

Aerosol size distributions in urban Jinan: Seasonal characteristics and variations between weekdays and weekends in a heavily polluted atmosphere

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Abstract Aerosol size distributions, trace gas, and PM_{2.5} concentrations have been measured in urban Jinan, China, over 6 months in 2007 and 2008, covering spring, summer, fall, and winter time periods. Number concentrations of particles (10–2,500 nm) were 16,200, 13,900, 11,200, and 21,600 cm⁻³ in spring, summer, fall, and winter, respectively. Compared with other urban studies, Jinan has higher number concentrations of accumulation-mode particles (100–500 nm) and particles (10–2,500 nm), but lower concentrations of ultrafine particles (10–100 nm). The number, surface and volume concentrations, and size distributions of particles showed obvious seasonal variation and are also influenced by traffic emissions. Through correlation analysis, traffic emissions are proposed to be a more important contributor to Atkien-mode and accumulation-mode particles than coal firing. Around midday, the presence of nanoparticles and new particle formation is limited to pre-existing particles from traffic emissions and the mass transport of particles from suburban

and rural areas. Compared with other studies in urban areas of Europe and the USA, the variation of particle number concentration and related gas concentration in Jinan between weekdays and weekends is smaller and the reasons has been deduced.

Keywords Aerosol · Particle number concentration · Seasonal variation · Diurnal variation · Weekday · Weekend

Introduction

Atmospheric aerosols can modify the radiation budget of a geographic area directly by scattering and absorbing solar radiation or by reflecting it back into space (IPCC 2001; Engler et al. 2007). Aerosols can also have an indirect effect on radiation budget by influencing the properties and occurrence of clouds (Stott et al. 2000; Menon et al. 2002; Bellouin et al. 2005; Chung et al. 2005). Aerosol particles can be carriers and catalysts of atmospheric chemical reactions and are toxic to health, especially ultrafine particles ($D < 100$ nm) due to their large surface area-to-volume ratio and ability to penetrate deeper into the respiratory tract (Oberdörster 2000; Nemmar et al. 2003). Aerosol particles can also degrade atmospheric visibility which can influence human activities (Dockery and Pope 1994).

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Not only is particle concentration of atmospheric aerosols important but many researchers have indicated that the size distribution of particles is an important parameter. Therefore, there has been considerable interest in the size distribution of atmospheric particles and mechanism of formation and growth of particles (Holmes 2007). These parameters and phenomena have been studied in many different atmospheric environments such as coastal, polar, remote boreal forest, and continental rural and urban areas (Wu et al. 2007). Kulmala et al. (2004a) and Holmes (2007) have reviewed many measurements taken around the world and pointed out that different characteristics of size distributions and mechanisms for new particle formation were observed in different areas.

Within urban areas, particle concentrations and size distributions are strongly influenced by traffic emissions (Hussein et al. 2004) and two mechanisms for new particle formation exist (Holmes 2007). The first mechanism involves the formation of particles either inside or within the first meter of exiting the exhaust pipe (Morawska et al. 1998). The second process appears to be associated with nucleation, probably arising from SO₂ oxidation, and subsequent condensational growth of particles (Holmes 2007). In recent years, many studies have been done on urban aerosol number size distributions, including long-term measurements. However, most of these projects were conducted in developed countries, and only a few studies have been carried out in the heavily polluted megacities of newly industrialized countries. The characteristics of particle size distributions in these megacities are usually different from those in cities in developed countries, because of larger populations, fast industrialization, and rapid urbanization (Yue et al. 2009).

Shandong Province, P. R. China, with an area of 150,000 km², is located in one of the most heavily air polluted areas in the world from gases (<http://www.temis.nl/airpollution/no2.html>; <http://sacs.aeronomie.be/archive/month.php>) and aerosols (http://ladsweb.nascom.nasa.gov/browse_images/l3_browser.html) arising from coal and vehicle usage without extensive emission controls and is the largest emitter of SO₂ in China (<http://www.stats.gov.cn/tjsj/ndsj/2009/indexeh.htm>). SO₂

emission in Shandong Province totaled more than 1.692 million tons in 2008 (<http://www.stats.gov.cn/tjsj/ndsj/2009/indexeh.htm>). The city of Jinan, in the center of the province, is the capital of Shandong Province, and has an urban population of about 3.6 million and a total population of more than six million in 2009 (<http://www.jinan.gov.cn/col/col12/index.html>). Like many cities in the northern area of China, Jinan is suffering serious air pollution from aerosol particles and SO₂ (Yang et al. 2007; <http://www.stats.gov.cn/tjsj/ndsj/2009/indexch.htm>). Yang et al. 2007 showed that annual mean mass concentration of PM_{2.5} of urban Jinan was 138 µg·m⁻³, about 14 times of WHO Air Quality Guidelines (10 µg·m⁻³; WHO 2005). For a better understanding of aerosol size distributions of urban Jinan and characteristics of aerosol size distributions under serious air pollution, continuous measurements of particle number concentrations in the range of 10–2,500 nm are reported for all four seasons in urban Jinan, under heavily polluted conditions. Additionally, mass concentration of PM_{2.5}, SO₂, NO_x, and O₃ were measured simultaneously. The concentrations and size distributions of number, surface, and volume of particles were investigated. The seasonal, diurnal variations of particle number concentrations, and other related parameters are discussed. Additionally, variations between weekdays and weekends and correlations of several main parameters have been analyzed.

Methodology

Site description

The sampling site was on the roof of public teaching building in Hongjialou Campus of Shandong University located in the north-eastern part of urban Jinan (Fig. 1, 36°40' N, 116°57' E), about 20 m above ground level. There are no obvious industrial pollutant sources near the sampling site because all the districts nearby are residential or commercial. The sampling site is affected by two major roads with heavy traffic: east of the site is East Second Ring Road with a distance of about 240 m and south of the site is Huayuan Road with a distance of about 300 m.

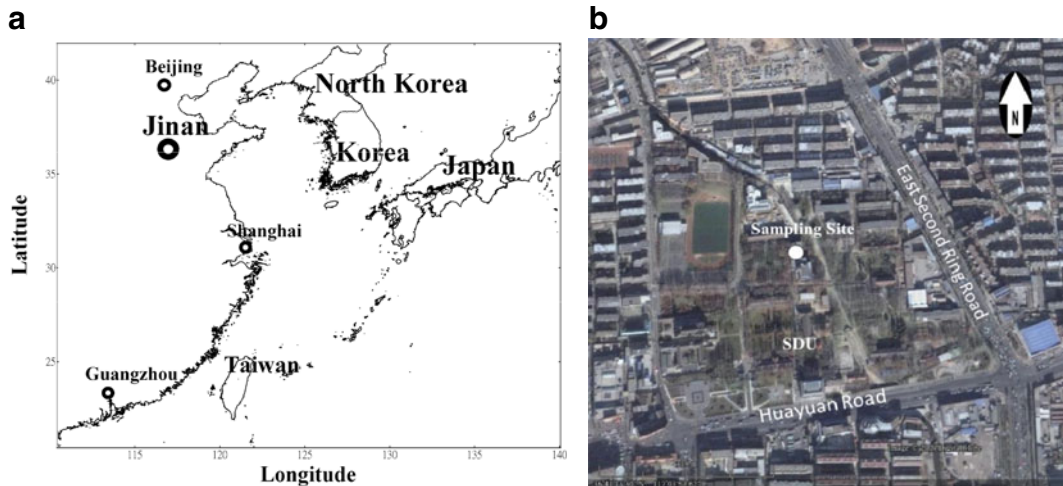


Fig. 1 Location of the atmospheric measurement site. **a** Location of Jinan. **b** Sampling site (white dot in the image from Google Earth™)

Instrumentation

A wide-range Particle Spectrometer™ (WPS-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™ 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 less than 10,000 V, using a sheath flow rate of 3 L·min⁻¹. The CPC portion of the instrument 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 using 24 additional channels. Particles are drawn into the aerosol inlet at a flow rate of 0.70 L·min⁻¹ and focused with a 3 L·min⁻¹ 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. The LPS was calibrated with four NIST traceable sizes of PSL (0.701, 1.36, 1.6, and 4.0 μm mean diameter). Each sample takes 8 min 16 s to be analyzed. Over the course of all measurements, the WPS experienced no malfunctions.

The mass concentration of PM_{2.5} was sampled using Reference Ambient Air Samplers (RAAS) PM_{2.5} samplers (TEI RAAS 2.5–400), and using Teflon filter weighing using a Sartorius ME5-F balance (0.001 mg). Ozone was monitored using a UV photometry analyzer (TEI model 49C), SO₂ by a pulsed UV fluorescence analyzer (TEI model 43C), and NO_x by a MoO/chemiluminescence analyzer (TEI Model 42i-TL). The TEI model 49C was fully calibrated using a TEI Model 49C Primary Standard UV photometric O₃ calibrator every month. The TEI Model 43C and TEI Model 42i-TL were fully calibrated each week using a TEI Model 111 zero air supply and a TEI Model 146C calibrator with mixed standard gas consisting of NO, SO₂, and CO. The TEI Model 42i-TL, 43C, and 49C were simultaneously zero checked every 3 to 4 days over the course of the measurements.

Meteorological data were obtained from an automatic meteorological station, located 10 m northeast of the sampling site. In spring, southwest wind is most frequent and northeast wind

takes the second, and it is on the contrary in winter. In both summer and fall, northeast wind dominates and southwest wind takes the second but is far less frequent the northeast wind.

Sampling duration and data treatment

As the larger instrumental error originates from too small concentration values of particles with diameters larger than 2.5 μm and the particle number concentrations are dominated by ultrafine particles (Seinfeld and Pandis 1998), only particle data not larger than 2.5 μm was analyzed. Particles in the diameter from 10 nm to 2.5 μm were divided into six sub-ranges—10–20 nm (N_{10-20} or N_{nuc} , nuclei mode), 20–50 and 50–100 nm (N_{20-50} , N_{50-100} , Atkien mode), 100–200, 200–500, and 500–2,500 nm ($N_{100-200}$, $N_{200-500}$, $N_{500-2,500}$, accumulation mode).

In order to analyze seasonal differences, the available dataset was subdivided according to four seasons: spring (03/26/2008–05/03/2008), summer (05/31/2008–06/26/2008), fall (10/02/2008–10/26/2008), and winter (12/03/2007–01/07/2008, 02/02/2008–03/25/2008). Considering that 03/25/2008 was the end of the heating season (residential steam heat production ceases) in Jinan, we regarded the whole heating season as winter. After data screening, there are 7,308, 3,377, 3,763, and 12,014 WPS samples for spring, summer, fall, and winter, respectively.

Results and discussion

Number concentration and seasonal variety

Various descriptive statistics regarding the measured hour average number concentrations of the four seasonal periods are included in Table 1. The

particle number concentration with the diameter between 10 and 2,500 nm ($N_{10-2,500}$) in winter was the largest of the four seasons, while N_{20-50} , N_{50-100} , $N_{100-200}$, and $N_{200-500}$ had the same seasonal variation. The N_{20-200} range accounted for most of the total particle number concentration in Jinan, so its variety dominated the seasonal variety of the total number concentration. Differing from the larger diameter particles, N_{10-20} in fall was far larger than other seasons indicating more frequent new particle formation events occurring in fall; a detailed explanation has been presented in “[Diurnal variation](#)”. The $N_{200-500}$ value was the largest in winter and second largest in summer. In fall, N_{20-50} , N_{50-100} , $N_{100-200}$, $N_{200-500}$, and $N_{500-2,500}$ were the lowest and are attributed to good atmospheric dispersion.

Similar with Jinan (I have analyzed the particle sources of Jinan in “[Number, surface, and volume concentration size distribution](#)”), in Pittsburgh, a USA city, nucleation and vehicle emissions were the most important sources of particles (Stanier et al. 2004a, b; Zhang et al. 2004). Compared with the study on urban Pittsburgh (Table 2; Stanier et al. 2004a), urban Jinan possesses a lower number concentration of particles smaller than 20 nm (nucleation mode) and 50 nm but has a higher number concentration of accumulation mode particles and those between 50 and 100 nm. The mass concentrations of $\text{PM}_{2.5}$ are generally dominated by particles of accumulation mode, and higher number concentrations of accumulation mode indicated higher mass concentrations of $\text{PM}_{2.5}$ in urban Jinan. During the experimental period in Pittsburgh, the mass concentrations of $\text{PM}_{2.5}$ were 20 $\mu\text{g}\cdot\text{m}^{-3}$ in summer and 12 $\mu\text{g}\cdot\text{m}^{-3}$ in winter, compared to 173.2 and 159.6 $\mu\text{g}\cdot\text{m}^{-3}$ in summer and winter for Jinan, respectively. Otherwise, particle surface concentrations are dominated by the accumulation mode as well, so higher particle

Table 1 Descriptive statistics of the measured particle number concentrations ($\#\cdot\text{cm}^{-3}$) of the four seasons (diameter unit: nm)

Season	10–20	20–50	50–100	100–200	200–500	500–2,500	10–2,500
Spring	500	6,500	4,600	3,300	1,200	25	16,200
Summer	900	5,500	3,300	2,900	1,200	77	13,900
Fall	2,200	4,800	2,100	1,500	600	22	11,200
Winter	900	7,700	6,600	4,700	1,700	29	21,600

Table 2 Comparison of descriptive statistics of sub-ranges for Jinan and Pittsburgh, annual (Unit: #·cm⁻³)

Particle size range (nm)	10–20	20–50	50–100	100–200	200–500	500–1,000	1,000–2,500
Pittsburgh, USA	4,100	6,500	3,600	1,710	460	18	0.59
Jinan, China	1,100	6,100	4,100	3,100	1,200	40	6.7

number concentration of accumulation mode in urban Jinan indicates higher particle surface concentration. New particle formation and the existence of newly formed nanoparticles are strongly limited by surface concentrations of pre-existing aerosol particles (Kulmala et al. 2001, 2004a, b) So that may be an important reason for why lower nucleation mode particle number concentrations were observed in Jinan, under a high SO₂ concentration (annual mean 26 ppb).

For Tables 2 and 3, data were acquired using different types of instruments.

Compared with other studies in urban areas (Table 3), the value of *N*_{10–100} in Jinan was observed to be the least of six cities listed, while the value of *N*_{100–500} was the highest, as was the mass concentrations of PM_{2.5}.

Number, surface, and volume concentration size distribution

Mean particle number, surface, and volume size distributions of the four seasons in Jinan are shown in Fig. 2; the surface and mass concentrations were calculated with the assumption that the particles are spherical. As can be seen in Fig. 2, there is obvious seasonal variation and the number concentrations are mainly determined by Atkien mode particles. The number distribu-

tion in winter shows a single peak around 49 nm dominated by Atkien mode particles; the number distribution in spring is similar to winter but has a slightly lower peak maximum (41 nm). A bimodal log-normal (Atkien mode and accumulation mode) distribution in summer and fall are also shown in Fig. 2a, and in fall, higher value for nucleation mode is observed. The differences of number size distribution in various seasons are determined by factors such as meteorology, the sources and sinks of particles, etc., so further information from particle size distributions cannot be determined. However, the fact that new particle formation events occur more frequently in fall than in other seasons in Jinan can be deduced.

Surface and volume (mass) size distributions (Fig. 2b, c) are mainly determined by the accumulation mode particles. Measurements in the fall not only showed the highest nucleation mode particle number concentration but also had the lowest surface concentration. Single peaks with inconspicuous secondary peaks were observed in spring, fall, and winter. The main peaks of surface and volume (mass) size distributions are located around 300 and 400 nm, respectively. In contrast, there is a conspicuous peak located near 1,095 nm for both surface and volume (mass) size distributions in summer. Venkataraman and Rao (2001) have observed biomass burning can produce a

Table 3 Comparison of descriptive statistics of sub size ranges for Jinan and other urban studies

Location		Number conc. (#·cm ⁻³)		PM _{2.5} (µg·m ⁻³)	Source
		10–100 nm	100–500 nm		
Alkmaar, Netherlands	Winter	18,300	2,120	27.0	Ruuskanen et al. (2001)
Erfurt, Germany	Winter	17,700	2,270	41.9	Ruuskanen et al. (2001)
Helsinki, Finland	Winter	16,200	973	9.42	Ruuskanen et al. (2001)
Pittsburgh, USA	Annual	14,300	2,170	16	Stanier et al. (2004a)
Atlanta, USA	Annual	21,400		19.3	Woo et al. (2001)
Jinan, China	Spring	11,700	4,500	102.7	This work
Jinan, China	Summer	9,700	4,100	159.2	This work
Jinan, China	Fall	9,100	2,100	99.3	This work
Jinan, China	Winter	15,200	6,400	173.2	This work
Jinan, China	Annual	11,400	4,300	133.6	This work

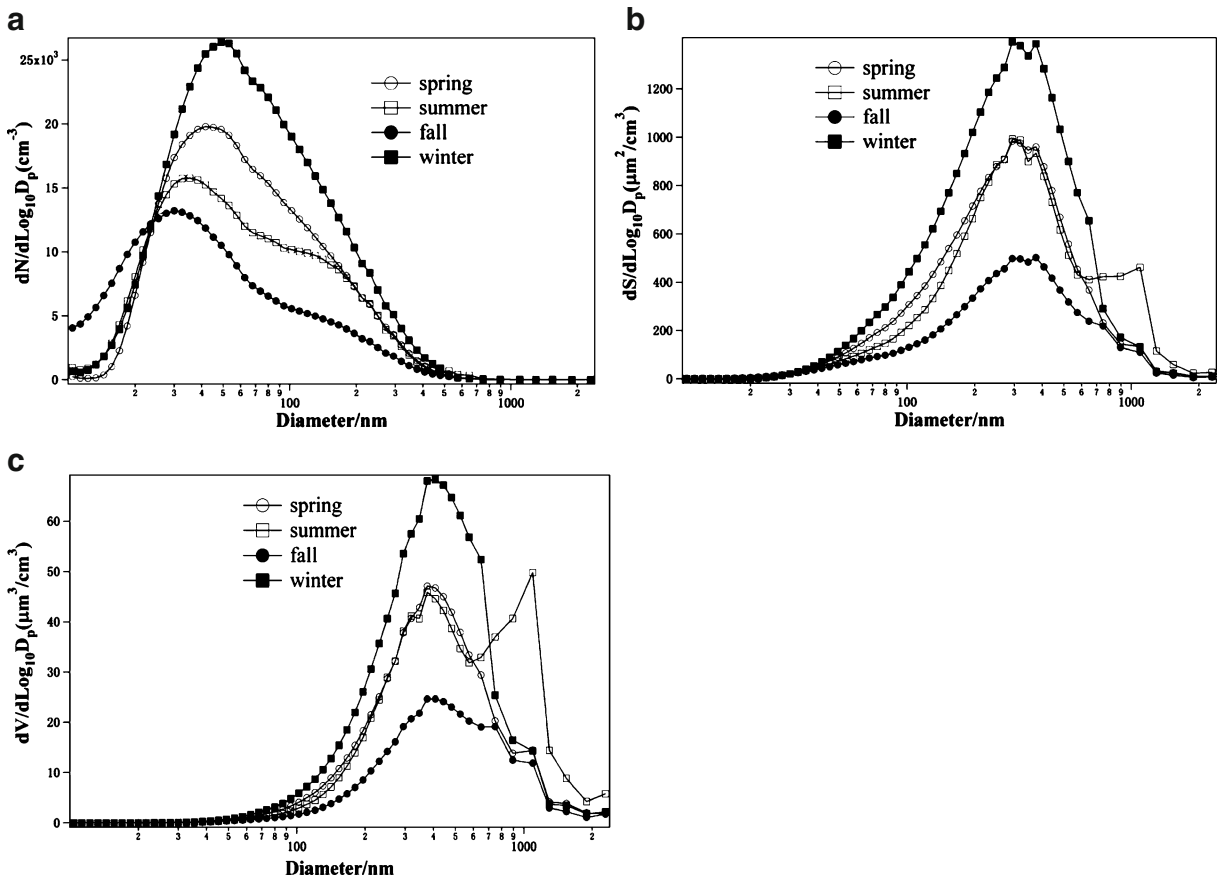


Fig. 2 Mean number (a), surface (b), and volume (mass) (c) size distributions in four seasons, Jinan

particle mass size distribution with a peak near 1 μm , and our summer measurement was operated during June, a frequent biomass burning period in the northern China (Fig. 3a), so the conspicuous peak located near 1,095 nm for both surface and volume (mass) size distributions in summer may have originated from agricultural biomass burning.

Diurnal variation

Figure 3 illustrates the diurnal variations of particle size distribution between 10 and 500 nm with diameter and geometric mean diameter (GMD) of particles (10–2,500 nm) on the y-axis, time of the day on the x-axis, and particle concentration ($dN/d\text{Log}_{10}D_p$) by shading contour for the various seasons. The simultaneous diurnal variations of

the concentrations of NO_x , SO_2 , and O_3 , as well as aerosol surface area are shown in Fig. 4.

As seen in Fig. 4, in spring and winter, a high concentration of N_{20-200} was observed during day and night, indicating the accumulation of particles from traffic emissions under a poor atmospheric dispersion capacity and a high concentration of background particles in the Jinan urban area. In summer and fall, that the N_{20-200} was lower indicates a good dispersion capacity, especially in fall. Considering the diurnal of NO_x concentration, for all seasons (cf. Fig. 5), the contour area plots can be divided into three regions of high particle concentration: morning rush hour, evening rush hour and daytime around noon, with particles mainly arising from the accumulation mode. Compared with morning rush hour, the high particle number concentrations in evening rush hour lasted for longer time due to the poor dispersion capacity

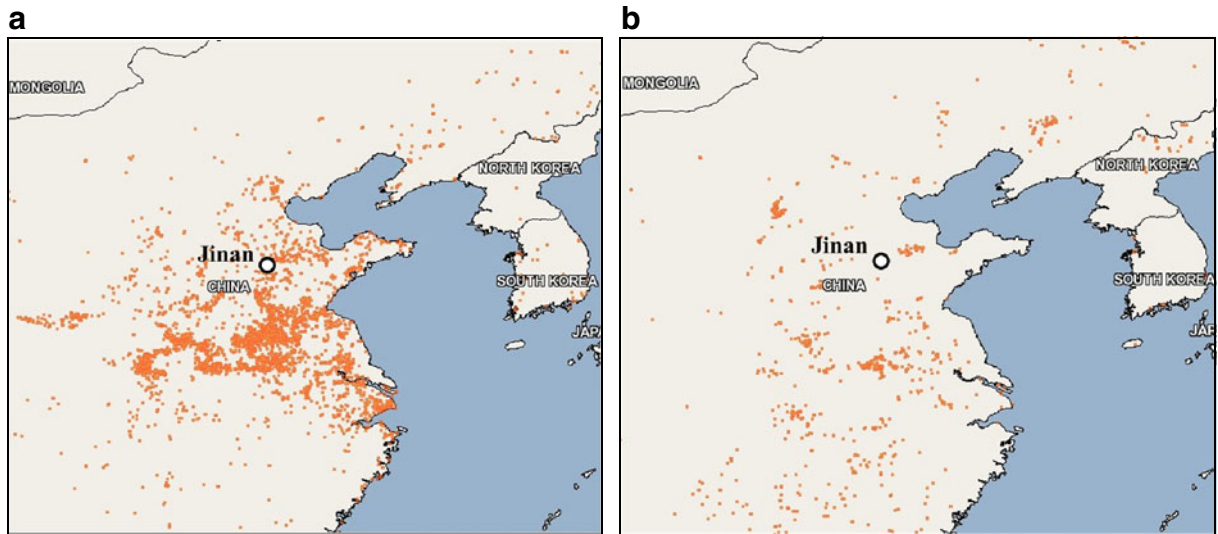


Fig. 3 MODIS images of fire spots, **a** summer (05/31/2008–06/26/2008), **b** fall (10/02/2008–10/26/2008), (Aqua & Terra, <http://firefly.geog.umd.edu/firemap/>)

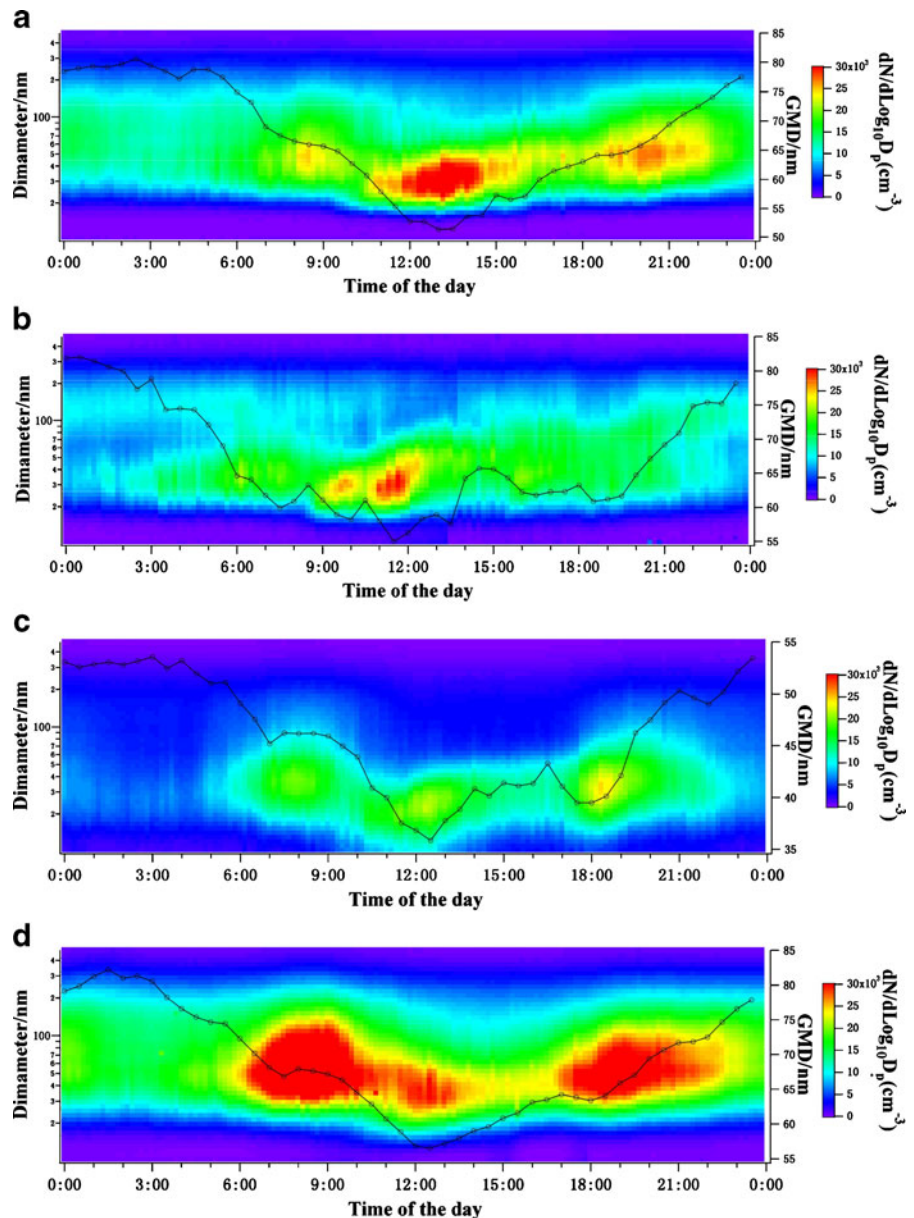
at night. In winter, with a temperature inversion layer, lower wind speed, and a lower altitude of the atmospheric mixing layer, it is obvious that particle number concentrations are influenced by traffic emissions in rush hour.

An event of new particle formation was identified if there was a spontaneous burst of particle number concentrations in the nucleation mode (10–20 nm, the smallest channel of the WPS system) followed by their subsequent growth at the rate of a few nanometers per hour over a time span of hours. This event must also be, without obvious local emissions affecting the number concentrations of nucleation-mode particles (Gao et al. 2008). In this work, this criterion was used to identify the overall diurnal frequency of new particle formation in various seasons. In Fig. 5, although peaks of SO₂ concentration (indicating high concentrations of precursors for new particle formation and growth) and O₃ (indicating strong photochemical oxidation to a certain extent) occurred simultaneously in spring, summer, and fall, no obvious new particle formation was observed in spring, summer, and winter. Figure 4c shows that more frequent new particle formation events occurred in fall. New particle formation and the existence of newly formed nanoparticles are strongly limited by surface concentrations of

pre-existing aerosol particles (Kulmala et al. 2001, 2004a, b). As can be seen in Fig. 5, compared with other seasons, fall has the lowest particle surface concentration at noon, so it may be concluded that high number concentrations of particles in spring, summer and winter limit new particle formation in urban Jinan.

The diurnal variations of surface concentration were highly accordant with that of NO_x concentration (Fig. 5), especially in the morning and evening rush hours. In the morning and evening, atmospheric dispersion capacity and the influence of air mass transport from the suburbs and rural areas are weaker, so it is reasonable that the surface concentration is dominated by traffic emissions. While both NO_x and surface concentrations are the lowest around noon, it cannot be confirmed that surface concentrations are also dominated by traffic emissions like the morning and evening, because both NO_x and surface concentrations may be affected strongly by atmospheric dispersion around midday. The correlation coefficients (*R*) between *N*_{nuc}, concentrations of NO_x and SO₂, and surface concentration around midday are shown in Table 4. Compared to the complete data set (Table 6 below), *R*_{surface vs NO_x} (the correlation coefficients between concentrations of particle surface area and NO_x) around midday

Fig. 4 Contour plots of diurnal variations of particle number concentration in 10–500 nm and diurnal variations of GMD (black line, 10–2,500 nm), spring (a), summer (b), fall (c), winter (d)



are lower and R_{surface} vs SO_2 around midday are higher. This suggests that surface concentrations are affected more strongly by air mass transport from suburbs and rural areas, and more weakly affected by traffic emissions. Because most new particle formation events occurred around noon, it can be concluded that surface concentrations of pre-existing particles from traffic emissions and transporting from suburban and rural areas limit new particle formation in urban Jinan.

These data suggest that in the Jinan urban area, traffic emissions are important sources of particle number and surface concentrations. However, particle surface concentrations also decrease the latent concentration of ultrafine particles derived from new particle formation under the high SO_2 concentration levels in Jinan. Around midday, N_{20-60} was higher than during the morning and evening rush hours, although there is a smaller traffic flow and stronger dispersion capacity

Fig. 5 Diurnal variations of SO₂, O₃, NO_x, and surface concentrations of PM_{2.5}, spring (a), summer (b), fall (c), winter (d)

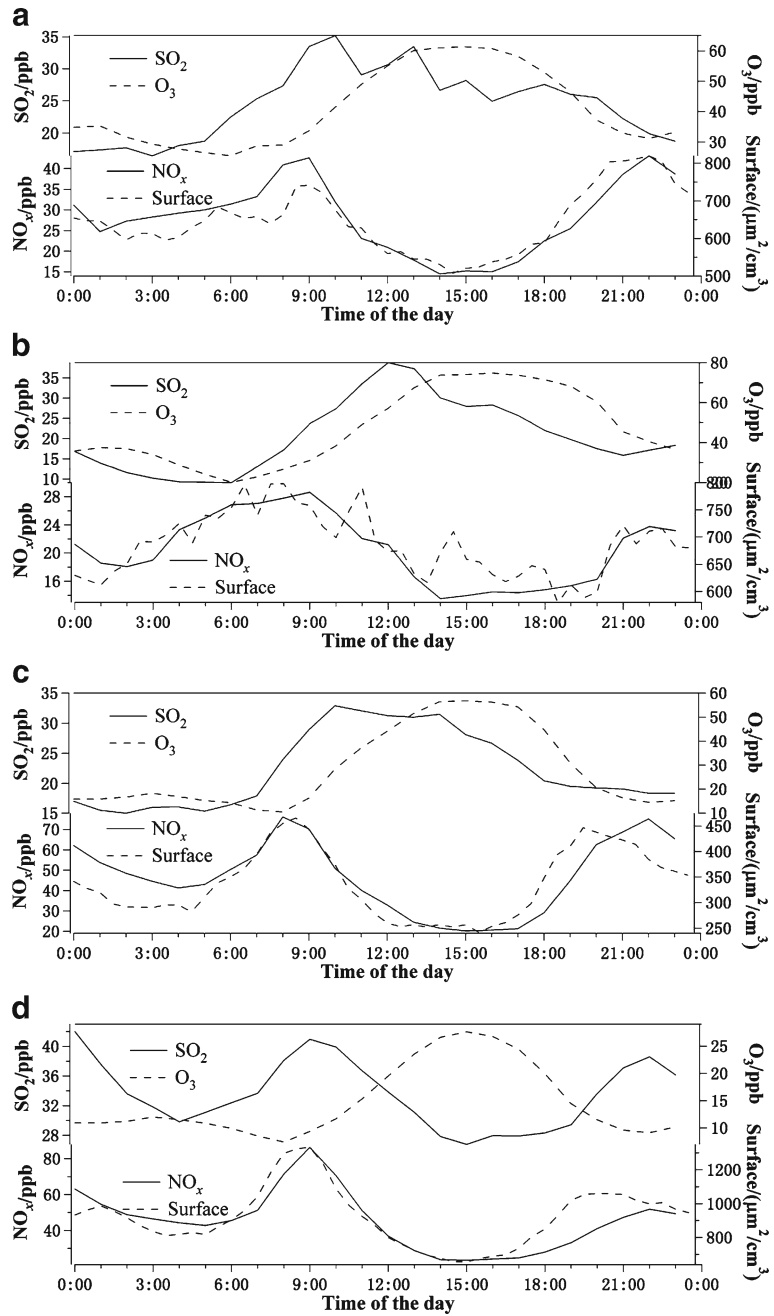


Table 4 Correlation coefficients (*R*) between hour average of *N*_{nuc}, surface concentration of PM_{2.5}, and NO_x and SO₂ concentrations

	Spring			Summer			Fall			Winter		
	Surface	NO _x	SO ₂	Surface	NO _x	SO ₂	Surface	NO _x	SO ₂	Surface	NO _x	SO ₂
<i>N</i> _{nuc}	-0.28	-0.02	0.09	-0.34	-0.01	-0.05	-0.39	-0.18	0.07	-0.24	-0.02	-0.06
Surface	1.00	0.53	0.56	1.00	0.56	0.48	1.00	0.59	0.33	1.00	0.44	0.40

Spring 10:00–17:00; summer 9:00–16:00; fall 9:00–16:00; winter 10:00–16:00. |*R*| ≥ 0.5 in bold

during midday. Also, N_{10-20} is lower at midday, indicating that N_{20-60} particles arose from other sources instead of new local particle formation. This suggests that atmospheric transport from rural and suburban areas of newly formed particles are an important particle source at noon. Although measurements in the upwind direction of urban Jinan are lacking, it may be deduced that frequent new particle formation events occur in rural or suburban areas around Jinan under the high concentration levels of SO_2 ; similar phenomena were observed in Beijing (Shi et al. 2007) and Pittsburgh (Stanier et al. 2004b). After transport to the urban of Jinan and thorough mixing with urban air, the newly formed particles gradually become larger through heterogeneous reaction or coagulate with pre-existing particles. The subsequent growth of GMD indicates good conditions for particle growth in the heavily polluted air of urban Jinan.

Variations in particle number concentration during weekdays versus weekends

Studies on the differences in particle number concentration between weekdays and weekends can provide insight on the impact of motor vehicle emissions on particle concentrations in urban air (Morawska et al. 2002). Variations of particle number concentration and other pollutant parameters between weekdays (Monday to Friday

and other workdays, except holidays) and weekends (include holidays except workdays) were also studied, in order to investigate the influence of traffic flow rates.

Several parameters divided into weekdays and weekends in the four seasons are presented in Table 5. Compared to weekdays, the spring and winter averages of N_{nuc} (N_{10-20}), N_{Ait} (N_{20-100}), and N_{acc} ($N_{100-2,500}$), as well as surface concentration of $\text{PM}_{2.5}$, mass concentration of $\text{PM}_{2.5}$, and NO_x concentrations were lower on weekends. In contrast, summer and fall, variations between weekdays and weekends, were different than winter and spring. Generally, in urban areas, concentrations of NO_x are dominated by traffic emissions, so NO_x concentrations on weekends are potentially lower than on weekdays as traffic flow rates decrease. However, in Jinan, NO_x concentrations on weekends were higher than weekdays in summer and fall; SO_2 concentrations are similarly higher. Biomass burning can increase concentrations of NO_x , SO_2 , and other pollutants (Crutzen and Andreae 1990; Scholes et al. 1996), and the summer and fall measurements taken in June and October, respectively, were influenced by agricultural biomass burning (Fig. 3a, b). It should be noted that agricultural production in China is not influenced by holidays and weekends; thus, it is expected that the sampling site was influenced by agricultural biomass burning in summer and fall, especially in the fall. For all

Table 5 Hour average particle number, surface, mass concentration of $\text{PM}_{2.5}$, SO_2 , O_3 , and NO_x concentrations in Jinan during weekdays and weekends

Seasons		N_{nuc}	N_{Ait}	N_{acc}	Surface	$\text{PM}_{2.5}$	SO_2	O_3	NO_x
Spring	Average	500	11,100	4,500	644	102.7	26.1	39.9	29.8
	Weekdays	600	11,700	4,600	653	100.1	28.1	39.8	32.1
	Weekends	500	9,900	4,200	625	110.0	22.2	40.3	25.8
Summer	Average	900	8,800	4,200	695	173.2	21	46.3	21.2
	Weekdays	1,000	8,600	4,000	655	167.1	20.0	39.2	20.7
	Weekends	800	9,200	4,800	778	186.4	23.0	58.6	22.0
Fall	Average	2,200	6,900	2,100	338	99.3	23.9	34	50.8
	Weekdays	2,200	7,300	2,300	360	95.7	19.0	36.4	48.4
	Weekends	2,200	6,100	1,900	296	104.7	30.2	30.9	53.8
Winter	Average	900	14,300	6,500	922	159.6	33.9	14.9	38.1
	Weekdays	900	14,600	6,500	924	170.7	30.8	14.3	37.5
	Weekends	800	14,100	6,300	859	125.7	33.9	14.7	36.5

N_{nuc} , N_{Ait} , and N_{acc} : Number concentrations of N_{10-20} , N_{20-100} , and $N_{100-2,500}$, unit: #·cm⁻³. Surface concentration of $\text{PM}_{2.5}$, unit: $\mu\text{m}^2\cdot\text{cm}^{-3}$. Mass concentration of $\text{PM}_{2.5}$, unit: $\mu\text{g}\cdot\text{m}^{-3}$. SO_2 , O_3 , and NO_x , unit: ppb

seasons, there was no obvious difference between N_{nuc} on weekdays and weekends. In addition, traffic emissions have been considered an important source of N_{nuc} and N_{Ait} (Morawska et al. 1998; Harris and Maricq 2001; Kittelson et al. 2004; Burtscher 2005), so it is an important reason for lower N_{nuc} and N_{Ait} on weekends, except N_{Ait} in summer.

The variation between weekdays and weekends of several parameters included in Table 5 are smaller than measurements in urban areas in developed countries (Pekkanen et al. 1997; Ruuskanen et al. 2001; Woo et al. 2001; Morawska et al. 2002; Wehner et al. 2002, Stanier et al. 2004a, b; Laakso et al. 2003; Voigtlander et al. 2006; Costabile et al. 2009). The same phenomenon was also observed in the Beijing

urban area (Shi et al. 2007). Two reasons for this characteristic feature of the particles in Jinan may be attributed to social practices and terrain, respectively. First, in China, traffic, agricultural, and industrial activities do not decrease significantly on weekends compared to weekdays. Second, Jinan is surrounded by mountains on the east, south, and west sides, so the terrain favors the accumulation of pollutants, delays atmospheric mass transfer, and attenuates the variation between weekdays and weekends.

The correlation coefficient matrix subdivided into weekdays and weekends for several parameters for the four seasons is shown in Table 6. As mentioned above, surface concentrations are dominated by N_{nuc} , so N_{acc} and surface concentration have almost the same relationship with

Table 6 Correlation coefficients between particles and gases in spring, summer, fall, and winter, hour average, subdivided into weekdays and weekends

Seasons	Weekdays						Weekends						
	N_{nuc}	N_{Ait}	N_{acc}	Surface	NO_x	SO_2	N_{nuc}	N_{Ait}	N_{acc}	Surface	NO_x	SO_2	
Spring	N_{nuc}	1.00					1.00						
	N_{Ait}	0.33	1.00				0.42	1.00					
	N_{acc}	-0.16	0.56	1.00			-0.16	0.54	1.00				
	Surface	-0.17	0.60	0.97	1.00		-0.16	0.52	0.97	1.00			
	NO_x	-0.10	0.48	0.71	0.70	1.00	-0.08	0.51	0.76	0.74	1.00		
	SO_2	0.09	0.44	0.56	0.52	0.36	1.00	0.31	0.49	0.53	0.46	0.39	1.00
	O_3	0.21	-0.24	-0.55	-0.47	-0.72	-0.20	0.24	-0.31	-0.59	-0.53	-0.81	-0.22
Summer	N_{nuc}	1.00					1.00						
	N_{Ait}	0.53	1.00				0.49	1.00					
	N_{acc}	-0.26	0.23	1.00			-0.34	0.25	1.00				
	Surface	-0.23	0.20	0.91	1.00		-0.32	0.28	0.96	1.00			
	NO_x	-0.10	0.30	0.63	0.58	1.00	-0.04	0.38	0.60	0.63	1.00		
	SO_2	0.07	0.27	0.22	0.21	0.30	1.00	0.27	0.39	0.23	0.25	0.10	1.00
	O_3	0.17	-0.01	-0.44	-0.37	-0.65	0.16	0.09	-0.18	-0.28	-0.31	-0.78	0.24
Fall	N_{nuc}	1.00					1.00						
	N_{Ait}	0.46	1.00				0.31	1.00					
	N_{acc}	-0.21	0.47	1.00			-0.14	0.73	1.00				
	Surface	-0.23	0.42	0.98	1.00		-0.14	0.69	0.97	1.00			
	NO_x	-0.09	0.49	0.67	0.60	1.00	-0.19	0.39	0.70	0.71	1.00		
	SO_2	0.21	0.18	0.15	0.09	0.14	1.00	0.22	0.23	0.37	0.40	0.09	1.00
	O_3	-0.09	-0.47	-0.33	-0.24	-0.59	0.08	0.36	-0.08	-0.24	-0.18	-0.48	0.22
Winter	N_{nuc}	1.00					1.00						
	N_{Ait}	0.25	1.00				0.16	1.00					
	N_{acc}	-0.17	0.62	1.00			-0.11	0.58	1.00				
	Surface	-0.21	0.57	0.96	1.00		-0.08	0.57	0.98	1.00			
	NO_x	0.02	0.38	0.49	0.39	1.00	-0.04	0.32	0.53	0.47	1.00		
	SO_2	-0.02	0.31	0.43	0.35	0.78	1.00	0.13	0.19	0.53	0.44	0.69	1.00
	O_3	0.37	-0.26	-0.40	-0.41	-0.17	-0.15	0.37	-0.27	-0.49	-0.48	-0.20	-0.22

$|R| \geq 0.5$ in bold

the other parameters. For all seasons, surface concentration and N_{Ait} have apparent correlations with NO_x , and have secondary correlations with SO_2 , indicating they are more influenced by traffic emissions than by coal firing. Particle surface concentrations have negative correlations with N_{nuc} . On weekends, N_{nuc} and N_{Ait} have more apparent positive correlation with SO_2 than on weekdays, indicating that more ultrafine particles may be related with coal firing. There are no obvious differences for coefficients between weekdays and weekends.

Summary and conclusion

Particle number concentration and size distribution were measured in Jinan urban site in spring, summer, fall and winter, during 2007 and 2008, along with parameters including concentration measurements of SO_2 , NO_x , and O_3 , and the mass concentration of $\text{PM}_{2.5}$. Number concentrations of particles (10–2,500 nm) were 16,200, 13,900, 11,200, and 21,600 cm^{-3} in spring, summer, fall, and winter, respectively. Compared with other urban studies, Jinan has higher particle number concentrations of accumulation-mode particles (100–500 nm), and lower concentrations of ultrafine particles (10–100 nm), because the high concentration of pre-existing particles limits new particle formation and existence of the existence of nanoparticles. All the particle number, surface, and volume (mass) concentrations and size distributions showed seasonal variations in urban Jinan.

Diurnal variations of particle number concentrations (10–500 nm) with GMD (10–2,500 nm) and concentrations of NO_x , SO_2 , and O_3 for four seasons have been studied and showed obvious seasonal variations. Infrequent new particle formation events were observed in spring, summer and winter, but not fall. The diurnal variation of particle surface concentration was highly correlated with NO_x in all seasons, indicating that the variation is predominantly determined by traffic emissions, especially during the morning and evening rush hours, and influenced by greater air mass transport from suburban and rural areas at midday. In Jinan, traffic emissions are main

source of particle number concentrations, and air mass transport from suburban and rural areas turn more important at midday. The subsequent growth of GMD in the afternoon indicates the suitable conditions for particle growth in the heavily polluted air of urban Jinan.

Compared with other studies in urban areas, the variations of particle number and related gas concentrations, between weekdays and weekends are smaller in Jinan. Two reasons for this characteristic have been deduced from society social practices and terrain, respectively. Through correlation analysis, particle number concentrations are predominantly influenced by traffic emissions and coal firing for all seasons and these sources have different levels of impact depending on day of the week.

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References

- Bellouin, N., Boucher, O., Haywood, J., & Reddy, M. S. (2005). Global estimate of aerosol direct radiative forcing from satellite measurements. *Nature*, *438*, 1138–1141.
- Burtscher, H. (2005). Physical characterization of particulate emissions from diesel engines: A review. *Journal of Aerosol Science*, *36*(7), 896–932.
- Chung, C. E., Ramanathan, V., Kim, D., & Podgorny, I. A. (2005). Global anthropogenic aerosol forcing derived from satellite and ground-based observations. *Journal of Geophysical Research-Atmospheres*, *110*, D24207. doi:10.1029/2005JD006356.
- Costabile, F., Birmili, W., Klöse, S., Tuch, T., Wehner, B., Wiedensohler, A., et al. (2009). Spatio-temporal variability and principal components of the particle number size distribution in an urban atmosphere. *Atmospheric Chemistry and Physics*, *9*(9), 3163–3195.
- Crutzen, P. J., & Andreae, M. O. (1990). Biomass burning in the tropics: Impact on atmospheric chemistry and biogeochemical cycles. *Science*, *250*(4988), 1669–1678.
- Dockery, D. W., & Pope, C. A. (1994). Acute respiratory effects of particulate air pollution. *Annual Review of Public Health*, *15*, 107–132.
- Engler, C., Lihavainen, H., Komppula, M., Kerminen, V. M., Kulmala, M., & Viisanen, Y. (2007). Continuous measurements of aerosol properties at the Baltic

- Sea. *Tellus Series B-Chemical and Physical Meteorology*, 59(4), 728–741.
- Gao, J., Wang, T., Zhou, X., Wu, W., & Wang, W. (2008). Measurement of aerosol number size distributions in the Yangtze River delta in China: Formation and growth of particles under polluted conditions. *Atmospheric Environment*, 43(4), 829–836.
- Harris, S. J., & Maricq, M. M. (2001). Signature size distributions for diesel and gasoline engine exhaust particulate matter. *Journal of Aerosol Science*, 32(6), 749–764.
- Holmes, N. S. (2007). A review of particle formation events and growth in the atmosphere in the various environments and discussion of mechanistic implications. *Atmospheric Environment*, 41(10), 2183–2201.
- Hussein, T., Puustinen, A., Aalto, P. P., Makela, J. M., Hameri, K., & Kulmala, M. (2004). Urban aerosol number size distributions. *Atmospheric Chemistry and Physics*, 4, 391–411.
- IPCC (Intergovernmental Panel on Climate Change) (2001). Climate change 2001: The scientific basis. In J. T. Houghton, Y. Ding, D. J. Griggs, M. Noguer, P. J. van der Linden, X. Dai, et al. (Eds.), *Contribution of working group I to the third assessment report of the intergovernmental panel on climate change*. Cambridge: Cambridge University Press.
- Kittelson, D. B., Watts, W. F., & Johnson, J. P. (2004). Nanoparticle emissions on Minnesota highways. *Atmospheric Environment*, 38(1), 9–19.
- Kulmala, M., Maso, M., & Makela, J. M. (2001). On the formation, growth and composition of nucleation mode particles. *Tellus Series B-Chemical and Physical Meteorology*, 53(4), 479–490.
- Kulmala, M., Petäjä, T., Mönkkönen, P., Koponen, I. K., Dal Maso, M., Aalto, P. P., et al. (2004a). On the growth of nucleation mode particles: Source rates of condensable vapor in polluted and clean environments. *Atmospheric Chemistry and Physics*, 4(5), 409–416.
- Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V. M., et al. (2004b). Formation and growth rates of ultrafine atmospheric particles: A review of observations. *Journal of Aerosol Science*, 35(2), 143–176.
- Laakso, L., Hussein, T., Aarnio, P., Komppula, M., Hiltunen, V., Viisanen, Y., et al. (2003). Diurnal and annual characteristics of particle mass and number concentrations in urban, rural and Arctic environments in Finland. *Atmospheric Environment*, 37(19), 2629–2641.
- Menon, S., Del Genio, A. D., Koch, D., & Tselioudis, G. (2002). GCM simulations of the aerosol indirect effect: Sensitivity to cloud parameterization and aerosol burden. *Journal of the Atmospheric Sciences*, 59(3), 692–713.
- Morawska, L., Bofinger, N. D., Kocis, L., & Nwankwoala, A. (1998). Submicrometer and supermicrometer particles from diesel vehicle emissions. *Environmental Science & Technology*, 32(14), 2033–2042.
- Morawska, L., Jayaratne, E. R., Mengersen, K., Jamriska, M., & Thomas, S. (2002). Differences in airborne particle and gaseous concentrations in urban air between weekdays and weekends. *Atmospheric Environment*, 36(27), 4375–4383.
- Nemmar, A., Hoet, P. H., Dinsdale, D., Vermylen, J., Hoylaerts, M. F., & Nemery, B. (2003). Diesel exhaust particles in lung acutely enhance experimental peripheral thrombosis. *Circulation*, 107(8), 1202–1208.
- Oberdörster, G. (2000). Toxicology of ultrafine particles: In vivo studies. *Philosophical Transactions of the Royal Society of London Series A—Mathematical Physical and Engineering Sciences*, 358(1775), 2719–2740.
- Pekkanen, J., Timonen, K. L., Ruuskanen, J., Reponen, A., Mirme, A., et al. (1997). Effects of ultrafine and fine particles in urban air on peak expiratory flow among children with asthmatic symptoms. *Environmental Research*, 74(1), 24–33.
- Ruuskanen, J., Tuch, T., Ten Brink, H., Peters, A., Khlystov, A., Mirme, A., et al. (2001). Concentrations of ultrafine, fine and PM_{2.5} particles in three European cities. *Atmospheric Environment*, 35(21), 3729–3738.
- Scholes, R. J., Ward, D. E., & Justice, C. O. (1996). Emissions of trace gases and aerosol particles due to vegetation burning in southern hemisphere Africa. *Journal of Geophysical Research-Atmospheres*, 101(D19), 23677–23682.
- Seinfeld, J. H., & Pandis, S. N. (1998). *Atmospheric chemistry and physics: From air pollution to climate change*. New York: Wiley.
- Shi, Z. B., He, K. B., Yu, X. C., Yao, Z. L., Yang, F. M., Ma, Y. L., et al. (2007). Diurnal variation of number concentration and size distribution of ultrafine particles in the urban atmosphere of Beijing in winter. *Journal of Environmental Sciences-China*, 19(8), 933–938.
- Stanier, C. O., Khlystov, A. Y., & Pandis, S. N. (2004a). Ambient aerosol size distributions and number concentrations measured during the Pittsburgh Air Quality Study (PAQS). *Atmospheric Environment*, 38(20), 3275–3284.
- Stanier, C. O., Khlystov, A. Y., & Pandis, S. N. (2004b). Nucleation events during the Pittsburgh Air Quality Study: Description and relation to key meteorological, gas phase, and aerosol parameters. *Aerosol Science and Technology*, 38(12), 253–264.
- Stott, P. A., Tett, S. F. B., Jones, G. S., Allen, M. R., Mitchell, J. F. B., & Jenkins, G. J. (2000). External control of 20th century temperature by natural and anthropogenic forcings. *Science*, 290, 2133–2137.
- Venkataraman, C., & Rao, G. U. M. (2001). Emission factors of carbon monoxide and size-resolved aerosols from biofuel combustion. *Environmental Science & Technology*, 35(10), 2100–2107.
- Voigtlander, J., Tuch, T., Birmili, W., & Wiedensohler, A. (2006). Correlation between traffic density and particle size distribution in a street canyon and the dependence on wind direction. *Atmospheric Chemistry and Physics*, 6, 4275–4286.

- Wehner, B., Birmili, W., Gnauk, T., & Wiedensohler, A. (2002). Particle number size distributions in a street canyon and their transformation into the urban-air background: Measurements and a simple model study. *Atmospheric Environment*, *36*(13), 2215–2223.
- WHO (2005). *World health organization air quality guidelines*. Global Update, E87950.
- Woo, K. S., Chen, D. R., Pui, D. Y. H., & McMurry, P. H. (2001). Measurement of Atlanta aerosol size distributions: Observations of ultrafine particle events. *Aerosol Science and Technology*, *34*(1), 75–87.
- Wu, Z. J., Hu, M., Liu, S., Wehner, B., Bauer, S., Andreas, M. B., et al. (2007). New particle formation in Beijing, China: Statistical analysis of a 1-year data set. *Journal of Geophysical Research-Atmospheres*, *112*(D9), D09209.
- Yang, L. X., Wang, D. C., Cheng, S. H., Wang, Z., Zhou, Y., Zhou, X. H., et al. (2007). Influence of meteorological conditions and particulate matter on visual range impairment in Jinan, China. *Science of the Total Environment*, *383*, 164–173.
- Yue, D. L., Hu, M., Wu, Z. J., Wang, Z. B., Guo, S., Wehner, B., et al. (2009). Characteristics of aerosol size distributions and new particle formation in the summer in Beijing. *Journal of Geophysical Research-Atmospheres*, *114*, D00G12, doi:[10.1029/2008JD010894](https://doi.org/10.1029/2008JD010894).
- Zhang, Q., Stanier, C. O., Canagaratna, M. R., Pandis, S. N., & Jimenez, J. L. (2004). Insights into the chemistry of new particle formation and growth events in Pittsburgh based on aerosol mass spectrometry. *Environmental Science & Technology*, *38*(18), 4797–4809.