**Atmospheric
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Quantitative evaluation of emission control of primary and secondary organic aerosol sources during Beijing 2008 Olympics

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Abstract

To explore the primary and secondary sources of fine organic particles after the aggressive implementation of air pollution controls during 2008 Beijing Olympic Games, 12-h PM$_{2.5}$ concentrations were measured at one urban and one upwind rural site during the CAREBeijing-2008 (Campaigns of Air quality REsearch in Beijing and surrounding region) summer field campaign. The PM$_{2.5}$ concentrations were $72.5 \pm 43.6 \, \mu g \, m^3$ and $64.3 \pm 36.2 \, \mu g \, m^{-3}$ at the urban site and rural site, respectively, which were the lowest in recent years due to the implementation of drastic control measures and favorable weather conditions. Five primary and four secondary fine organic particle sources were quantified using a CMB (chemical mass balance) model and tracer-yield method. Compared with previous studies in Beijing, the contribution of vehicle emission increased, with diesel engines contributing $16.2 \pm 5.9\%$ and $14.5 \pm 4.1\%$ to the total organic carbon (OC) concentrations and gasoline vehicles accounting for $10.3 \pm 8.7\%$ and $7.9 \pm 6.2\%$ of the OC concentrations at two sites. Due to the implementation of emission control measures, the OC concentrations from important primary sources have been reduced, and secondary formation has become an important contributor to fine organic aerosols. Compared with the non-controlled period, primary vehicle contributions were reduced by 30% and 24% in the urban and regional area, and reductions in the contribution from coal combustion were 57% and 7%, respectively. These results demonstrate the emission control measures significantly alleviated the primary organic particle pollution in and around Beijing. However, the control effectiveness of secondary organic particles was not significant.

1 Introduction

With rapid economic development and increasing energy consumption, Beijing has experienced severe air pollution and has become one of the hotspots for air pollution in the world (Shao et al., 2006). Ambient particles, especially fine particles, have been

reported as a crucial air pollutant in Beijing (Guo et al., 2010; Hu and Guo, 2009; Chan and Yao, 2008). Thus, a comprehensive understanding of the fine particle sources is very important for regulatory agencies to develop effective control policies.

Many particle source apportionment studies have been conducted in Beijing during the last a few years. Elemental tracers were previously applied to apportion the particle sources (Song et al., 2006, 2007; Sun et al., 2006). The problem of the elemental method is that it cannot characterize some important sources that emit mainly organic matter other than elemental compounds, e.g., cooking and diesel/gasoline engine exhaust. Sun et al. (2012) used radiocarbon (^{14}C) analysis to apportion the sources of carbonaceous aerosol. However, radiocarbon analysis can only distinguish fossil carbon from contemporary carbon. Particulate organic matter (POM) accounted for a large fraction in fine particles (PM$_{2.5}$) (Kanakidou et al., 2005). For instance, 30 %–50 % of the PM$_{2.5}$ was organic matter in Beijing (Lin et al., 2009). For their unique source indication, organic molecular tracers have been successfully used in particle source apportionment recently (Schauer et al., 1996; Stone et al., 2009; Guo et al., 2012). Organic tracers have been applied in several fine particle source apportionment studies in Beijing (Zheng et al., 2005; Wang et al., 2009). Zheng et al. (2005) and Wang et al. (2009) reported and quantified the source contributions for fine particles in the year of 2000 and 2005 to 2007, respectively, using a chemical mass balance (CMB) model. However, the particle sources have changed significantly in recent years, due to the change of energy structures, removal of industries, as well as new air quality control policies.

As the host of the 2008 Olympics, more attention was attracted to Beijing for its severe fine particle pollution. To alleviate this pollution, a series of control measures were implemented by Beijing municipality. Guo et al. (2012) briefly reported the major particle sources in Beijing 2008 but mainly focusing on the secondary organic aerosol formation. It is very important to compare the particle source contributions with previous studies to explore the primary particle source changes in recent years.

It is also essential to evaluate the effectiveness of the control measures for policy making, as well as better scientific understanding. Satellite-based and modeling

studies indicated significant particle pollution reduction during the Olympic period (Cermak and Knutti, 2009; Wang et al., 2010c). Huang et al. (2010) also reported the high time resolution particulate chemical compositions by using Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS), and found all major species

5 during the Olympics were low due to both control measures and favorable weather conditions. However, simply exploring variations in fine particle pollution cannot distinguish the effects between emission control and meteorological conditions. Liu et al. (2012) used a statistical model to evaluate the $PM_{2.5}$ control effects. Nevertheless, the results only showed the integrated effects of all control measures but not specific sources. To

10 quantitatively evaluate the control effect to specific sources, source apportionment is a direct and more effective way.

In the present study, ambient fine particles ($PM_{2.5}$) were collected at an urban site and an upwind rural site in Beijing before, during and after the Olympic Games in 2008. Particulate organic matter (POM) including primary and secondary organic trac-

15 ers was measured by gas chromatography/mass spectrometry (GC-MS). CMB model and tracer-yield method were employed to apportion the primary and secondary particulate organic sources. The results were compared with precious work to explore the change of particulate sources in the recent years. The variations of both primary and secondary particulate organic sources in different control periods were also discussed

20 to evaluate the emission control effectiveness. The results of this work can improve our understanding of the emission control effect on air pollution and provide a basis data set for regional air quality model.

2 Experiment

2.1 Sampling

25 12-h ambient fine particle ($PM_{2.5}$) samples were collected by using four-channel samplers (TH-16A, Tianhong, China) simultaneously at an urban site Peking University

(PKU) and an upwind rural site at Yufa during the CAREBeijing-2008 (Campaigns of Air Quality REsearch in Beijing and surrounding region) summer field campaign from 16 July to 31 August 2008. The detailed site descriptions are in previous papers (Huang et al., 2006; Guo et al., 2010; Zheng et al., 2011). PKU (39° 59′ 21″ N,

5   116° 18′ 25″ E), the urban site, is located at the campus of Peking University. Yufa, (39° 30′ 49″ N, 116° 18′ 15″ E) the rural site, is 53 km south from the PKU site. During the campaign the wind was mainly from south and southwest, so Yufa was an upwind rural site. Around Yufa site was nothing but farmland. The pollution concentrations at Yufa can be considered as the regional pollution level (Guo et al., 2012).

10   Each set of the samples consists of one Teflon filter and three quartz filters. In Total 74 sets (36 day samples and 38 night samples) of samples at PKU and 99 sets (44 day samples and 45 night samples) of samples at Yufa were collected, respectively.

2.2   Chemical analysis

The Teflon filters were ultrasonically extracted by de-ionized water, and analyzed by

15   ion-chromatograph (Dionex, ICS-2500) to measure major water-soluble ionic compounds. The analysis methods were in the previous paper (Hu et al., 2005). An area of $1.45\,cm^2$ of the quartz filters was punched to analyze EC (elemental carbon) and OC (organic carbon) using a Sunset Laboratory-based carbon analyzer (Birch and Cary, 1996). The remaining two quartz filters were then extracted and analyzed to determine

20   the chemical composition of particulate organic matter. The detailed analytical method for particulate organic species has been described previously (Guo et al., 2012). In brief, 29 kinds of internal standard compounds (including ketopinic acid (KPA), 26 deuterated species and two carbon isotope (^{13}C)) were spiked in the samples, and then the filters were ultrasonically extracted with organic solution and concentrated to

25   0.5–1 mL. About a 200–300 μL extract of each sample was derivatized with BSTFA (BSTFA/TMCS, 99 : 1; Supelco). The derivatized and underivatized fractions were analyzed using an Agilent GC-MS system (6890 plus GC-5973N MSD). In total 164 organic compounds were quantified, including primary and secondary organic tracers.

The names and the concentrations of these particulate organic species are listed in Table S2 in Supplement.

2.3 Source apportionment

A chemical mass balance (EPA, version CMB 8.2) receptor model was employed to apportion the primary sources of OC. Five major primary sources were chosen in the model, and the source profiles were from literature, including vegetative detritus (Rogge et al., 1993), biomass burning (Fine et al., 2004), coal burning (Zheng et al., 2005), non-catalyzed engines and diesel engines (Lough et al., 2007; Schauer et al., 1999, 2002). The tracers that were applied to CMB model are marked in Table S2 in Supplement. Cholesterol, historically used as a tracer for Chinese cooking (Zhao et al., 2007), was not detected in most samples, indicating that cooking source was an insignificant contributor during the study period, so cooking was not considered in the model. However, if cooking or other sources contributed to OC, they would be apportioned as other OC. The criteria for acceptable fitting results included the square regression coefficient of the regression equation $R^2 > 0.85$, the sum of square residual Chi-square value $\chi^2 < 4$, and the percentage of explained mass to total mass 80–120 %. A summary of the statics for CMB fitting results is listed in Table S1 in Supplement.

The contributions of secondary sources were estimated by using tracer-yield method. In this approach, fixed ratios of tracers to OC on the basis of laboratory experiments were used (Kleindienst et al., 2007). Although several uncertainties remained, this method is very useful and the only way to roughly estimate biogenic and anthropogenic SOA (secondary organic aerosol) from specific VOC (volatile organic compound) precursors (Offenberg et al., 2007).

3 Results and discussion

3.1 PM$_{2.5}$ chemical composition of and definition of different periods

Fine particles were reported to be the most important air pollutant of Beijing during the last decade. Table 1 lists the mass concentrations of PM$_{2.5}$ in summer of Beijing (2000–2008). Zheng et al. (Zheng et al., 2005) reported PM$_{2.5}$ as high as 118.3 µg m^{-3} in 2000. The concentrations gradually decreased from 2000 to 2006 (Dan et al., 2004; Song et al., 2007; Zhao et al., 2009; Wang et al., 2009). Due to unfavorable meteorological conditions, PM$_{2.5}$ increased to 91.7 µg m^{-3} in 2007 (Zhao et al., 2009). During the measurement period of our work, PM$_{2.5}$ fluctuated drastically from 8.6–174.4 µg m^{-3} at PKU and 14.3–167.6 µg m^{-3} at Yufa, respectively. The concentrations were the lowest in 2008. Although the fine particle pollution of Beijing decreased significantly, the concentrations were still very high compared with those in other regions of the world.

PM$_{2.5}$ chemical compositions during the measurement are presented in Fig. 1. Sulfate and organics (OC*1.6) were the two most important components in PM$_{2.5}$. Sulfate accounted for the largest fraction with 37.0 % and 37.2 % at PKU and Yufa, respectively. In total, secondary inorganic compounds represented 63.9 % and 65.8 % of the fine particles at two sites, indicating the secondary particles were dominant in Beijing summer. Carbonaceous compounds including organic carbon (OC) and elemental carbon (EC) were also important in fine particles. Organics were the second abundant component in PM$_{2.5}$, accounting for 20.3 % and 22.0 %. EC was from incomplete combustion, responsible for 4.5 % and 4.0 % of PM$_{2.5}$. A comparison of OC and EC concentrations between this study and previous work is listed in Table 2 (He et al., 2001; Zheng et al., 2005; Wang et al., 2009). The variation of OC was not significant from 2000–2006. However EC decreased a lot due to the primary emission control strategies these years. In 2008 both OC and EC were the lowest since 2000 (OC 9.2 ± 3.3 µg m^{-3} at PKU and 8.9 ± 3.1 µg m^{-3} at Yufa; EC 2.6 ± 1.3 µg m^{-3} at PKU and 2.3 ± 1.0 µg m^{-3} at Yufa). In one hand, the drastic control measures reduced primary

emission. On the other hand, the favorable weather conditions also contributed to this pollution alleviation (Huang et al., 2010).

For easier discussion and comparison, the measurement was divided into five periods based on the Olympic Games, control measures (mainly focusing traffic control) and weather conditions. The detailed control measures in different periods are listed in Table S3 in Supplement with 15–20 July defined as non-control period (NC). The "odd/even plate number rule" for vehicles (vehicles with odd and even plates can only be driven alternatively) was not implemented, and there was no precipitation during this period. Two periods were defined as Before Olympic period (21–31 July: BO-I, 1–8 August: BO-II) based on the pollution episode. During both periods, the weather conditions were stagnant until the precipitation process at the end of the each period; during Olympic period (DO) was from 9–24 August. There were several precipitation instances during this period. The "odd/even plate number rules" were implemented during BO-I, BO-II and DO periods throughout the Beijing area. After Olympic period (AO) was 25–31 August. The "odd/even plate number rule" was only implemented in the urban area (inside the 5th Ring Road).

3.2 Particulate organic species in different periods

In total, 164 kinds of particulate organic species were measured, accounting for $6.4 \pm 1.6\%$ and $8.0 \pm 1.3\%$ of the total organic matter (OC*1.6) at PKU and Yufa, respectively. The average concentrations of these species during the measurement are listed in Table S2 in Supplement.

The organic compounds (not including secondary organic tracers) were cataloged into 12 classes, and their concentrations in 5 different periods are shown in Fig. S1 in Supplement. Detailed information for each class is also given in Supplement (Figs. S2–S5). During the measurement, dicarboxylic acid was the most abundant component in all periods except NC period of PKU. Previous studies suggested dicarboxylic acids in Beijing were mainly from secondary formation (Yao et al., 2002; Guo et al., 2010). The abundance of dicarboxylic acids indicated secondary formation was important in

summer of Beijing. *n*-alkanoic acids were the major component during NC period at PKU. Although *n*-alkanoic acids could also be from chemical transformation, vehicle emission was their major source in Beijing (Huang et al., 2005). Thus, more traffic in NC period may be responsible for the high *n*-alkanoic acid concentration. Compared with previous work, most primary compounds such as PAHs (polycyclic aromatic hydrocarbons), hopanes and sugar compounds were much lower in 2008, maybe due to the strict emission control measures for the Olympics.

A series of *n*-alkanes ranging from C_{12}–C_{36} were measured, and their concentrations in different periods are presented in Fig. S2 in Supplement. The *n*-alkanes concentrations at two sites were both the highest in NC period and lowest during the Olympics. The maximum concentration species (C_{max}) were C27 or C29, and an odd carbon preference was found, indicating the biogenic sources (vegetative matter and biomass burning) were important (Zhang et al., 2007; Rogge et al., 1993). However, low carbon number species such as C19, C20 and C21 were also high especially at the urban site. Low carbon predominance index (CPI) was found at both sites (PKU 1.5, Yufa 1.7). These results suggested fossil fuel combustion was also important in Beijing (Schauer et al., 2002, 1999).

PAHs are generally from a variety of combustion and so are not specific tracers. Natural combustion like from a volcano could contribute to PAHs, but the majority was from anthropogenic emissions (Ravindra et al., 2008). In this study, total PAH concentrations in five different periods were $30.28\,\mathrm{ng\,m^{-3}}$, $23.89\,\mathrm{ng\,m^{-3}}$, $20.86\,\mathrm{ng\,m^{-3}}$, $15.71\,\mathrm{ng\,m^{-3}}$, $22.40\,\mathrm{ng\,m^{-3}}$ at PKU and $25.55\,\mathrm{ng\,m^{-3}}$, $25.37\,\mathrm{ng\,m^{-3}}$, $24.89\,\mathrm{ng\,m^{-3}}$, $15.76\,\mathrm{ng\,m^{-3}}$, $26.29\,\mathrm{ng\,m^{-3}}$ at Yufa. High PAHs concentrations were found in NC period at PKU and NC and AO period at Yufa when traffic control was not conducted. Compared with previous study, PAHs were much lower than those before (Wang et al., 2009; Zheng et al., 2005). The distributions of PAHs are exhibited in Supplement (Fig. S3). Five-ring PAHs were dominant at both sites, followed by four-ring and six-ring PAHs. These three kinds of PAHs comprised 60 % and 70 % of the measured PAHs at two sites. This distribution

was consistent with the previous work and was attributed to the high ambient temperature in summer time.

Saccharide is considered to be mainly from biomass burning (Simoneit, 1999). Three sugar compounds were quantified: levoglucosan, manosan and galactosan, among which levoglucosan is considered as the best tracer for biomass burning. During the study period levoglucosan concentrations were $97 \pm 72\,\mu g\,m^{-3}$ and $120 \pm 78\,\mu g\,m^{-3}$ at PKU and Yufa, respectively, which was the lowest in recent years. The higher levoglucosan concentration at Yufa indicated more frequent biomass burning emission at the rural area of Beijing. The highest levoglucosan concentrations were found in NC period, the lowest in DO period at both sites.

Hopanes are not abundant in atmospheric particles, but they can well indicate the vehicle emission (Simoneit, 1986). Three hopanes were measured, whose total concentrations in different periods were $3.97\,ng\,m^{-3}$, $1.85\,ng\,m^{-3}$, $1.97\,ng\,m^{-3}$, $1.34\,ng\,m^{-3}$, $2.00\,ng\,m^{-3}$ at PKU and $2.59\,ng\,m^{-3}$, $2.63\,ng\,m^{-3}$, $2.41\,ng\,m^{-3}$, $1.85\,ng\,m^{-3}$, $2.83\,ng\,m^{-3}$ at Yufa (see Fig. S5). At PKU hopanes were much lower in control periods than those in the non-control period due to the traffic control. The reduction at Yufa was not as significant as that in the urban area. Nevertheless the hopane concentrations were also lower in control period, suggesting the traffic control improved the regional air quality.

Detailed discussion about SOA tracers has taken place in previous work (Guo et al., 2012). In brief, high concentrations of SOA tracers were found at both sites because of the high regional SOC (secondary organic carbon) background ($\sim 2\,\mu g\,C\,m^{-3}$). Biogenic SOA tracers were higher at a rural site due to higher precursor concentrations. Anthropogenic SOA tracers were comparable between urban and rural sites, implying the anthropogenic particle pollution was widespread in large scale of Beijing and affected regional air pollution.

3.3 Source apportionment of ambient organic carbon

Primary and secondary organic tracers were applied in CMB model and tracer-yield method to estimate the contributions of primary and secondary particulate organic sources to total OC. Five primary sources and four SOCs derived from isoprene, α-pinene, β-caryophyllene were apportioned, accounting for $73.8 \pm 9.7\%$ and $79.6 \pm 10.1\%$ of the measured OC at PKU and Yufa, respectively. The results are listed in Table 3, compared with those from previous source apportionment studies in Beijing (Wang et al., 2009; Zheng et al., 2005).

Vehicle emission was a very important source of fine organic particles in Beijing. It was reported that the number of vehicles in Beijing has increased 20–25 % per year (China Statistical Yearbook 2011, http://www.stats.gov.cn/tjsj/ndsj/2011/indexeh.htm). The increasing vehicle amount has led to serious fine particle pollution in Beijing (Chan and Yao, 2008). In our work, diesel and gasoline vehicle emissions were the major contributors to ambient organic particles with the contributions of $16.2 \pm 5.9\%$ and $14.5 \pm 4.1\%$ by diesel engines and $10.3 \pm 8.7\%$ and $7.9 \pm 6.2\%$ by gasoline vehicles at PKU and Yufa, respectively. The higher contributions at PKU were reasonable, because there were more vehicles in the urban area. Compared with previous work, vehicle emission contributed more in 2008 than previous years. However, considering the OC concentrations, the absolute concentrations attributing to vehicle emission were lowest in 2008. A diesel engine contribution reduced more significantly than gasoline engines. On one hand, gasoline cars were more constrained during the Olympics. On the other hand, the number of public vehicles like buses increased during the Olympics, and these vehicles mainly used diesel engines.

Biomass burning was also an important source for ambient particles in Beijing. Although open biomass burning is strictly forbidden in Beijing, and its contribution to organic particles could be considered to be zero (Cao et al., 2005), the open fire from nearby province could still influence the air quality of Beijing. Moreover, wood consumed as domestic fuel in rural areas also greatly contributed to ambient fine particles.

In our work biomass burning contributed $7.5\pm5.5\,\%$ and $8.9\pm4.9\,\%$ to measured OC at PKU and Yufa. The higher contribution at Yufa was mainly due to more domestic wood use and transport from surrounding area. The biomass burning contribution was lower than those in the year 2005 and 2006.

Coal is the dominant fuel in Beijing. About 68.7 % of the total energy consumption of China in 2008 was coal (China Statistical Yearbook 2008, http://www.stats.gov.cn/tjsj/ndsj/2008/indexeh.htm). In our work, coal burning accounted for $5.8\pm5.5\,\%$ and $7.8\pm4.6\,\%$ of the measured OC. This result was comparable with the result in 2000, but higher than that reported in 2005 and 2006. Because the total OC was much lower in 2008, the absolute coal burning OC was the lowest in recent years. The alleviation of coal burning pollution was mainly due to the more strict restriction and some emission reductions such as temporary closures of factories. However, the rural area was still affected significantly by coal combustion. Although coal cannot be used within the central urban area of Beijing, large amount of coal was still consumed in rural area of Beijing. In addition, the most common coal usage in rural area was residential coal, which has higher emission factors (Zhang et al., 2008). Some emission control measures were also implemented in nearby province, but the measures were much less strict. The industry and other coal usage in surroundings of Beijing also had great contribution to the organic particles of Beijing.

Because there was no previous study on SOCs derived from specific VOCs, no comparison can be made to investigate their variations in recent years. However, unapportioned OC by CMB model was likely mainly comprised of secondary source particles, so their changes can be to an extent to estimate the variation of the particle secondary sources. In this study, $60.0\pm13.2\,\%$ and $59.0\pm11.0\,\%$ of the OC cannot be apportioned by CMB, which was considered mainly to consist of secondary organic carbon, and this fraction was the largest in 2008, suggesting SOC has become an important contributor to organic particles.

3.4 Variation of organic particle sources in different periods

In order to improve air quality for the Olympics, Beijing municipal government imple-
mented a series of measures to alleviate the particle pollution in Beijing. It provided
a good opportunity to understand the impact of reduced emissions on air pollution.
The OC concentrations from major primary and secondary sources and their contribu-
tions to total ambient organic carbon are shown in Fig. 2 and Table 4. To evaluate the
emission control effectiveness, F-tests were first used between every two periods to
test whether the two periods had significant difference (significance level $\alpha = 5\%$), and
then the mass concentrations of specific sources, as well as their contributions to total
OC, during different periods were compared to quantitatively estimate the variation of
different sources.

Vehicles were the major controlled emission during the Olympics. The detailed con-
trol measures are listed in Table S1 in Supplement. At the urban site PKU, vehicle
emission was the major source of organic particles during non-control period, con-
tributing $1.69 \pm 0.42 \, \mu g \, m^3$ by diesel vehicles and $1.54 \pm 1.21 \, \mu g \, m^3$ by gasoline vehicles.
Their contributions decreased considerably during the next four controlled periods. An
F-test result showed the OC concentration attributed by vehicles in NC period was
significantly higher than those in the other controlled periods. The OC concentrations
from gasoline vehicle decreased significantly, reduced by 58 %, 47 %, 62 % and 44 %
compared with the NC period, with the overall average reduction of 54 %. The OC
concentration from diesel vehicles also decreased but not as significantly as that from
gasoline vehicles. The OC concentrations were reduced by 4 %, 7 %, 31 % and 28 %
during the four controlled periods. The average reduction was 13 %. The reduction of
absolute concentrations was also affected by weather conditions, so we cannot better
evaluate the effectiveness of the control measures. Exploring the variations of source
contributions (percentages of specific sources to total OC) is a better way to reflect
the impact of the emission control measures. During the NC period, gasoline vehicle
emission contributed 15.4 % of the measured OC. The contribution was reduced by

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61 %, 48 %, 49 % and 36 % during the BO-I, BO-II, AO and AO periods. Compared between non-control and the whole control period, the average reduction was 52 %. Correspondingly, the contributions of diesel vehicle emission were reduced by 11 %. All in all, the contributions of vehicle emissions were reduced by 30 % on average. Although official estimation of the traffic reduction was about 60 %, there were no exact measured data. However, our result was consistent with the statistical model result about total $PM_{2.5}$ reduction from Liu et al. (2012).

Coal combustion OC also decreased during the four control periods with the reduction of 57 %, 66 %, 65 % and 39 % by concentration and 61 %, 66 %, 55 % and 30 % by contribution (ratio of specific source to total OC). The average reduction was 59 % and 57 % in concentration and contribution, respectively. The contributions of biomass burning and vegetative detritus showed no obvious differences between control and non-control periods (F-test result), indicating these two sources did not have great changes. The results in this study indicated the control measures have significantly improved the urban air quality of Beijing.

Yufa was an upwind rural site and a regional site. The variation in source contributions at Yufa can evaluate the control effectiveness at the regional scale of Beijing. Vehicle emission was the major source of organic particles at Yufa, and their contributions decreased during traffic control periods. Although the reduction was not as much as that at PKU, the contributions of gasoline vehicle emission also decreased considerably by 45 %, 62 %, 55 % and 54 % in four control periods, and the diesel vehicle contribution was reduced by 10 %, 27 %, 20 % and 14 %. The average reduction of the contribution was 39 % for gasoline vehicles and 14 % for diesel vehicle. The contribution of two kinds of vehicle emission was on average reduced by 24 %. Particle pollution from coal burning was also cut down by 17 % in concentration and by 7 % in contribution to total OC. The reduction of coal burning pollution was not as significant as that in the urban area, maybe because Yufa was more affected by the emissions from neighboring provinces. Nevertheless, we can still conclude that the control measures greatly alleviated the regional primary organic particle pollution.

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Secondary formation processes including nucleation, gas-to-particle transformation and multiphase reaction are still poorly understood (Zhang et al., 2012; Hallquist et al., 2009; Zhang, 2010). The SOA formation is very complicated, and can be affected by various factors such as precursors, ambient temperature, humidity and particle acidity (Wang et al., 2010b; Zhao et al., 2006; Saathoff et al., 2009). In this work, SOCs derived from isoprene, α-pinene, β-caryophyllene and toluene were apportioned, and their concentrations and contributions to measured OC in different periods are presented in Table 4 and Fig. 2. Three major SOCs from toluene, isoprene and α-pinene showed similar trends at PKU and Yufa. The highest SOC concentrations appeared during the BO-I period at PKU and BO-II period at Yufa when the conditions were favorable for SOA formation and accumulation: high radiation, temperature, humidity, O_3 concentration and no precipitation.

At the urban site PKU, the contributions of apportioned SOCs to measured OC during the five periods were 35.8 %, 38.4 %, 31.0 %, 33.4 % and 27.9 %. An F-test result showed that the anthropogenic SOC contributions did not have significant differences between controlled and non-controlled period. However, the biogenic SOC contributions were a little higher during controlled period. Because biogenic VOC emission cannot be controlled, the biogenic SOCs were considered to be affected mainly by weather conditions. The ratio of anthropogenic to biogenic SOC can simply explore whether the control measures had impact on anthropogenic SOC formation. This ratio was higher in non-control period, implying the emission control to a certain extent constrained the anthropogenic SOC formation in the urban area. Wang et al. (2010a) reported the toluene mixing ratio decreased during the traffic control period. The lower precursor concentrations may lead to the lower anthropogenic SOC concentration.

At rural site Yufa, SOCs contributed 27.4 %, 34.7 %, 45.3 %, 39.1 % and 32.7 % to measured OC. Both concentrations of biogenic and anthropogenic SOCs were higher in the controlled period. The anthropogenic to biogenic SOC ratios did not have significant difference between control and non-control period. In conclusion, emission

control to an extent constrained the anthropogenic SOC formation but not significantly. Weather conditions were still the major influencing factors on SOC formation.

4   Conclusions

PM$_{2.5}$ and carbonaceous components during 2008 Beijing Olympic Games were the
lowest in the recent years. The improvement of air quality was mainly due to the drastic emission control measures and also favorable weather conditions. Five major primary sources and four SOCs were identified by CMB model and tracer-yield method. Compared with the previous source apportionment work, vehicle emission has become the major source of organic particles these years, at both urban and regional scale.
However, due to the emission control measures, the absolute concentrations from vehicles were gradually reduced, especially in 2008 when more strict measures were implemented. For the same reason, coal combustion and biomass burning OC both decreased and were the lowest in 2008. Organic particles transported from nearby province also contributed organic particles in rural Beijing. Secondary formation be-
came a significant contributor to ambient fine organic particles in Beijing. Nevertheless the absolute SOC concentrations also decreased compared with those in previous years.

The effectiveness of emission control measures was evaluated. The vehicle contributions were reduced by 30 % in the urban area and 24 % at a regional scale. Contribution
from coal combustion was also reduced by 57 % in the urban area and 7 % at regional scale. All these results suggested the emission control measures significantly alleviated the organic particle pollution in and around Beijing. The anthropogenic SOC formation was to a certain extent constrained due to the reduction of precursor concentrations. However, the SOC control effectiveness was not significant. Weather conditions were
the major influencing factor on SOC formation. To improve the regional air quality, especially to reduce the SOC pollution, more strict control measures should be implemented at a larger regional scale in the future.

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Table 1. PM$_{2.5}$ mass concentrations in summer of Beijing 2000–2008, μg m^{-3}.

Year/Month	2000/07	2001/07–08	2002/06–07	2003/06	2004/08	2005/06–08	2006/08	2007/06–08	2008/07–08	2008/07–08[a]
N[b]	5	8	10	3	5	-	29	-	74	99
Concentration	118.3	104.1 ± 45.1	99.4 ± 33.9	80.2 ± 80.2	100.4	84.5[c]	83.3	91.7[c]	72.5 ± 43.6	64.3 ± 36.2
Literature	Zheng et al. (2005)	Dan et al. (2004)	Dan et al. (2004)	Dan et al. (2004)	Song et al. (2007)	Zhao et al. (2009)	Wang et al. (2009)	Zhao et al. (2009)	This study	This study

[a] Rural site Yufa; all other measurements were conducted at urban site.
[b] Number of samples.
[c] PM$_{2.5}$ was measured by online instrument tapered element oscillating microbalance (TEOM).

Table 2. Concentrations of organic carbon (OC) and elemental carbon (EC) in $PM_{2.5}$, $\mu g\,m^3$.

Year/Month	1999/07	2000/07	2005/08	2006/08	2008/07–08[a]	2008/07–08[b]
N[c]	12	5	46	43	74	99
OC	6.27	20.9	16.1	22.1	9.2 ± 3.3	8.9 ± 3.1
EC	9.4	3.0	3.2	5.5	2.6 ± 1.3	2.3 ± 1.0
Reference	He et al. (2001)	Zheng et al. (2005)	Wang et al. (2009)	Wang et al. (2009)	This study	This study

[a] PKU site.
[b] Yufa site.
[c] Number of samples.

Table 3. Comparison of source contributions to fine particulate organic carbon in summer of Beijing (%).

| Year/Site	2000	2005	2006	2008	2008
	PKU	PKU	PKU	PKU	Yufa
Gasoline vehicles	19.0	10.6 ± 2.8	7.1 ± 1.5	10.3 ± 8.7	7.9 ± 6.2
Diesel vehicles		9.6 ± 6.2	5.7 ± 1.5	16.2 ± 5.9	14.5 ± 4.1
Vegetative detritus	1.5	0.3 ± 0.2	0.3 ± 0.2	1.4 ± 0.8	1.9 ± 1.5
Dust	25.6				
Wood burning		10.4 ± 3.5	8.2 ± 2.0	7.5 ± 5.5	8.9 ± 4.9
Straw burning		1.3 ± 0.7	1.7 ± 2.8		
Coal burning	5.8	2.1 ± 2.2	1.8 ± 0.8	5.8 ± 5.5	7.8 ± 4.6
Cigarettes	2.2				
Cooking		24.5 ± 4.2	23.8 ± 4.3		
Other OC	45.9	41.1 ± 9.7	51.4 ± 5.6	60.7 ± 13.2	58.8 ± 11.0
Reference	Zheng et al. (2005)	Wang et al. (2009)		This study	

Table 4. The organic carbon concentrations from major primary and secondary sources during different periods ($\mu g\,m^{-3}$).

	NC PKU	BO-I	BO-II	DO	AO	NC Yufa	BO-I	BO-II	DO	AO
Isoprene SOC	0.82	1.26	1.04	0.67	0.57	1.42	1.46	1.86	1.00	0.95
α-pinene SOC	0.67	0.67	0.39	0.38	0.36	0.45	0.49	0.69	0.50	0.44
Toluene SOC	1.66	2.05	1.61	1.34	1.29	0.99	1.43	2.24	1.55	1.52
β-caryophyllene SOC	0.42	0.33	0.08	0.16	0.19	0.09	0.11	0.21	0.18	0.19
Vegetative detritus	0.21	0.13	0.16	0.08	0.12	0.27	0.18	0.18	0.16	0.24
Biomass burning	0.67	0.87	0.65	0.49	0.59	0.79	0.73	0.93	0.89	0.90
Diesel vehicles	1.69	1.63	1.57	1.17	1.22	1.76	1.54	1.25	1.12	1.38
Gasoline vehicles	1.54	0.65	0.81	0.59	0.86	1.23	0.84	0.60	0.57	0.67
Coal burning	1.08	0.46	0.37	0.38	0.66	0.92	0.82	0.78	0.59	0.92

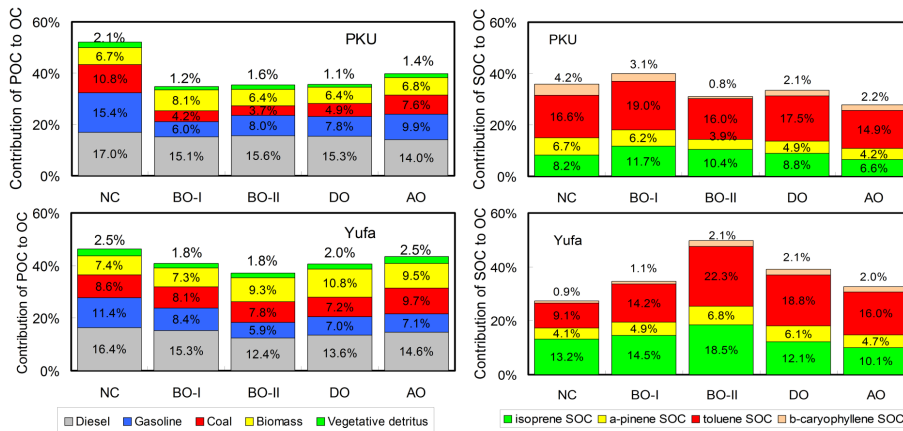Note: NC = Non-Control, BO = Before Olympics, DO = During Olympics, AO = After Olympics.
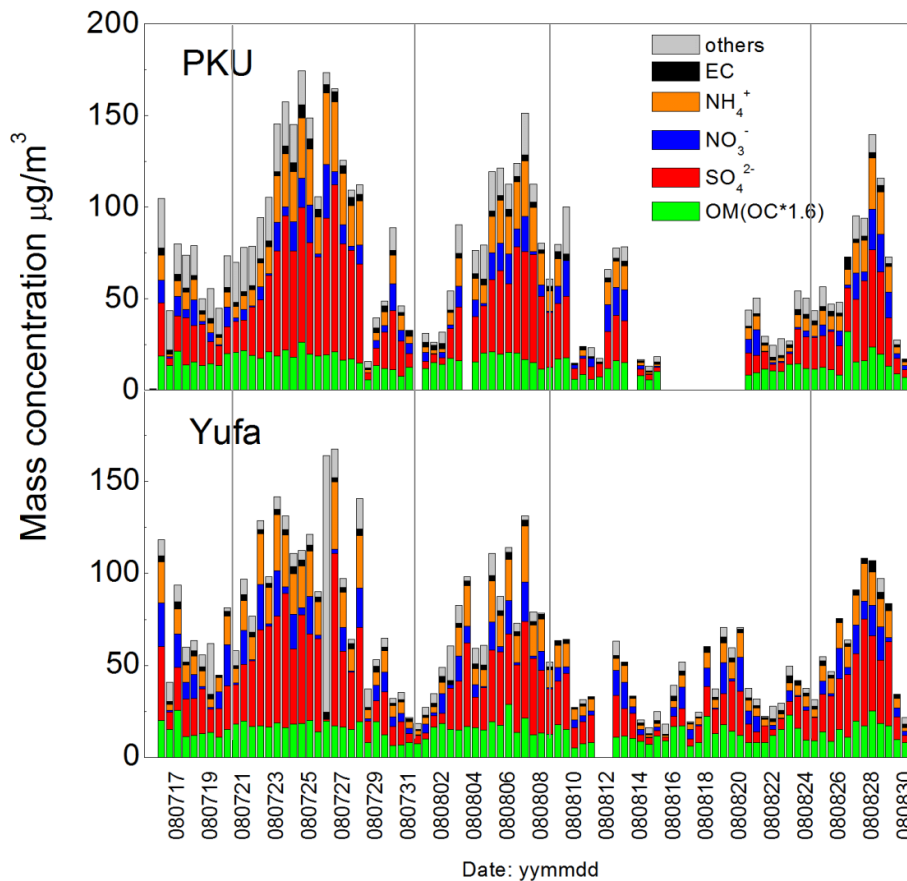
Fig. 1. Chemical composition of PM$_{2.5}$ at an urban site PKU and an upwind rural site Yufa site.

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Fig. 2. Contributions of primary and secondary sources to ambient organic carbon in different periods. NC = Non-Control, BO = Before Olympics, DO = During Olympics, AO = After Olympics.