

Size Distributions of Aerosol Sulfates and Nitrates in Beijing during the 2008 Olympic Games: Impacts of Pollution Control Measures and Regional Transport

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(Received 7 March 2012; revised 15 May 2012)

ABSTRACT

For the 2008 Olympic Games, drastic control measures were implemented on industrial and urban emissions of sulfur dioxide (SO₂), nitrogen oxides (NO_x) and other pollutants to address the issues of poor air quality in Beijing. To investigate the effects of SO₂ and NO_x reductions on the particulate sulfate and nitrate concentrations as well as their size distributions, size-segregated aerosol samples were collected using micro-orifice uniform deposit impactors (MOUDIs) at urban and downwind rural sites in Beijing before and after full-scale controls. During the sampling period, the mass concentrations of fine particles (PM_{1.8}) at the urban and rural sites were 94.0 and 85.9 μg m⁻³, respectively. More than 90% of the sulfates and ~60% of nitrates formed as fine particles. Benefiting from the advantageous meteorological conditions and the source controls, sulfates were observed in rather low concentrations and primarily in condensation mode during the Olympics. The effects of the control measures were separately analyzed for the northerly and the southerly air-mass-dominated days to account for any bias. After the control measures were implemented, PM, sulfates, and nitrates were significantly reduced when the northerly air masses prevailed, with a higher percentage of reduction in larger particles. The droplet mode particles, which dominated the sulfates and nitrates before the controls were implemented, were remarkably reduced in mass concentration after the control measures were implemented. Nevertheless, when the polluted southerly air masses prevailed, the local source control measures in Beijing did not effectively reduce the ambient sulfate concentration due to the enormous regional contribution from the North China Plain.

Key words: size distribution, aerosol, sulfate, nitrate, control effect, Olympic Games

Citation: Wang, X. F., T. Wang, R. K. Pathak, M. Hallquist, X. M. Gao, W. Nie, L. K. Xue, J. Gao, R. Gao, Q. Z. Zhang, W. X. Wang, S. L. Wang, F. H. Chai, and Y. Z. Chen, 2013: Size distributions of aerosol sulfates and nitrates in Beijing during the 2008 Olympic Games: Impacts of pollution control measures and regional transport. *Adv. Atmos. Sci.*, **30**(2), 341–353, doi: 10.1007/s00376-012-2053-4.

1. Introduction

Aerosols are one of the most important atmospheric pollutants. Aerosols are associated with adverse human health, visibility reduction, and climate

change (Heintzenberg, 1989; Charlson and Heintzenberg, 1995; Vedal, 1997; Chang et al., 2009; Wang et al., 2010c). The size distributions of aerosols are crucial for understanding aerosol formation, conversion, transportation, and removal processes, and in assess-

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ing the effects of aerosols on human health and the global radiation budget (Liu et al., 2008)). Most fine particles are believed to be normally formed through secondary processes (gas to particle or gas to liquid conversion), and coarse particles are generally directly emitted from the anthropogenic and/or natural sources (primary processes). Anthropogenic pollutants (gases and particles), generated largely in urban and industrial regions, are considered to be the principal contributors to the generation of fine aerosols in the atmosphere.

The recent rapid industrialization and urbanization in China are major concerns regarding the deteriorating regional air quality and global climate change (Chan and Yao, 2008). Beijing (39.13° – 41.08° N, 115.22° – 117.50° E) is the capital of China, with a population of 17.55 million people (NBSC, 2011). It is located on the northern edge of the North China Plain. Urbanization, coal combustion, and increasing traffic in recent decades in Beijing have significantly contributed to severe atmospheric particulate pollution (Zhang et al., 2000; Yao et al., 2002; Wang et al., 2008). In Beijing, fine particles dominate the particulate matter (Guo et al., 2010). The fine particles of Beijing are primarily composed of sulfates, nitrates, ammonium, and carbonaceous aerosols (Yao et al., 2002; Sun et al., 2004; Hu et al., 2005; Roger et al., 2009). Sulfates and nitrates, which are significantly responsible for the regional haze, are primarily formed through atmospheric oxidation of SO_2 emitted from coal combustion and NO_x from vehicle exhaust, respectively (Seinfeld and Pandis, 2006). Previous studies suggest that the size distribution of sulfates often exhibits a unimodal distribution in fine mode and nitrates exhibits a bimodal distribution in urban Beijing (Yao et al., 2003; Hu et al., 2005). Furthermore, sulfates are found generally formed regionally, but the nitrates can be produced locally. The in-cloud process primarily contributes to the generation of sulfate during the summer (Guo et al., 2010).

The 29th Olympic Games were hosted in Beijing from 8 to 24 August 2008. The Chinese government made air-quality improvement a top priority and concern. To significantly improve the air quality in Beijing during the Games, a series of control measures for reducing the emissions of air pollutants from coal combustion, road traffic, industries, construction, and other anthropogenic sources were intensively implemented from 20 July to 20 September 2008. Coal-fired factories in Beijing were shut down, and industrial production in the city region was also reduced. The vehicles that failed to meet the Exhaust Emission Standard of Euro-III were restricted from entering into the urban areas, and all private cars were sub-

ject to the alternate day-driving scheme. The production from 27 cement plants, and 106 quarry and lime plants were also halted, and all construction activity was completely stopped (UNEP et al., 2009; Wang et al., 2009a, 2010a). Moreover, another 105 corporations in Beijing either halted or limited work, and dozens of polluting factories in Hebei were also closed before the Olympics. According to a preliminary assessment from the UNEP, the emissions of SO_2 , NO_x and PM_{10} in the Beijing area were reduced by 14%, 38% and 20%, respectively, after the implementation of the control measures. In more recent studies, much larger emission reductions of SO_2 (41%), NO_x (46%–47%), and PM_{10} (52%–55%) are suggested to have occurred during the Olympics when compared to the non-Olympic period (Wang et al., 2010a, Zhou et al., 2010).

A significant reduction in the source emissions in the local Beijing area provided a unique opportunity to investigate the impacts of the source controls on air quality and their implications on secondary aerosols in particular. Many studies on the effects of control measures on air quality have been published recently. Without exception, all such studies have observed a significant reduction of primary air pollutants in Beijing. For example, on-road and urban measurements indicate a remarkable decrease of ambient SO_2 (70%), NO_x (41%), and BC (12%) and coarse particulate matter (43%) during the Olympics compared to the non-Olympic period (Wang et al., 2009a, c). During August 2008, the surface concentrations of SO_2 and NO_y in urban plumes dropped by 21%–61% when compared to the same period in previous years (Wang et al., 2009b). For secondary aerosols, however, $\text{PM}_{2.5}$ and fine sulfates and nitrates exhibited rather complex or different responses to the control measures due to various processes, e.g., regional transport driven by meteorology. The control measures appear to have not significantly reduced on the level of secondary aerosols during the first 2 weeks after the full-scale controls (Wang et al., 2010b; Okuda et al., 2011; Xing et al., 2011).

In this study, we analyzed the data on the size-resolved water soluble ions in atmospheric aerosols that were simultaneously collected at urban and rural sites in Beijing during the unique Olympic Games period. We compared the concentrations and size distributions of PM, sulfates and nitrates for periods before and after the implementation of full-scale controls, as well as during the Olympics. The effects of source reductions on aerosol sulfate and nitrate were separately evaluated in the framework of northerly and southerly air-mass-dominated days to minimize any bias from the meteorological conditions. The production and transportation of sulfates and nitrates in Beijing are

also discussed via comparisons of data from the urban site with those from the rural site.

2. Experiment and methods

2.1 Sampling sites

In this study, two sites were involved in collecting size-resolved aerosol samples (shown in Fig. 1), which are the same as those in our previous studies (Wang et al., 2010b). One site is in the Chinese Research Academy of Environmental Science (CRAES) beside the North Fifth Ring Road in urban Beijing (abbreviated to “CR”; 40°02'N, 116°25'E, 44 m a.s.l.). The CR site is near the center of Beijing. Another site is located in Heishanzai, Changping district, Beijing (abbreviated to “HSZ”; 40°22'N, 116°18'E, 303 m a.s.l.). The HSZ site is in rural Beijing, where elevated concentrations of ozone and fine nitrates were observed in our previous study in the summer of 2005 (Wang et al., 2006; Pathak et al., 2009). The distance between the urban and rural sites is ~ 37.4 km.

2.2 Measurements and analysis

During this study, two sets of Micro-Orifice Uniform Deposit Impactors (MOUDIs) were deployed to collect aerosol samples at a flow rate of 30 L min^{-1} . There were 10 stages of the MOUDI (Model 110 with rotator) at the urban site, with size ranges of $>18 \mu\text{m}$ (inlet), $10\text{--}18 \mu\text{m}$, $5.6\text{--}10 \mu\text{m}$, $3.2\text{--}5.6 \mu\text{m}$, $1.8\text{--}3.2 \mu\text{m}$, $1.0\text{--}1.8 \mu\text{m}$, $0.56\text{--}1 \mu\text{m}$, $0.32\text{--}0.56 \mu\text{m}$, $0.18\text{--}0.32 \mu\text{m}$, $0.1\text{--}0.18 \mu\text{m}$ and $0.05\text{--}0.1 \mu\text{m}$ (Marple et al., 1991). The MOUDI (Model 100 with rotator) at the rural site had only eight stages and did not include

the two smallest stages listed above. Size-segregated aerosol samples were collected at both sites from 11 July to 24 August 2008. The aerosol samples were mostly collected over 24-hour periods from 0700 LST in the morning to the next 0700 LST. During pollution episodes, aerosol samples were collected over 12-hour periods from 0700 to 1900 LST for the daytime samples and from 1900 to 0700 LST for the nighttime samples. Samples were not collected during most rainy days, because the sampler had no device for water removal. Once the sampling was completed, all impactors in the MOUDI were cleaned using absorbent cotton soaked with absolute alcohol, followed by the use of high-purity nitrogen gas to remove the remaining alcohol and to unblock the micro-orifice nozzles.

Aluminum substrates (MSP) were used in both MOUDIs to collect aerosol samples in this study. Before sampling, the substrates were pre-heated at 500°C using a muffle furnace for four hours to remove residual organics. After sampling, the aluminum substrates were placed in plastic Petri dishes and then stored in a refrigerator at a temperature below -5°C for subsequent gravimetric and chemical analysis in laboratory. The substrates were weighed using a microbalance (ME5, Sartorius) with precision of $1 \mu\text{g}$ before and after collecting the samples. Before weighing, the substrates were balanced for 48 hours in the conditions of constant temperature ($20^\circ\text{C} \pm 0.5^\circ\text{C}$) and constant relative humidity ($50\% \pm 2\%$).

The atmospheric aerosol samples collected on the aluminum substrates were completely dissolved by treating the substrates with ultrasonic waves in 10 mL of deionized water. Inorganic water-soluble ions in the resulting sample solutions were detected with an ion

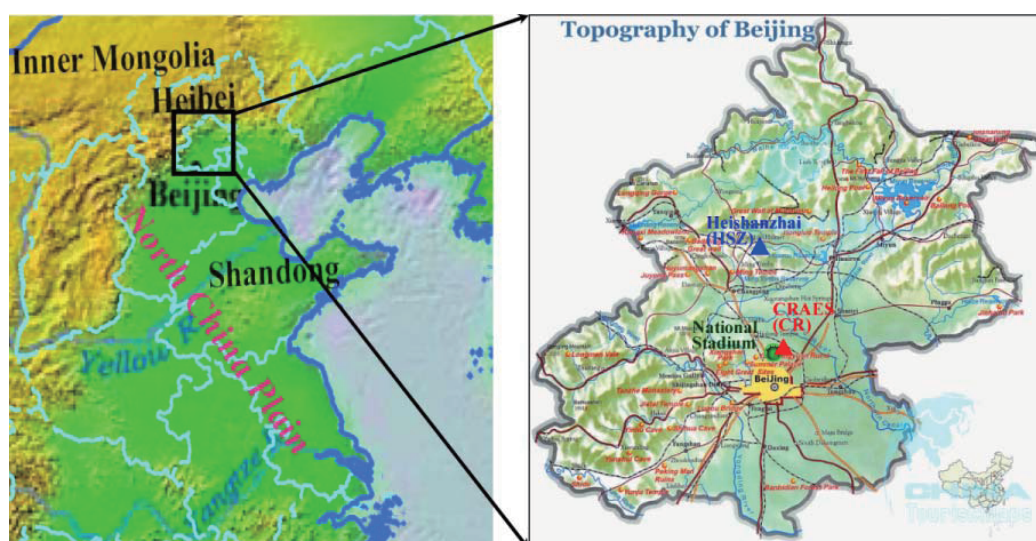


Fig. 1. Locations of the sampling sites.

chromatograph (Dionex ICS 90). The anions (F^- , Cl^- , NO_2^- , NO_3^- , SO_4^{2-}) were analyzed using an AS14A Column and an AMMS 300 Suppressor with eluent of $3.5 \text{ mmol L}^{-1} \text{ Na}_2\text{CO}_3$ – $1.0 \text{ mmol L}^{-1} \text{ NaHCO}_3$ at a flow rate of 1.2 mL min^{-1} . The cations (Na^+ , NH_4^+ , K^+ , Mg^{2+} , and Ca^{2+}) were analyzed using a CS12A Column and a CSRS Ultra II Suppressor with eluent of 20 mmol L^{-1} methanesulfonic acid (MSA) at a flow rate of 1.0 mL min^{-1} . The method of analysis used was the same as that reported by Gao et al. (2011). During the chemical analysis using the ion chromatograph in the laboratory, multi-point calibrations were performed for all of these ions every day after the eluent solutions were changed.

Concurrently, a number of other air pollutants and parameters were measured at the urban and rural sites during this study. SO_2 was measured using a pulsed UV fluorescence analyzer (TEI model 43C), O_3 was measured using a UV photometric analyzer (TEI model 49i), and CO was measured using a non-dispersive infrared analyzer (API model 300EU). NO and NO_2 were measured by a commercial chemiluminescence analyzer (TEI model 42i) equipped with a photolytic converter (blue light converter, Meteorologie Consult GmbH). Water-soluble ions in $PM_{2.5}$ were measured using an ambient ion monitor (AIM, URG-9000B) at both sites. $PM_{2.5}$ samples were also collected by a Reference Ambient Air Sampler (RAAS 2.5-400, Thermo Electron Corporation, Waltham, MA, USA) with Teflon filters and followed by subsequent gravimetric analysis to determine the $PM_{2.5}$ concentrations. In addition, the ambient temperature, relative humidity, wind direction, and speed were measured by a portable automatic meteorological station. A detailed description of the measurement techniques used is described in Wang et al. (2010b).

2.3 Sampling artifacts and data correction

Aluminum substrates are stable and quite suitable for determining the mass of aerosols collected on them. In past decades, aluminum substrates have been extensively used in MOUDI sampling (e.g., Chen et al., 1997; Maenhaut et al., 2002; Moya et al., 2004; Duan et al., 2005, 2007; Lin et al., 2008; Massling et al., 2009). However, the use of aluminum substrates for aerosols is subject to sampling artifacts due to bounce-off and evaporation loss (Stein et al., 1994; Duan et al., 2005; Nie et al., 2010). The bounce-off loss is generally significant at low RH. The evaporation loss of semi-volatile components such as nitrate ammonium depends upon the ambient temperature, RH, aerosol loading on the substrate or filter and the pressure drop across the sampler (e.g., Dasch et al., 1989; Zhang

and McMurry, 1992; Cheng and Tsai, 1997; Pathak et al., 2004, 2009). The average RH and temperature in urban and rural Beijing during the sampling period were $58\% \pm 17\%$ and $80\% \pm 16\%$, respectively, and $27^\circ\text{C} \pm 4^\circ\text{C}$ and $25^\circ\text{C} \pm 4^\circ\text{C}$, respectively. In our study, apparent losses of both types were observed in some aerosol samples collected by MOUDI.

To examine the sampling artifacts of the MOUDIs at the urban and rural sites, the $PM_{2.5}$ mass concentrations from the RAAS and the $PM_{2.5}$ ion concentrations from the AIM were used as benchmarks. For comparison, the PM and ion concentrations in particle sizes $< 2.5 \mu\text{m}$ for MOUDI were estimated as

$$PM_{2.5} = PM_{1.8} + PM_{1.8-3.2} * \lg(2.5/1.8) / \lg(3.2/1.8).$$

Compared to the RAAS, the $PM_{2.5}$ mass concentrations from the MOUDI had an average sampling loss of 29% at the urban site and 33% at the rural site. Compared to the AIM, the average sampling loss (bounce-off loss) of MOUDI for $PM_{2.5}$ sulfates concentrations was 19% at the urban site and 35% at the rural site. For semi-volatile components of fine nitrates and chlorides, the sampling loss included both bounce-off and evaporation types, and thus was much higher, that is, a loss of $\sim 50\%$ at the two sites. Assuming that the bounce-off loss of the MOUDI for the fine nitrates was the same as that for the fine sulfate, the average evaporation loss of the $PM_{2.5}$ nitrates was $\sim 30\%$. Generally, the sampling losses of PM and water-soluble ions at the rural site were higher than those at the urban site, which was likely because of the different operating conditions and the errors in the manual operation of the MOUDIs. Sampling loss is common for the MOUDI, and the degree of loss largely depends on the ambient weather conditions, the aerosol loading and composition, the working condition of the collector, and the manual operation errors. However, in most studies, the MOUDI data have not been corrected because the sampling loss is generally small or because the degree of sampling loss is not clear due to lack of additional instruments/measurements for comparison. In this study, the sampling losses of the MOUDIs were large, and the concurrent measurements of $PM_{2.5}$ mass concentration using RAAS and $PM_{2.5}$ ion concentrations using AIM provided the possibility to correct the sampling artifacts at both sites.

To eliminate or avoid significant uncertainty involving bounce-off or evaporation losses, the sampling artifacts of size-segregated samples were corrected. The MOUDI samples were divided into two or three groups according to the degree of sampling loss for $PM_{2.5}$ or each ion. For each group of samples, an average integrated loss degree was determined from a regression plot ($R^2 > 0.93$) between MOUDI and RAAS/AIM and was used to further constrain the

sampling loss in different size bins. The loss function for each size bin for a certain degree of integrated loss was derived from the differences between the MOUDI samples with and without sampling loss. The size-resolved MOUDI data were then corrected based on the constrained loss percentage in each size range. Only bounce-off loss was considered for PM and sulfates in this correction. For semi-volatile components such as nitrates and chlorides, bounce-off and evaporation loss were corrected separately. The bounce-off loss was regarded to be the same as that for sulfates, and the remaining loss was considered to be evaporation loss. After correction of the sampling loss, excellent correlation was obtained between the MOUDI data and that from RAAS/AIM ($R^2 > 0.98$ for $PM_{2.5}$, sulfate and nitrate), and the corrected MOUDI data were believed to exhibit reduced uncertainty.

2.4 Air mass backward trajectory analysis

In this study, air-mass backward trajectories for each sample were calculated using HYSPLIT Trajectory Model with Global Data Assimilation System data online (Draxler and Rolph, 2012). For each sample, six 48-h backward trajectories were calculated at 50 m above ground level and used to determine the direction of the air masses during the sampling time. For each sampling day air masses arrived from approximately the same direction, thus, the trajectory shown in Fig. 2 was used in the middle of sampling time. The air masses at the rural site had very little difference from those in the urban site.

3. Results and discussions

3.1 Overall results

Because there is no 2.5- μm cutoff point for MOUDI, the diameter of 1.8 μm was defined as the cutoff point that divides fine and coarse particles in this study; that is, $PM_{1.8}$ and $PM_{1.8-10}$ represented the fine and coarse particles, respectively. The average concentrations ($n=26$) of $PM_{1.8}$ and $PM_{1.8-10}$ were 94.0 ± 72.5 and $32.2 \pm 17.8 \mu\text{g m}^{-3}$, respectively, at the urban site, and 85.9 ± 73.4 and $22.7 \pm 16.7 \mu\text{g m}^{-3}$, respectively, at the rural site (see Table 1). Compared with previous studies during summer in Beijing (Hu et al., 2005; Guo et al., 2010), the $PM_{1.8}$ and PM_{10} concentrations in urban Beijing were slightly lower than those measured in August 2006; however, the urban $PM_{1.8}$ concentration was still higher than that in July 2001 and 2002. The downwind rural $PM_{1.8}$ concentration was slightly higher than that in the upwind rural site (Yufa) in August 2006.

The ratios of $PM_{1.8}$ to PM_{10} were 0.70 ± 0.06 and 0.74 ± 0.09 at the urban and rural sites, respectively, indicating the dominance of fine particulates in urban and rural aerosols in Beijing. More than 90% of the sulfates were found in the fine particles; however, only $\sim 60\%$ of the nitrates were found in the fine particles. Ammonium was primarily found in the fine particles, and is likely primarily associated with fine sulfates and nitrates. The average concentrations of sulfates and nitrates in the fine particles were 28.6 ± 26.0 and

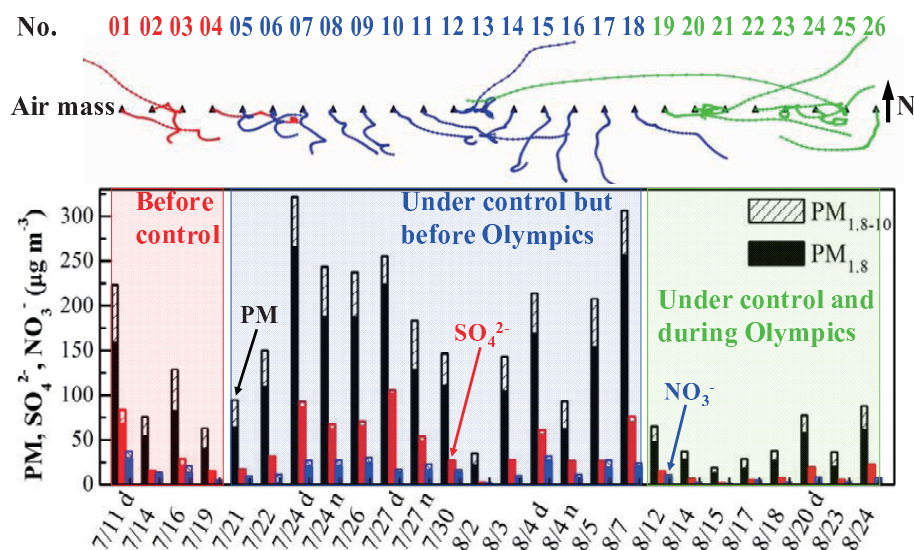


Fig. 2. Sample numbers corresponding 48-h backward trajectories with the endpoint in the urban site at 50 m above the ground, and concentrations of PM, sulfate, and nitrate in fine and coarse particles. Note “d” for daytime and “n” for nighttime of the day.

Table 1. Average concentrations of aerosol and ions in fine and coarse particles, ratio of $PM_{1,s}$ to PM_{10} and the ratios of ions in fine particles ($\mu\text{g m}^{-3}$).

	CR (23 days, 26 sets)				HSZ (22 days, 26 sets)			
	$PM_{1,s}$	$PM_{1,s-10}$	$PM_{1,s}/PM_{10}$	Ion/ $PM_{1,s}$	$PM_{1,s}$	$PM_{1,s-10}$	$PM_{1,s}/PM_{10}$	Ion/ $PM_{1,s}$
	PM	94.0±72.5	32.2±17.8	0.70±0.06	0.27±0.07	85.9±73.4	22.7±16.7	0.74±0.09
SO_4^{2-}	28.6±26.0	2.7±3.6	0.92±0.05	0.10±0.04	29.8±30.0	1.9±2.1	0.94±0.02	0.11±0.07
NO_3^-	9.9±8.7	4.2±2.6	0.63±0.17	0.01±0.03	11.4±11.1	3.9±3.0	0.65±0.18	0.00±0.00
Cl^-	0.8±0.8	0.3±0.2	0.56±0.25	0.15±0.05	0.3±0.3	0.1±0.1	0.65±0.13	0.16±0.04
NH_4^+	16.0±15.2	1.4±1.5	0.90±0.05	0.01±0.00	16.4±15.7	1.2±0.9	0.90±0.05	0.01±0.00
K^+	0.7±0.5	0.1±0.1	0.87±0.04	0.00±0.00	0.7±0.7	0.1±0.1	0.82±0.07	0.00±0.00
Na^+	0.2±0.1	0.1±0.0	0.69±0.09	0.00±0.00	0.1±0.1	0.0±0.0	0.71±0.11	0.00±0.00
Ca^{2+}	0.4±0.3	1.7±0.9	0.21±0.13	0.01±0.02	0.2±0.1	0.7±0.5	0.23±0.09	0.00±0.00

9.9±8.7 $\mu\text{g m}^{-3}$, respectively, in urban Beijing, and 29.8±30.0 and 11.4±11.1 $\mu\text{g m}^{-3}$, respectively, at the rural site. The average sulfate and nitrate concentrations in fine particles at the rural site were slightly higher than those measured at the urban site, which may be attributed to the aging process of aerosols that led to the formation of more secondary aerosols over the distance at high RH (a detailed analysis can be seen in section 3.4). The concentrations of crustal and sea salt species were generally small. K^+ was distributed primarily in fine particles, Ca^{2+} in coarse particles and Cl^- and Na^+ approximately evenly in both fine and coarse particles. Generally, the size-resolved inorganic ion compositions of particulate matter were similar at the urban and downwind rural sites. Thus, the temporal variations, size distributions and control effects were only analyzed for the urban samples.

3.2 Characteristics of PM, sulfate and nitrate in different periods

According to the dates when the control measures were implemented and the Olympics began, the sampling periods were classified as: (1) BC—the period before the control measures were enforced (11 to 19 July), (2) CBO—period under source control but before Olympics (20 July to 7 August), (3) CDO—period under source control and during Olympics (8 to 24 August). The periods of both CBO and CDO were after control measures were implemented, and thus the sum of the two was designated as AC.

The concentrations of PM and of sulfates and nitrates in both fine and coarse particles measured at the urban site are shown in the time series in Fig. 2. Before full-scale controls were implemented, moderately high concentrations of PM, sulfates and nitrates were observed, with concentration peaks observed in droplet mode (0.56–1.8 μm) (Fig. 3). Notably, heavy particulate pollution episodes were also encountered after the enforcement of control measures but before the Olympics began. The highest total sulfate concentration of 105 $\mu\text{g m}^{-3}$ (12-hour average) was recorded for the daytime of 27 July. The average concentrations of PM and sulfates were even higher than those in the first period, particularly within the size range of 0.32–1.8 μm . During the Olympic Games, the mass concentrations of PM, sulfate and nitrate were rather low. Sharp decreases were observed for PM, sulfates, and nitrates in both fine and coarse particles. Furthermore, the concentration peaks of PM and sulfates decreased, and they shifted to smaller particles in condensation mode (0.1–0.56 μm) compared to those before the Olympics.

In our previous study (Wang et al., 2010b), a dramatic increase of ozone and fine sulfates was observed

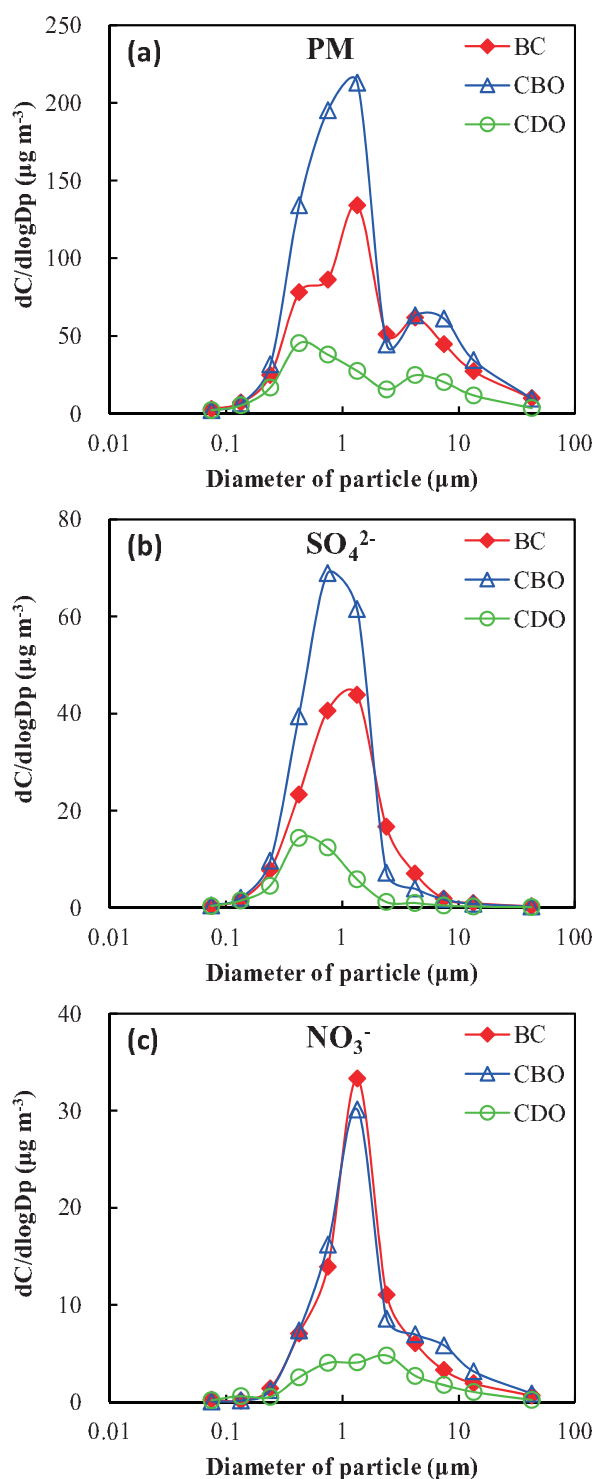


Fig. 3. Average size distributions of PM, sulfate, and nitrate concentrations in urban Beijing for the three periods. Numbers of samples in periods of BC, CBO and CDO were 4, 14 and 8, respectively.

during the second period, which was associated with significantly more air masses from the SE–S–SW di-

rections when compared to the other two periods. This finding is consistent with the results shown in the present dataset. Upon examination of the corresponding 48-hour backward trajectories for each sample (Fig. 2), a generally elevated concentration of sulfates was observed when the air masses prevailed from south-sector (south, southeast, and southwest), where most of the coal-fired power plants and industries are located. Whenever the air masses prevailed from the north corridor—a region of minimal industrial activities—the concentrations were generally below the annual averages (Zhang et al., 2006; Chan and Yao, 2008; Zhao et al., 2009). In the CBO period, the air masses generally prevailed from the southern corridor, and Beijing city experienced a large number of particulate pollution episodes characterized by the fine particles being primarily in droplet mode. In contrast, during the Olympics period, the air masses primarily prevailed from the north. With cleaner air masses and occasional precipitation, the city experienced better than expected air quality (Wang et al., 2009c; Wang et al., 2010b). The concentration peaks of sulfates shifted to a smaller size (0.32–0.56 μm), which might be associated with advantageous meteorological conditions as well as emissions reduction. This observed shift is discussed in detail in the next section.

We also noted that the average size distributions of nitrate in the three periods were nearly unimodal, primarily because the nitrate size distributions for individual samples were not very regular. For most of the aerosol samples, the nitrate size distributions were bimodal with one peak in the fine mode and another in the coarse mode; however, concentration peaks were likely present in different size bins. Sometimes the nitrate concentration peak in the coarse mode was so small that it was masked by the large peak in the fine mode.

3.3 Effects of control measures on PM, sulfate and nitrate

As shown in Fig. 2, the size-resolved aerosol sampling days were categorized as northerly or southerly air mass dominated based on the routes of the air masses arriving at our sampling site. The air masses from the east, northeast, north, northwest or west areas are classified as northerly, and those from southeast, south and southwest regions are classified as southerly. When northerly air masses prevailed, the urban particulate concentration embodied the degree of air pollution from local emissions in combination with a dilution effect from the clean northerly air masses, and thus primarily represented the local emissions. While under the influence of the polluted southerly air masses, the measured data in urban Bei-

ing represented the regional rather than local characteristics. Furthermore, due to the particular terrain in Beijing, featuring with mountains in the north and plains in the south, the northerly wind helps diffuse air pollutants, while the southerly wind is conducive to the accumulation of pollutants. The alternating appearance of clean and polluted episodes is common in Beijing (Wang et al., 2009c; Guo et al., 2010); however, this alternating behavior complicated the evaluation of the effects of control measures on aerosol sulfates and nitrates. Therefore, in this study, the effects of control measures were separately analyzed for northerly and southerly air-mass dominated days, which can diminish or eliminate the influence of different meteorological conditions (especially the air masses and the pollutant dispersion). A limitation of this study was the limited number of samples for each type in the BC and AC periods.

3.3.1 Effects for northerly air-mass dominated days

For northerly air-mass-dominated days, the average concentrations of size-segregated aerosols during periods before and after the controls are compared in Figs. 4a-c. Concentrations of PM, sulfates and nitrates in different size bins exhibited remarkable reduction ($\sim 50\%$) after the control measures were implemented. Moreover, the amount of reduction differed with particle sizes and tended to increase with increasing particle diameters (Table 2). The coarse particles reduced by the largest degree ($> 60\%$), which can be attributed to the strict controls on construction dust and road dust. The droplet mode, which dominated the mass concentrations before the implementation of control measures (Fig. 4), exhibited sharp reductions of 53.3%, 64.6%, and 64.1% for PM, sulfates, and nitrates, respectively. Because the days that were dominated by clean northerly air masses are believed to primarily represent the local pollution, these results suggest that a reduction in source (SO_2 , NO_x , and particle) emissions consequently reduced the PM loadings and secondary formations of sulfates and nitrates locally. However, this reduction was partially biased due to the occasional local rainfall. In addition, during the period after the control measures were implemented, the sulfate peak shifted to smaller size bins with the GMAD (geometric mean aerodynamic diameter) varying from $0.75 \mu\text{m}$ (droplet mode) to $0.42 \mu\text{m}$ (condensation mode), which indicates some changes in the formation pathways.

The drastic reduction of droplet sulfates and nitrates in northerly air-mass dominated days implied that their formation in droplet mode was largely restrained after the reduction of SO_2 , NO_x and other pollutants in the local Beijing area. The possible rea-

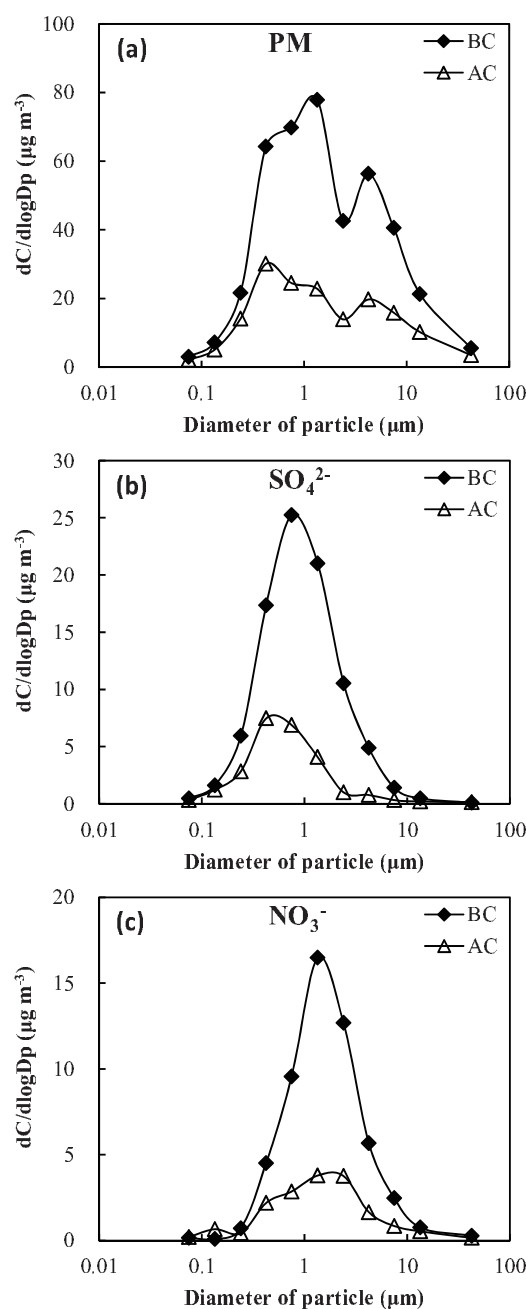


Fig. 4. Average size distributions of PM, sulfate, and nitrate concentrations in urban Beijing for northerly air-mass-dominated days before and after the implementation of control measures.

sons for this reduction are the following: (1) Emissions of precursory pollutants of SO_2 and NO_x , and thus the ambient oxidants, were all reduced. Consequently, the oxidation rates of sulfates and nitrates (SOR and NOR) were reduced (see Table 3). (2) The reduced emission as well as the ambient concentration of fine particles led to reduced cloud condensation nuclei

Table 2. Percentage reduction of PM, sulfate and nitrate concentrations in different modes for northerly air masses dominated days in urban Beijing after the implementation of control measures compared to those before the control measures were implemented. Particle modes of Aitken, condensation, droplet and coarse modes are derived from particle size ranges of 0.056–0.1 μm , 0.1–0.56 μm , 0.56–1.8 μm and 1.8–10 μm , respectively (Guo et al., 2010; Roger et al., 2009).

Mode	PM	SO ₄ ²⁻	NO ₃ ⁻
Total	53.50%	63.80%	63.00%
Aitken	41.90%	46.10%	41.10%
Condensation	43.20%	50.50%	16.40%
Droplet	53.30%	64.60%	64.10%
Coarse	62.80%	82.90%	74.70%

(CNN) activity and consequently fewer clouds (Jiménez-Escalona and Peralta, 2010), which in turn reduced the formation of sulfates and nitrates in droplet-mode via in-cloud processes. After the control measures were implemented, formation involving in-cloud processes became less important, and sulfates were likely produced via the homogeneous oxidation of SO₂ with the OH radical followed by nucleation and condensational growth in condensation mode.

3.3.2 Effects for southerly air-mass dominated days

The average concentrations of size-resolved aerosols in southerly air-mass-dominated days before and after the controls are shown in Figs. 5a–c. The effect of local emission reduction on aerosol nitrates was obvious, particularly in the droplet mode. Nevertheless, a different response was observed for the PM and sulfates after the control measures were implemented: no significant reduction in any mode. Moreover, the corresponding primary gas pollutants exhibited similar results (Table 3). After the control measures were implemented, the ambient concentration of NO_x (NO and NO₂) declined remarkably in southerly air-mass dominated days, but the SO₂ concentration increased. These observations suggest that the nitrogen compounds were primarily locally pro-

duced, while the sulfur was primarily from regional sources during pollution episodes under the influence of southern air masses.

When the southerly air masses prevailed, elevated concentrations of air pollutants were transported to Beijing from the North China Plain, where large amounts of power plants and industries were located. Under these conditions, the terrain in Beijing, with mountains in the north and plains in the south, was adverse for pollutants to diffuse, and thus heavy air pollution (especially particulate matter pollution) often occurred. The sulfates mainly formed over a regional scale. Thus, the ambient concentration of sulfates depended more on the regional emissions than the local sources and varied with the degree of the regional particle pollution. Consequently, the source reduction of SO₂ in the local Beijing area did not significantly reduce the aerosol sulfate. In contrast, the nitrates were primarily produced locally, and the ambient nitrate concentration was largely linked to the local emission sources. Thus, even under the influence of southerly air masses, the source controls of NO_x in the Beijing area can result in some reduction of aerosol nitrates.

Overall, strict control measures for reducing the emissions of SO₂, NO_x and other air pollutants in the local Beijing area was effective at reducing the ambient concentrations of PM, sulfates, and nitrates when northerly air masses prevailed. However, the sulfate level did not reduce significantly when southerly air masses dominated, because a large amount of both gas SO₂ and aerosol sulfates were transported from the industry-intensive North China Plain. Therefore, large-scale regional controls of SO₂ are required to attain a reduction of aerosol sulfates in various weather conditions.

3.4 Production and transportation of sulfates and nitrates

As previously described, the urban site (CR) is situated in the heart of Beijing City, and the rural site (HSZ) is located 37.4 km north of the urban site. We

Table 3. Average meteorological parameters, trace gas concentrations, SOR and NOR in urban Beijing before and after the implementation of control measures for northerly and southerly air-mass-dominated days. Here SOR=[SO₄²⁻]_{total}/([SO₄²⁻]_{total}+ [SO₂]); NOR=[NO₃⁻]_{total}/([NO₃⁻]_{total}+ [NO₂]), mole ratio.

Type	Period	Temp (°C)	RH (%)	WS (m s ⁻¹)	CO (ppb)	SO ₂ (ppb)	NO (ppb)	NO ₂ (ppb)	O ₃ (ppb)	SOR	NOR
Northern	BC	27.3	54.8	0.6	740	7.1	0.5	12.8	54.3	0.44	0.27
	AC	25.1	64.4	0.4	581	4.2	0.3	8.3	42.9	0.32	0.18
Southern	BC	28.2	81.1	1.9	440	4.7	1.4	22.7	70.5	0.82	0.41
	AC	28.9	54.7	0.4	922	13.2	0.5	11.6	70.5	0.44	0.40

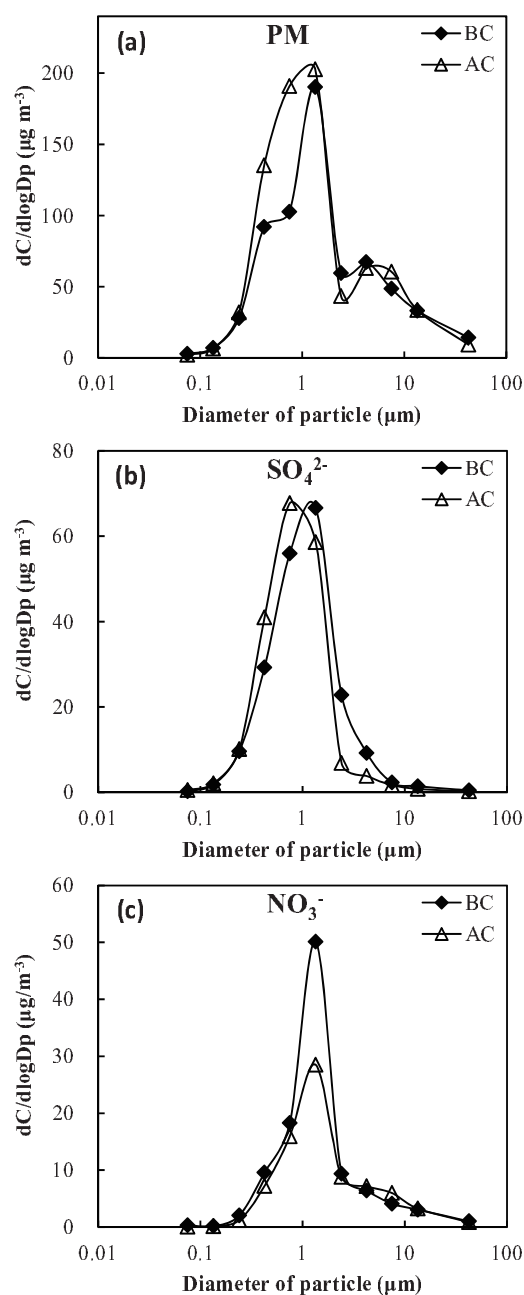


Fig. 5. Average size distributions of PM, sulfate, and nitrate concentrations in urban Beijing for southerly air-mass-dominated days before and after the implementation of control measures.

collected simultaneous samples at both sites to understand the production and transportation of sulfates and nitrates in Beijing by comparing the size-resolved samples at these two sites.

For samples collected under clean northerly air masses, the size distributions of the average sulfate and nitrate concentrations at the rural and “down-

wind” urban sites are shown in Figs. 6a and b. Generally, higher concentrations of sulfate and nitrate were observed in most of the size bins at the urban site compared to the rural site. When northerly air mass prevailed, the lower concentrations at the rural site represent the background air from the north; however, when this air travels into the urban areas of Beijing, it mixes with the locally produced pollutants, and hence elevated concentrations of sulfates and nitrates were observed. Compared to those at the rural site, much higher growth and sharp peaks were found in the accumulation mode for the urban samples, which indicates a local production of sulfates and nitrates in higher SO_2 and NO_x scenarios in the urban region.

Heavy pollution episodes were always observed when southerly air masses prevailed and passed into the urban area, feathering with extremely high atmospheric loading of sulfates and nitrates on a regional scale. The size distributions of the average sulfate and nitrate concentrations under southerly air masses are shown in Figs. 7a and b. Interestingly and in contrast to the days when northerly air masses dominated, the concentrations of sulfates and nitrates were significantly higher in droplet mode (0.56–1.8 μm) at the downwind rural site compared to the urban site. These data implied that more sulfates and nitrates were produced in the droplet mode when the polluted air plumes transported from the urban center to the downwind rural site. The high concentration of SO_2 and the high RH observed at the rural site (not shown here) indicate the plausible occurrence of heterogeneous oxidation of SO_2 to form sulfate in the droplet mode. Additionally, abundant NO_x and high humidity at the downwind rural site likely favored the enhancement of droplet mode nitrates by heterogeneous hydrolysis (Pathak et al., 2009, 2011).

4. Summary and conclusions

This study presents the size-resolved ionic compositions of atmospheric aerosols at urban and rural sites in the Beijing area during the particular period of the 2008 Olympic Games 2008 in conjunction with the preceding four weeks. Particulate matter pollution during the summer in Beijing was found to be centralized in fine particles, with an average $\text{PM}_{1.8}$ of $94.0 \mu\text{g m}^{-3}$ at the urban site and $85.9 \mu\text{g m}^{-3}$ at the rural site. Overall, both sulfates and nitrates were found to be dominant in the fine particles. During the 2008 Olympic Games period, particulate components exhibited rather low concentrations, and the concentration peak of sulfates shifted to smaller particles (condensation mode) as compared to the period before the Olympics. This finding was associated with

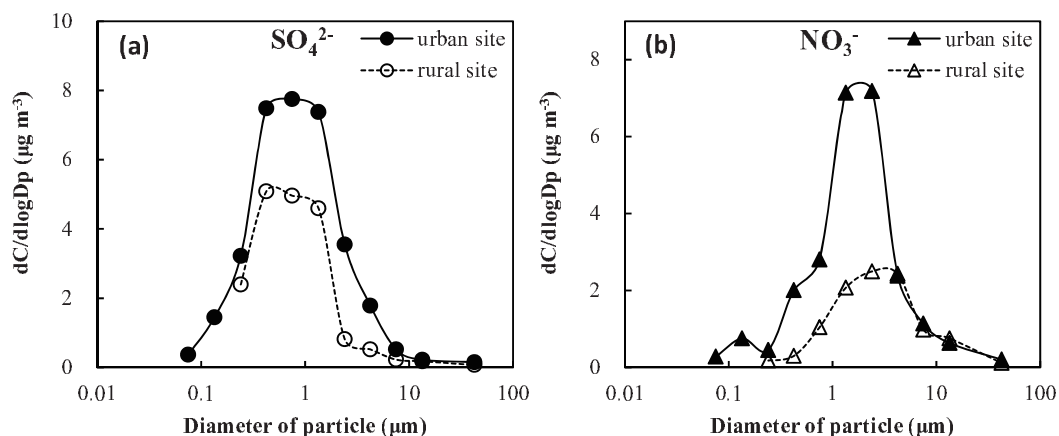


Fig. 6. Average size distributions of sulfate and nitrate concentrations at the rural site (hollow marker and dashed line) and the “downwind” urban site (solid marker and solid line) of Beijing when northerly air masses prevailed.

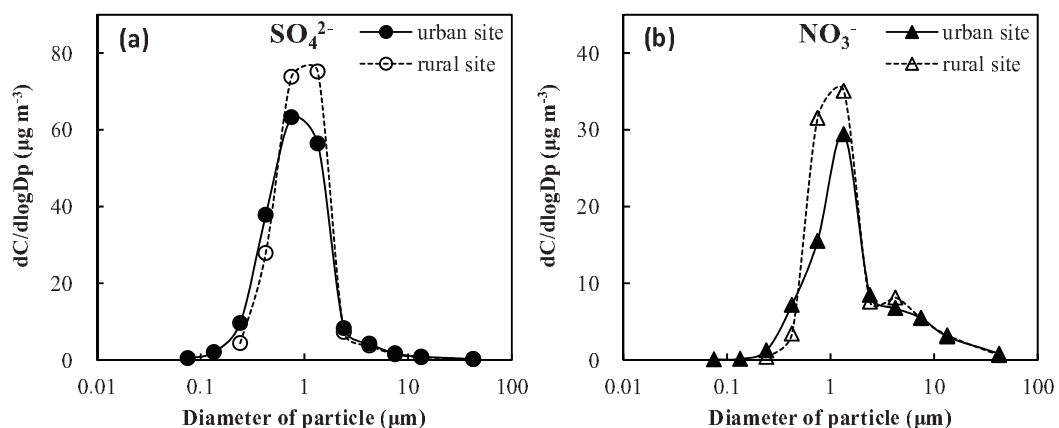


Fig. 7. Average size distributions of sulfate and nitrate concentrations at the urban site (solid marker and solid line) and the downwind rural site (hollow marker and dashed line) of Beijing when southerly air masses prevailed.

the clean northerly air masses, frequent precipitation and emission reductions. The control effects of particulate sulfates and nitrates were separately analyzed for northerly and southerly air-mass-dominated days to normalize the influence of meteorological factors. When the northerly air masses prevailed, the impacts of emission control measures appeared significant as the ambient concentration of PM, sulfates and nitrates were observed to be much lower in all modes, with an amplified reduction for the larger particle size. The in-cloud process was less important after implementation of controls, and thus more sulfates formed in condensation mode instead of droplet mode. Nevertheless, when the polluted air masses from the south prevailed, effects of the regional transportation made the source controls in the local Beijing are less effective for reducing the ambient concentration of sulfates.

A comparison between the urban and the downwind rural sites under southerly air masses reveals that the polluted air masses from the North China Plain not only transported particulate matter to Beijing but also brought a significant amount of precursory pollutants and led to further production of droplet sulfates and nitrates via heterogeneous processes under the humid conditions in downwind rural Beijing.

Acknowledgements. This work was supported by the National Key Basic Research Program of China (973 program) (Grant No. 2005CB422203), a key project of the Shandong Provincial Environmental Protection Department (Grant No. 2006045), the Niche Area Development Scheme of the Hong Kong Polytechnic University (Grant No. 1-BB94), the Hong Kong Research Grants Council (PolyU; Grant No. 5294/07E) and the central

level, scientific research institutes for basic R & D special fund business (Grant No. 2008KYYW01). The authors thank SHOU Youping, WANG Jing, YUAN Chao, WANG Linlin and WANG Xuezhong for their contributions to the sample analysis and field work.

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