPS data) mass concentration (M) and the surface concentration (S). (a) N vs S in summer; (b) N vs M in summer; (c) N vs S in winter; (d) N vs M in winter.

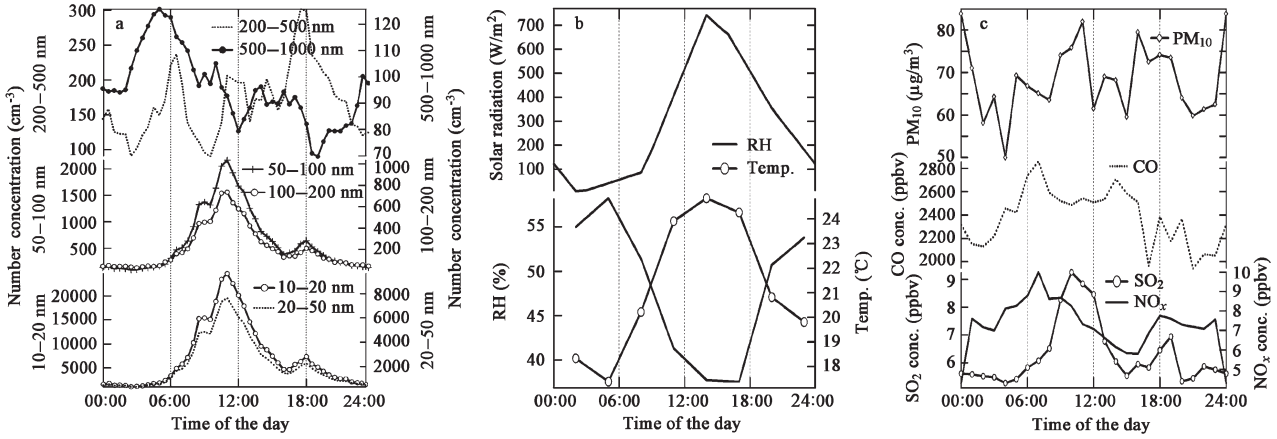

Fig. 5 Diurnal variation in summer (a) the number concentration, (b) the meteorological factors, and (c) the trace gases concentration (SO_2, NO_x, CO) and PM_{10} mass concentration.

concentration began to climb up since the sun raise and got the first peak at about 07:00 LT (239 cm^{-3}), with that the number concentration of the two modes got their evening peaks almost simultaneous around 18:00 LT (302 cm^{-3}), both of the two peaks appeared at the hours when traffic rush hour is expected. Therefore the dust during rush hours may be the possible contributor for these particles. The little peak appeared at noon hours may be also caused by the same reason or by growth of ultrafine particles under strong photochemical promotion. Despite the effect of dry deposition, it is also suggested that the pre-existing larger particles will depress the formation of new particle due to their coagulation effect on surface, and this may be the key reason which was responsible for the decrease of ultrafine particles after noon time.

2.3.2 Winter

Diurnal variations of ultrafine particles observed in winter were found to be much different from those in summer. Fig.6a indicates the diurnal variation of the particle

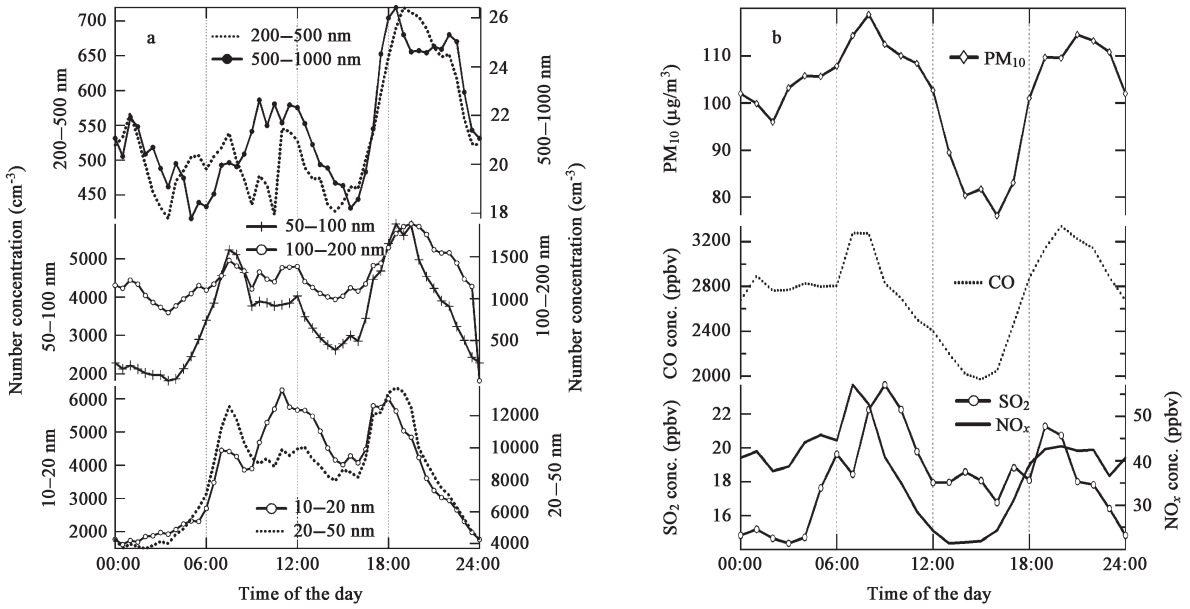

Fig. 6 Diurnal variation in winter (a) the number concentration, (b) the trace gases concentration (SO_2, NO_x, CO) and PM_{10} mass concentration.

counts and Fig.6b gives the diurnal pattern of the relative pollutants. What should be noticed is that the particle counts in the 4 modes smaller than 200 nm all showed three peaks profile. Number concentration in the size range of 10–20 nm got the major peak value of the day at about 11:00 LT (6260 cm^{-3}) with two shoulder peaks at 08:00 LT (4449 cm^{-3}) and 18:00 LT (5998 cm^{-3}), respectively. Note that the noontime peak appeared about 2 h later than the morning peak of SO_2, the high number concentration of this mode may mainly promoted by nucleation related with high mixing ratio of SO_2 and H_2SO_4. Compared with the diurnal variation of the same mode in summer, the two shoulder peaks at the jam hours strongly suggested the significant contribution from traffic emission.

For the number concentration in the size range of 20–200 nm, the peak values occurred at traffic hours (07:30 LT: 12578 cm^{-3} in 20–50 nm, 5230 cm^{-3} in 50–100 nm and 1456 cm^{-3} in 100–200 nm; 18:30 LT: 13757 cm^{-3} in 20–50 nm, 5911 cm^{-3} in 50–100 nm and 1861 cm^{-3} in 100–200 nm) are much pronounced than what appeared at noon hours (12:00 LT: 9925 cm^{-3} in 20–50 nm, 4026 cm^{-3} in 50–100 nm and 1385 cm^{-3} in 100–200 nm). The information strongly inferred an absolute different source or formation mechanism for ultrafine particles in winter from in summer, especially for the particles smaller than 200 nm. In winter, the cooler weather, more pre-existed larger particles and lower intensity of solar radiation restrained the nucleation and formation of new particles. While the particles in corresponding diameter emitted from diesel or petrolic engine may be the dominate contributor because of the lacking dilution caused by weaker convection.

Similar with what has been observed in summer, the particle counts in the size range of 200–500 nm also showed trip-peak profile during the day, in which the morning and evening peaks were highly correlated with the peaks of the traffic related trace gases (CO and NO_x). These distribution structures may supply significant indication on the characteristic of air masses influenced strongly by traffic emission. Still, it is rather interesting to know why the maximum value got in the afternoon, because the mixing height is high during that period, diluting the emissions.

3 Conclusions

Particle number concentration and size distribution were measured in Jinan urban site during the summer and winter months in 2006, together with some other parameters including SO_2, NO_x, CO, mass concentration of PM_{10}. Both the number concentration and size distribution showed strong seasonal variation in this study. For the particles in the size range of 10–500 nm, number concentration was 10685 cm^{-3} in summer and 17387 cm^{-3} in winter, inferring that the Jinan City has experienced relative serious pollution of ultrafine particles. The number mean diameter of fine particles was examined in the two seasons. The particle size of 21 nm was found to be the dominate mode during the summer observation, suggesting the major source of the ultrafine particle may caused by nucleation. While the mode of 36.3 nm in winter maybe the result of strong vehicular or burning emissions during the heating period.

Diurnal variation of particle counts in different diameter ranges have also been considered to identify the source or formation mechanism of the fine particles. The number concentration of particles smaller than 200 nm were found to get their diurnal peak at noontime, having a good correlation with the temperature and intensity of solar radiation and about one hour after the peak value of SO_2. The information suggested the nucleation and new particle formation promoted by photochemical process maybe the key drive of the ultrafine particles. However, the influence of direct emission can not be ignored because the counts of larger particles showed peak values during the rush hours. During winter observation, the particles in size range of 10–20 nm was thought to be influenced by the

comprehensive effect of nucleation and emission from traffic. While the obvious double-peak of those larger than 20 nm (20–50 nm, 50–100 nm 100–200 nm and 200–500 nm) can be identified to be caused by traffic emission. And the distinctness of property between two seasons was supposed to be the result of different weather condition and characteristic of boundary layer.

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