Atmos. Chem. Phys. Discuss., 13, 5165–5197, 2013
www.atmos-chem-phys-discuss.net/13/5165/2013/
doi:10.5194/acpd-13-5165-2013

Atmospheric Chemistry and Physics Discussions

Long-term measurements of particle number size distributions and the relationships with air mass history and source apportionment in the summer of Beijing

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Received: 31 December 2012 – Accepted: 4 February 2013 – Published: 22 February 2013

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

A series of long-term and temporary measurements were conducted to study the improvement of air quality in Beijing during Olympic Games period (8–24 August 2008). To evaluate actions taken to improve the air quality, comparisons of particle number and volume size distributions of August 2008 and 2004–2007 were performed. The total particle number and volume concentrations were 14 000 cm^{-3} and 37 μm^3 cm^{-3} in August of 2008, respectively. These were reductions of 41 % and 35 % compared with the mean values of August 2004–2007. A cluster analysis on air mass history and source apportionment were performed, exploring reasons of the reduction of particle concentrations. Back trajectories were classified into five major clusters. Air mass from south direction are always associated with pollution events during the summertime of Beijing. In August 2008, the frequency of air mass arriving from south has been twice higher compared to the average of the previous years, these southerly air masses did however not result in elevated particle volume concentrations in Beijing. This result implied that the air mass history was not the key factor, explaining reduced particle number and volume concentrations during the Beijing 2008 Olympic Games. Four factors were found influencing particle concentrations using a Positive matrix factorization (PMF) model. They were identified to local and remote traffic emissions, combustion sources as well as secondary transformation. The reductions of the four sources were calculated to 47 %, 44 %, 43 % and 30 %, respectively. The significant reductions of particle number and volume concentrations may attribute to actions taken, focusing on primary emissions, especially related to the traffic and combustion sources.

1 Introduction

The effects of aerosol particles on ecology, climate, and health-related issues are the central topics in the current environmental research. Particle mass concentration attracts more attention due to its negative impact on the air quality and urban visibility

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(Molina and Molina, 2004). Recent evidences indicate that the number concentration of ultrafine particles should get equal or even more attention than the particle mass concentration in terms of health effects (Oberdorster et al., 2005). Moreover, not only the particle number concentration is important, but also the particle number size distribution (Kumar et al., 2010). However, air quality standards (such as $PM_{2.5}$ and PM_{10}) are developed to control the particle mass concentration. The mass of ultrafine particles ($D_p < 100$ nm), which dominate the particle number concentration are negligible. The current air quality standards may thus be insufficient to draw a whole picture on the aerosol impacts, especially to ecology and health effects. Measurements of particle number distributions are one piece of the entire mosaic to understand better all kind of impacts of atmospheric aerosol particles.

Measurements of particle number size distributions have been conducted in diverse environments (Kulmala et al., 2004; Holmes, 2007). However, long-term measurements are mainly performed in Europe and North America (Kulmala et al., 2004; Asmi et al., 2011). Although a few intensive campaigns had been conducted in the heavy polluted environments such as Mexico (Dunn et al., 2004), New Delhi (Monkkonen et al., 2005) and the Pearl River Delta region (Liu et al., 2008; Yue et al., 2010), long-term measurements of particle number size distributions in developing countries were only carried out in urban Beijing since 2004 (Wehner et al., 2004) and in the North China Plain since 2008 (Shen et al., 2011). Previous results show that particle number concentrations especially in the accumulation mode range are much higher than in the cities of developed countries (Wehner et al., 2008; Wu et al., 2008).

With the rapid urbanization and motorization, Beijing is one of the biggest megacities in the world with a population of more than 15 million people and over 3.5 million vehicles up to 2008 (http://www.stats.gov.cn/). The air quality problem is characterized by high concentrations of fine particles and ozone in Beijing (Shao et al., 2006; Streets et al., 2007). As the host city of 29th Olympic Games, "Green Olympics" was proposed as one of the three themes. A series of control measures were conducted by the government gradually. The air quality in August 2008 had been the best for any

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summer periods over the last few years (UNEP, 2009). This achievement attributed to painstaking efforts in industrial restructuring and application of environment protection technologies since 1998 as well as temporary measures taken during Olympic Game periods, especially revolved around vehicles (Hao et al., 2006; Zhou et al., 2010). Many estimation results focusing on the air quality during the 2008 Olympic Games period have been reported (M. Wang et al., 2009; Wang and Xie, 2009; W. T. Wang et al., 2009; X. Wang et al., 2009; Y. Wang et al., 2009; Wang et al., 2010; Zhou et al., 2010). The model results showed that the average reductions of PM_{10}, CO and NO_2 were 28 %, 19.3 %, 12.3 %, respectively, in the case of a 32.3 % reduction of the traffic flow (Wang and Xie, 2009). A mobile laboratory study revealed that the on-road air pollutant concentrations decreased 54 % for CO, 41 % for NO_x and 12 % for Black carbon (BC) during the Olympics compared with the pre-control time period (Wang, M. et al., 2009). However, these studies only concern the variations of pollutants (such as pollutant gases and particle mass concentration) before and during the Olympic Games. Few studies were taken focusing on long-term measurements and none information about the particle number size distribution is published yet.

This investigation was based on the long-term measurements of particle number size distributions during five consecutive August from 2004 to 2008. Relationships between particle number size distributions and air mass origin or source apportionment were explored in order to answer the following questions: (1) how can one evaluate the air quality during the 2008 Olympic Games based on the particle number size distributions; (2) which actions were most responsible for the reductions of particle number and volume concentrations.

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2 Experimental

2.1 Sampling site

The sampling site was located on the 6th floor of an academic building (about 20 m above the ground level) on the campus of the Peking University (PKU; 39.99° N, 116.31° E), in the northwestern urban area of Beijing, outside the fourth-ring road. PKU site is assumed as representative for the Beijing urban background aerosol. Detailed descriptions of the measurement site we refer the reader to Wu et al. (2007).

2.2 Instrumentation

Number size distributions of atmospheric particles have been measured by a TDMPS (Twin Differential Mobility Particle Sizer) system with 10 min time resolution at PKU site since March 2004. The measured size range is 3–800 nm (mobility diameter) during 2004 to 2006 and 3–900 nm in 2007–2008. The system is composed of two Hauke-type Differential Mobility Analyzers (DMA) and two Condensation Particle Counters (CPC, model 3010 and 3025, TSI Inc., St. Paul, MN, USA). The relative humidity within the systems was kept below 30 % by adding a silica-gel dryer in the inlet line and in the sheath air cycle to avoid condensation of water in the inlet systems during humid days, especially in summertime. The particle number size distributions were corrected for particle losses inside the TDMPS and in the sampling, following the method of "equivalent length" as described in Wiedensohler et al. (2012). Particle volume concentrations were calculated from the measured number size distributions, assuming of spherical particles. Considering that the 2008 summer Olympic Games were held from 8 to 24 August in 2008, we choose only data of measurements taken in August from 2004 to 2008 in this investigation.

To evaluate the air quality during the Olympic Games, an intensive campaign called CAREBeijing 2008 was conducted from beginning of July to the end of September. The PM_1 particle mass concentration and chemical components measurements were

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carried out using an Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToFAMS). Detailed instrumentation description of HR-ToFAMS could be found in Huang et al. (2010). BC concentrations were measured using a Multi-Angle Absorption Photometer (MAAP, Thermo Model 5012).

Meteorological conditions including air temperature (T), relative humidity (RH), wind speed (WS) and precipitation were measured at a meteorological station in 200 m distance from the sampling site.

2.3 Methods

2.3.1 Back trajectory analysis

To investigate the influence of the air mass history and to obtain a better understanding on regional atmospheric aerosol properties, a back trajectory analysis was performed using the HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT4) model developed by NOAA/ARL to follow the history of air masses arriving at measurement site (Draxler and Rolph, 2012; Rolph, 2012). The meteorological input data used in the model was obtained from the NOAA ARL archives (http://ready.arl.noaa.gov/archives.php). In this investigation, 48 hours back trajectories were chosen and four trajectories per day (00:00, 06:00, 12:00, and 18:00) were calculated. The trajectories terminated on a height of 500 m above ground level. In addition, a k-means clustering algorithm was applied to group in total 620 backward trajectories (five years August) into different transport patterns according to their similarity in spatial distribution using the HYSPLIT4 software.

2.3.2 Source apportionment

Positive matrix factorization (PMF) is a powerful multivariate factor analysis tool to decompose the observed data into two matrices: factor contributions and factor profiles (Paatero and Tapper, 1994). In this study, the source apportionment is achieved using

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the EPA PMF 3.0 version (EPA, 2008). A specific data set was viewed as a data matrix **X** of *i* by *j* dimensions, in which *i* number of samples and *j* chemical species. The basic source-receptor model in matrix form can be simply presented as:

$$\mathbf{X}_{ij} = \sum_{k=1}^{p} \mathbf{G}_{ik}\mathbf{F}_{kj} + \mathbf{E}_{ij} \quad (1)$$

Here **X** is the matrix of observed particle number size distributions, **G** and **F** are the source contribution and size distribution profile of the source, respectively, and **E** is a matrix of residuals.

To evaluate the reductions of diverse sources, we assumed that the source profile was consistent in August from 2004 to 2008. It appears that some of the sampling days have nucleation and subsequent growth occurring. The mean frequency of new particle formation event was 10 % in August from 2004 to 2008. The basic assumption of the receptor model is that the ambient data is the sum of constant particle number size distribution profiles from the contributing sources (Zhou et al., 2004). Considering that nucleation is a strong source of newly formed particles, the days with intense nucleation events were thus excluded for this study. These day were classified as described in Wu et al. (2007). Overall, the whole data set includes 161 700 individual particle number size distributions.

Measurement uncertainty is one input parameter for the PMF analysis. In this study, an equation-based method was taken to calculate the measurement uncertainty. Two parameters including the method detection limit (MDL) and error fraction are involved into the equation-based method (EPA, 2008). The detection limit of the TDMPS was calculated by particle number concentration divided by raw counts of CPC for each size bins. The mean result is shown in the Fig. 1. Error sources for the measurements of particle number size distribution include the aerosol flow rate, the DMA, CPC counting efficiency and the correction for losses as described above. Considering these error sources, the error fraction was estimated as 15 (the percent uncertainty multiplied by 100) for particles smaller than 25 nm and 10 for particles larger than 25 nm. If the

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number concentration is less than or equal to the MDL provided, the uncertainty (Unc) was calculated by the following equation:

$$\text{Unc} = \frac{5}{6} \times \text{MDL} \tag{2}$$

If the particle number concentration is greater than the MDL provided, the uncertainty is calculated:

$$\text{Unc} = \sqrt{(\text{error fraction} \times \text{concentration})^2 + \text{MDL}^2} \tag{3}$$

Different numbers of factors and F_{peak} values have been explored to obtain the most meaningful results. F_{peak} is a parameter to control the rotation in PMF model by changing (adding or subtracting) the rows and columns of **F** and **G** matrices from each other depending on the sign of the F_{peak} value (Paatero et al., 2002). The PMF was run several times for the F_{peak} values between −2 to 2 in steps of 0.2. The Q-values are an assessment of how well the model fit the input data and calculated using Equation:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{\mathbf{X}_{ij} - \sum_{k=1}^{p} \mathbf{G}_{ik}\mathbf{F}_{kj}}{U_{ij}} \right] \tag{4}$$

with U as the uncertainty. The Q-value versus F_{peak} plot show a typical "U" shape with the lowest Q-value corresponding to F_{peak} of 0, which is selected in this study. The theoretical Q (Q_{exp}) is estimated by using the equation: $nm - p(n+m)$, where n is the bins number of the profile of the particle number size distribution, m is the number of samples in the data set, and p is the number of factors fitted by the model. If the assumption of the source profile and the estimation of the errors in the input data are accurate, solutions with numbers of factors that give Q/Q_{exp} near 1 should be obtained (Ulbrich et al., 2009). The value of Q/Q_{exp} is close to 1 when the factor number is chosen as 4 in this study.

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3 Results and discussion

3.1 General overview

The influence of the local meteorology on the air quality cannot be ignored. The meteorological condition can be characterized by high temperatures, high humidity, and low wind speed in Beijing in August (Wu et al., 2008). The statistics of various meteorological parameters in August from 2004 to 2008 are listed in Table 1. The highest mean temperature (28.2°) and relative humidity (71 %) were observed in 2007 and 2006, respectively. The average wind speeds (WS) were around 1 $m\,s^{-1}$ and the dominate wind was from north and northwest in five summers. Overall, the meteorological conditions did not show significant changes during the August periods from 2004 to 2008. The lowest total precipitation amount was 56 mm in 2004 and highest with 150 mm in 2008. However, the precipitation frequency was almost the same in August of these five years, indicating heavier rainfall events in August 2008, Precipitation is an important way for the removal of atmospheric aerosol particles. Previous studies (Gao et al., 2011; Zhang et al., 2012) indicated the important roles of atmospheric circulation and precipitation on the air quality during the Beijing 2008 Olympic Games. Hence, to avoid the influence of the precipitation, particle number size distributions during rainfall events were also excluded from the data set for further analysis.

Figure 2 displays variations of number and volume size distributions of aerosol particles as mean for August periods 2004–2007 and 2008. Peaks of the particle number and volume size distribution are around 50 nm and 450 nm, respectively. The lowest total particle (3–800 nm) number and volume concentrations were observed in August of 2008, with mean values of 14 000 cm^{-3} and 37 $\mu m^3\,cm^{-3}$, respectively. To obtain a more logical view on the reduction values of 2008, the comparisons between the average value of 2004–2007 (Ave. 2004–2007) and 2008 (Ave. 2008) are investigated in this study. Mode-related particle number and volume concentrations are listed in Table 2. Here, we defined the nucleation, Aitken and accumulation modes to the particle size ranges of 3–20 nm, 20–100 nm and 100–800 nm, respectively, following the

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two-year statistical analysis in previous study (Wu et al., 2008). We found significant decreases of number concentrations in the diverse modes. During the August 2008 period, the average number concentrations of nucleation, Aitken and accumulation mode particles were 2800 cm^{-3}, 7400 cm^{-3} and 3800 cm^{-3}, respectively, corresponding to the reduction for 46 %, 42 % and 40 % compared to the mean values of the August periods 2004–2007. The traffic emission is a major source in the urban environment of Beijing (Zheng et al., 2005; Song et al., 2006). Previous study indicated that Aitken mode particles correlate well with the traffic densities (Wu et al., 2008), however, the vehicle population increased from 2.3 million in 2004 to 3.5 million up to 2008 (http://www.bjstats.gov.cn/). The significant increase in vehicle population does however not result in an enhancement in particle number concentration in Aitken mode. We attribute this result to the temporal measures taken during the Olympic Games period: about half of the private vehicles were not allowed on the road during the summertime in 2008. To reduce the local combustion sources, companies (e.g. Beijing Steel Corporation) were forced to move to neighboring provinces prior the Olympic Games period (UNEP, 2009). As a result, the mean volume concentration in August 2008 was only 64 % compared to the average of the August 2004 to 2007 periods.

3.2 Back trajectory cluster analysis

The ground-based measurements in the summer of 2006 proved that the air pollution in Beijing is mainly a regional problem (Takegawa et al., 2009; Guo et al., 2010). The air quality of Beijing was determined by the source emissions from both the megacity and the provinces around depending on the meteorological air mass history. Hence, to explore the impacts of the air mass history on the reduction of particle number and volume concentrations in 2008, the HYSPLIT4 model was used to follow the history of air masses arriving at Beijing.

The five-cluster are the optimum solution according to the change in total spatial variance, with the mean back trajectory of each cluster presented in Fig. 3a. Each of the clusters represents between 75 and 250 back trajectories. The mean back trajectories

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show significant differences in direction and length. Three short length trajectory clusters 2, 3 and 4 arrived from southern and eastern sections, while the other two from northerly direction. The air masses from east (39 %) and south (20 %) direction are the dominate trajectories for August periods. Figure 4 exhibits the median particle number size distributions for five trajectory clusters from 2004 to 2007 and 2008. The particle number size distributions of different air mass types mostly peak around 40–200 nm, whereas the maximum of particle volume size distributions are at around 400–600 nm. The mean total particle number concentrations from northerly directions (NW and NE) were 31 200 cm^{-3} and 21 700 cm^{-3}, respectively, which are higher than the air masses come from other directions (Table 3). On the contrary, more volume concentrations were observed when the air masses from southwest and south, with the mean values of 80 $\mu m^3\,cm^{-3}$ and 70 $\mu m^3\,cm^{-3}$, respectively. This results consistent with the previous study that the slow southern wind controlled by high pressure systems always indicating the polluted situation for Beijing case (Wehner et al., 2008). However, the frequency of air mass from southern direction is 37 % in 2008, which is more than a factor of two compared with the mean frequency (16 %) in 2004–2007 (see Fig. 3b). Although, the air mass condition in 2008 was not favorable to reduce of particle concentration, still, the lowest particle volume concentration had been observed (Fig. 2b). This phenomenon indicates that the air mass condition was not the key factor, explaining the reductions during the Olympic Games 2008. Furthermore, no matter from which direction the air mass arrived, the lowest particle number and volume concentrations were observed in 2008. This implies that the reduction of the pollutants can be attributed to the abatement measures taken in Beijing and neighboring provinces during this period. The reductions of total particle number and volume concentrations calculated as the difference between the 2008 value and the mean of 2004–2007 are listed in Table 3. The reductions of particle number and volume concentrations varied from 24 % to 46 %, 23 % to 56 % for the five clusters, respectively. Highest reductions of total particle number concentrations (46 %) were observed in both the cluster 1 and the cluster 5. The reduction of total particle volume concentrations are more than 40 %

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in all clusters except cluster 3. This might be caused by the high frequency of trajectory cluster 3 in 2008.

3.3 Source apportionment

The results pointed out so far that the improved air quality in August 2008 could be more likely ascribed to the reductions of source emissions rather than the influence of different meteorological condition such as air mass history. Therefore, we tried to identify the sources based on particle number size distribution profiles using a PMF model and get detailed information on the variations of source emissions.

The mean modeled profiles of particle number size distribution for each factor are presented in Fig. 5a. The four factors are arranged in order of increasing peak diameter from factor 1 to factor 4. The modeled particle number size distribution is the sum of four factors. The differences between modeled and and observed values vary from 82 % (7 nm) to 99 % (120 nm). In total, the modeled particle number concentration could explain 96 % of the observation data. Source contributions of factors combined with gaseous and particle chemical components information could help us to identify the sources of four factors. However, we only have the complete data set during the CAREBeijing2008 intensive campaign. Hence, we choose here the data set in 2008 as an example to identify the major source of each factor.

The number size distribution of factor 1 has its peak diameter 16 nm as shown in Fig. 5a. Although this factor has only little contribution to the volume concentration (1 %), it represents however 25 % of the number concentration (see Fig. 5b and c). The shape of this factor profile is similar to the shape of particle number size distributions measured at a roadside (Wehner et al., 2002; Gramotnev and Ristovski, 2004; Virtanen et al., 2006), indicating its relation to car traffic. The strong diurnal variation shows the obvious peaking during the morning and evening rush hour, which is similar to the diurnal variation of NO_x, suggesting that factor 1 is likely to be particles nucleation induced by traffic emissions (Fig. 6). The pattern of this factor in Beijing is similar to the

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local traffic factor founding in the previous studies (Zhou et al., 2004, 2005; Yue et al., 2008).

The particle number size distribution of factor 2 is dominated by the ultrafine particles in the size range between 10 and 100 nm with the peak diameter around 50 nm. It provides 29 % of the particle number concentration but only 7 % to the volume concentration. The diurnal variation shows the peaks has a delay after the rush hour (Fig. 6). This should also assign to traffic-related particles, but it is not the same as the factor 1, possibly aged particles from more distant areas of the sampling site. In addition, evidences from other researches had shown that the diesel and gasoline engine vehicles emit particles in this range (Harris and Maricq, 2001; Kittelson et al., 2006). Factor 2 seems consistent with other findings (Kim et al., 2004; Zhou et al., 2005; Yue et al., 2008), and can interpreted titled as remote traffic. Nevertheless, the differences between these two factors only behaved on geometric mean diameter.

The peak of the number size distribution of factor 3 is at 100 nm. This factor accounts for 33 % of the total number concentration and 21 % of the total volume concentration. The similar diurnal variation trends are observed between number concentration of factor 3 and black carbon concentration (see Fig. 7a). Recent studies in China have turned out that the number size distribution of particles emitted by the combustion sources such as power station or biomass burning also shows the peak in the diameter around 100 nm (Yi et al., 2006; Li et al., 2007). Hence, we refer to the factor 3 is mainly contributed by the other combustion source besides car traffic.

A bimodal particle number size distribution with the peaks 30 nm and 200 nm characterizes factor 4. The concentration of this factor does not change obviously with time which is coincide with the previous study (Kim et al., 2004). Figure 7b depicts the high correlation between the daily pattern of factor 4 and secondary aerosol such as SNA (sulfate, nitrate and ammonium) and OOA (oxygenated organic aerosol organic). The sources of factor 4 seem to be rather complex, origination from the accumulation of secondary aerosol mass growing on pre-existing particles. This factor contributes to number (13 %) and significantly to the volume (71 %) concentration.

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Overall, we could assume that the factors 1–3 are related to the primary emission and the factor 4 represents the secondary aerosol mass. The box plot in Fig. 8 shows the statistics of the source contributions of each factor. The lower source contributions in all four factors of August 2008 were observed compared with the results between 2004 and 2007. The reductions of source contributions for the four factors were 47 %, 44 %, 43 % and 30 %, respectively. The obvious reductions of factors related to traffic and combustion turns out the measures taken focus on the primary emissions during Olympic Games periods are significant. Factor 4, which is assumed as secondary aerosol formation, does not show significant reduction compared with other factors. This discovery indicates that the atmospheric oxidation capacity does not show the corresponding rate of decline trend with the reduced primary emissions. This can be explained by long-range transport of aerosol precursors to be less affected by the reduction measures.

4 Conclusions

As the host city of the 29th Olympic Games, the municipal government of Beijing has been working on improve the air quality since last decade. In this study, the air quality improvement during the Olympic Games period was evaluated based on the long-term measurements of particle number size distributions since 2004.

The meteorological conditions were steady in August from 2004 to 2008. The mean total particle number and volume concentrations (in the size range 3–800 nm) were 14 000 cm^{-3} and 37 $\mu m^3\,cm^{-3}$ in August 2008, respectively. The lowest particle number concentrations in three modes of nucleation (3–20 nm), Aitken (20 100 nm) and accumulation (100–800 nm) are observed in August of 2008 among five years, with reductions of 46 %, 42 % and 40 %, respectively, compared with the mean values of August 2004 to 2007 periods.

Back trajectory cluster analysis was performed to investigate the impacts of the air mass history on the reduction of particle concentrations. In total five significant

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trajectory clusters have been identified. The results show that the air masses from easterly (39 %) and southerly (20 %) directions dominated during the Olympic Games in August 2008. These air masses always indicate pollution events with increased particle mass concentrations. For the Olympic Games period August 2008, a higher frequency (37 %) of these air masses was observed compared to the August periods 2004 to 2007 (16 %). In addition, the lowest particle number and volume concentrations were observed no matter from which direction the air masses came from originally. The reductions of total particle number and volume concentrations varied from 24 % to 46 % and 23 % to 56 %, respectively, for all trajectory clusters, with the mean values of 41 % and 35 %. These evidences imply that the air mass origins have not been the key factor, explaining the reductions of particle number and volume concentrations during the Olympic periods.

In this investigation, we also applied a PMF model to explore particle number size distributions for source apportionment. Four factors have been identified based on their unique size patterns and the correlations with other chemical components. We can assume that factors 1–3 are related the primary emission, in which factors 1 and 2 are related to the traffic emission and factor 3 to other combustion sources. These three factors account for 87 % of total particle number concentration. Factor 4 could be assigned to secondary aerosol formation, which has a great contribution to volume concentration (71 %). The reductions of source contribution for the four factors were 47 %, 44 %, 43 % and 30 %, respectively. These results indicate that the significant reductions of particle concentrations may attribute to the measures taken focus on the primary emissions, especially related to the traffic and combustion sources.

Acknowledgements. This work is supported by the National Natural Science Foundation of China (21025728, 20977001, 21190052), the China Ministry of Environmental Protection's Special Funds for Scientific Research on Public Welfare (201009002). The authors would like to thank Xiaoyang Liu and Jietai Mao (Department of Atmospheric and Oceanic Sciences, Peking University) for providing meteorological data.

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Table 1. Statistics of meteorological parameters in August from 2004 to 2008.

Year	T (°)	RH (%)	WS ($m\,s^{-1}$)	Total Precipitation (mm)/ Number of rainy days
2004	25.5 ± 3.6	60 ± 21	1.3 ± 0.8	56/6
2005	27.9 ± 3.6	68 ± 17	1.2 ± 0.8	92/6
2006	27.6 ± 3.7	71 ± 18	1.2 ± 1.0	98/5
2007	28.2 ± 4.4	59 ± 18	1.0 ± 1.2	116/5
2008	27.5 ± 4.0	69 ± 17	0.9 ± 0.9	150/5

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Table 2. Comparisons of particle number and volume concentrations between different modes (mean ± Standard deviation).

	Number concentration (cm^{-3})				Volume concentration ($\mu m^3\ cm^{-3}$)
	Nucleation	Aitken	Accumulation	Total	Total
Size range (nm)	3–20	20–100	100–800	3–800	3–800
Ave. (2004–2007)	5000 ± 5500	12 300 ± 6200	6400 ± 2800	23 900 ± 10 000	57 ± 36
Ave. 2008	2800 ± 4200	7400 ± 4400	3800 ± 2000	14 000 ± 7000	37 ± 30

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Table 3. Statistics of the particle number and volume concentrations for five trajectory clusters. The reduction is calculated by the equation: Ave. (2004–2007) – Ave. (2008)/Ave. (2004–2007).

	Total number concentration (cm^{-3})				Total volume concentration ($\mu m^3\ cm^{-3}$)			
	2004–2008	2004–2007	2008	reduction	2004–2008	2004–2007	2008	reduction
Cluster 1	31 200	36 000	19 500	46 %	36	37	16	56 %
Cluster 2	18 000	21 000	16 000	24 %	80	85	51	40 %
Cluster 3	15 800	19 100	11 900	38 %	67	78	60	23 %
Cluster 4	21 700	23 000	13 400	42 %	55	60	33	45 %
Cluster 5	27 100	30 200	16 500	46 %	32	32	14	55 %

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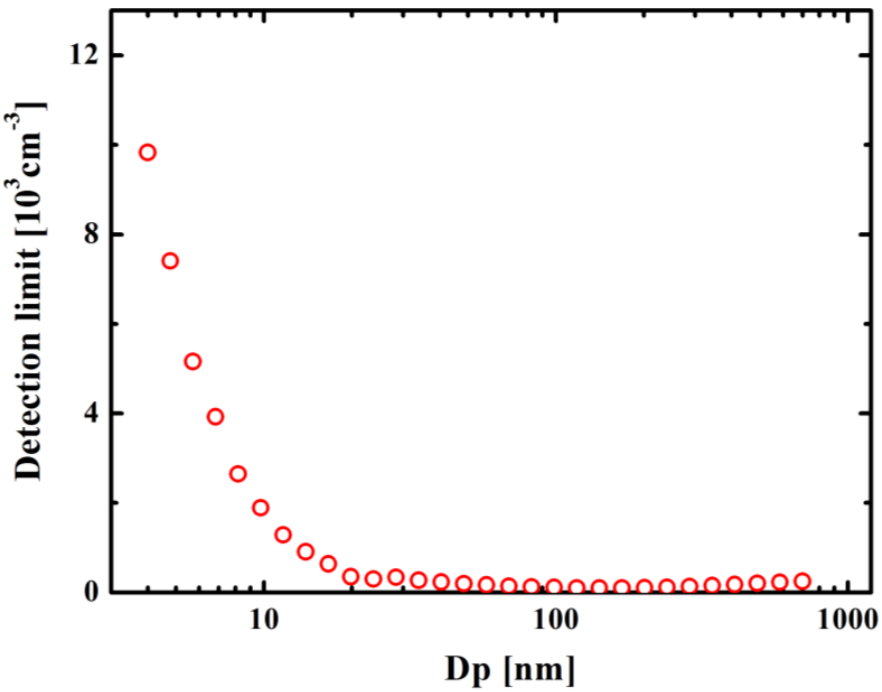

Fig. 1. TDMPS detection limit versus particle diameter.

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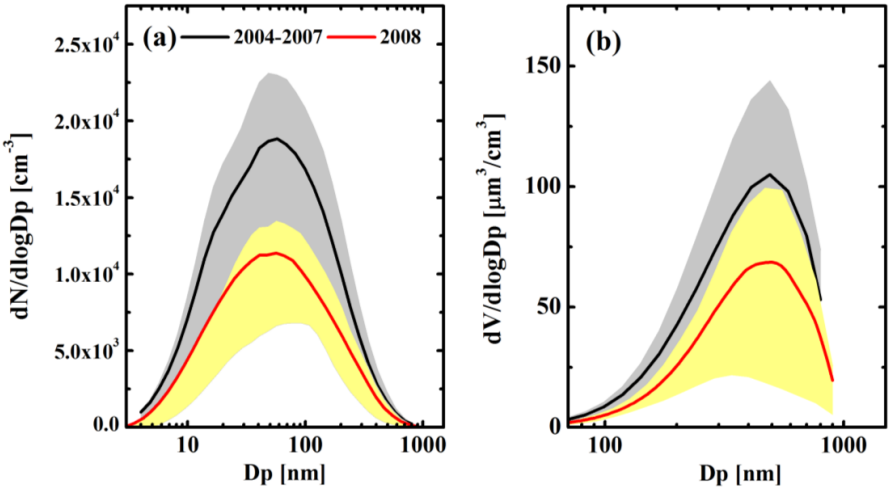

Fig. 2. Mean particle **(a)** number and **(b)** volume size distributions in August from 2004 to 2007 (black line) and 2008 (red line). The upper and lower boundaries of shaded areas indicate the 75th and 25th percentiles.

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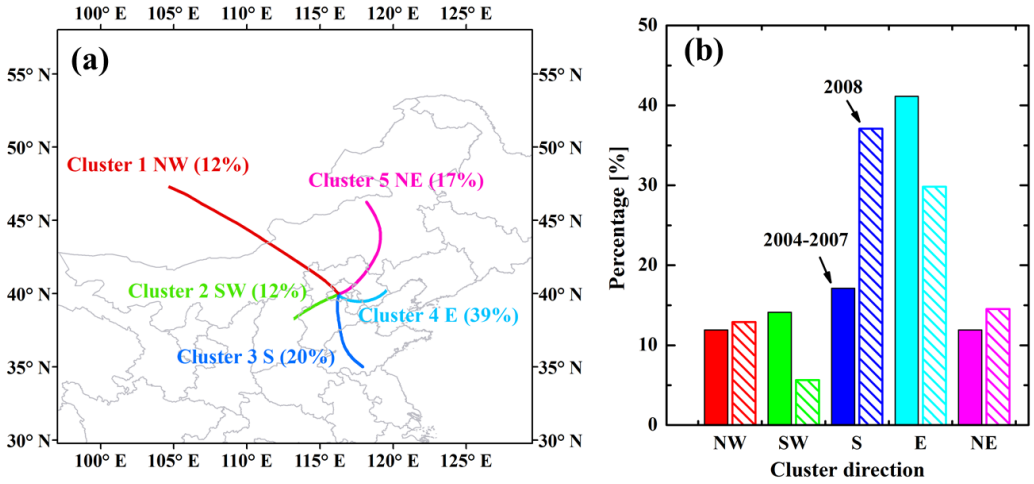

Fig. 3. Mean back trajectories for five trajectory clusters arriving at the PKU site in August 2004–2008 (left panel) and the mean frequencies of five trajectory clusters from 2004 to 2007 and 2008 (right panel). The directions of five trajectory clusters are Northwest (NW, red), Southwest (SW, green), South (S, blue), East (E, cyan) and Northeast (NE, pink), respectively.

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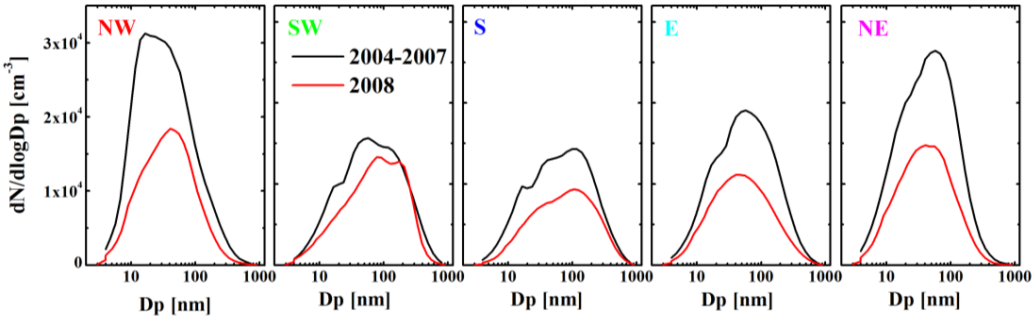

Fig. 4. Mean particle number size distributions for five trajectory clusters, according to the classification in Fig. 3. Black line indicate the mean values from 2004 to 2007, red line represent the mean values of 2008.

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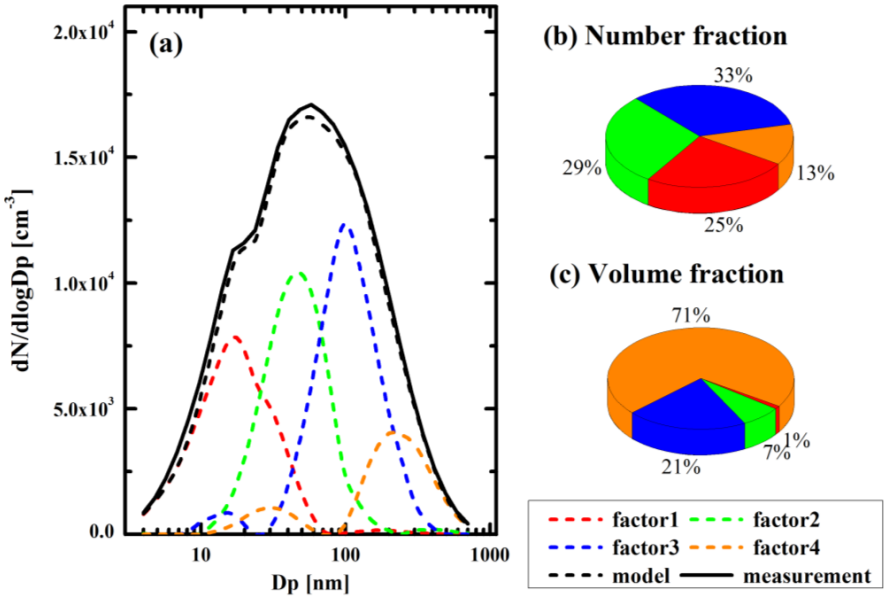

Fig. 5. (a) Number size distribution profiles of four factors. The black dotted and solid lines present the modeled and observed particle number size distributions, respectively; **(b)** and **(c)** are the fractions of four factors in particle number and volume concentrations.

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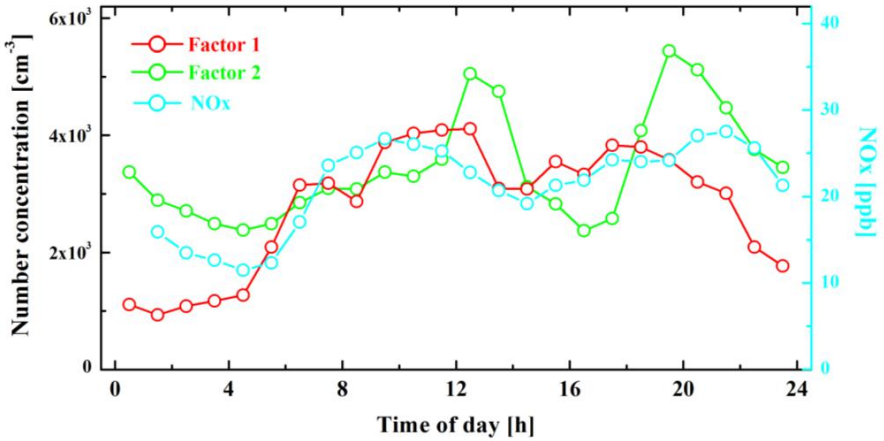

Fig. 6. Diurnal variations of NO_x (cyan) and particle number concentration of factor 1 (red) and factor 2 (green).

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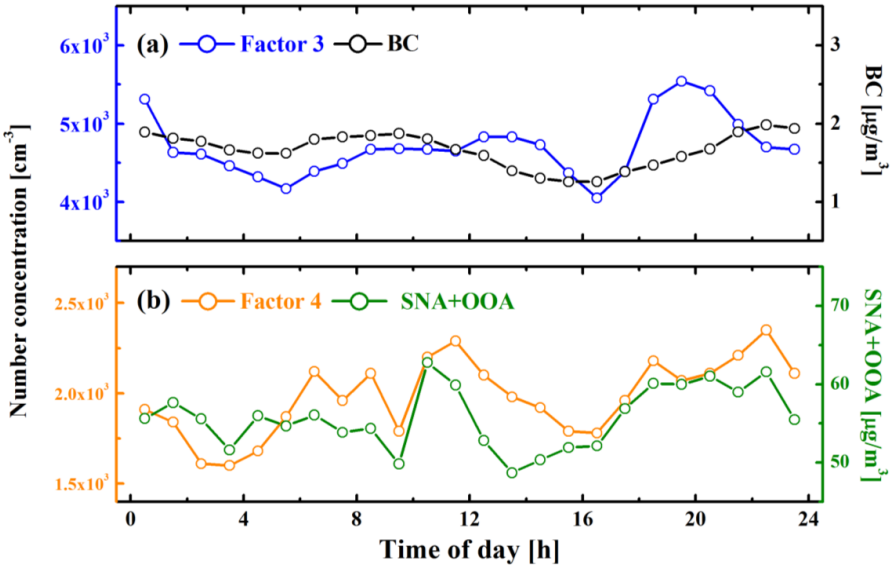

Fig. 7. Diurnal variations of **(a)** particle number concentration of factor 3 (blue) and BC concentration (black); and **(b)** particle number concentration of factor 4 (orange) and the sum of sulfate, nitrate, ammonium and oxygenated organic aerosol organic concentration (olive).

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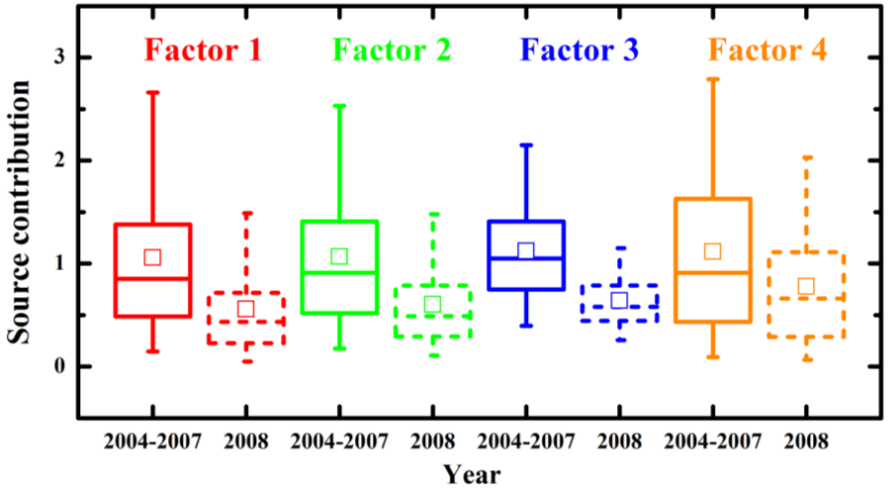

Fig. 8. Box plot of the source contributions from 2004 to 2007 and 2008 for 4 factors. Source contribution values are normalized. The upper and lower boundaries of boxes indicate the 75th and 25th percentiles; the line within the box marks the median; the whiskers above and below boxes indicate the 95th and 5th percentiles; and square symbols represent the means.

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