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Daily variations of size-segregated ambient particulate matter in Beijing

Bangtian Zhou, Huizhong Shen, Ye Huang, Wei Li, Han Chen, Yanyan Zhang, Shu Su, Yuanchen Chen, Nan Lin, Shaojie Zhuo, Qirui Zhong, Junfeng Liu, Bengang Li, Shu Tao*

Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China

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ABSTRACT

Daily, size-segregated particulate matter (PM) samples were collected at Peking University from March 2012 to April 2013. Seventeen indoor air samples were also collected over this period. Winter PM concentrations decreased compared with those reported a decade ago, but summer PM concentrations increased over the same time period. Increasing summer PM concentrations likely resulted from a shift in the major source of PM from primary coal burning to vehicle-associated secondary particle formation. A multiple regression model explained 62% of daily PM concentration variations, and wind direction was the most important factor controlling PM concentrations. Severe pollution was often associated with southeasterly winds, while westerly and northwesterly winds brought relatively clean air. Temperature, precipitation and relative humidity also affected PM concentrations. PM concentrations indoors were generally lower than, but significantly correlated with ambient concentrations. Indoor PM concentrations were also affected by wind speed and temperature.

1. Introduction

For decades China has been suffering from severe air pollution, and particulate matter (PM) is among the most important pollutants (He et al., 2001; Chan and Yao, 2008). This is particularly true in northern China where heating is needed (Li et al., 2014). A recent campaign conducted in seven cities across northern China revealed that annual mean concentrations of PM less than 10 μm in aerodynamic diameter (PM_{10}) reached 180 $\mu g/m^3$ (Li et al., 2014).

Robust associations between inhalation exposure to PM and a number of respiratory and cardiovascular diseases have been reported (Pope et al., 2002). According to the latest estimate, the population attributable fraction of total mortality from ambient PM pollution was as high as 16.2% in China, ranking fourth among major factors contributing to death (Global Burden of Disease (2014)).

The adverse health effects of PM are size dependent and it is believed that fine particles ($PM_{2.5}$), particularly ultrafine PM (<0.1 μm, $PM_{0.1}$), can penetrate deep into the lungs and even into the bloodstream (Brook et al., 2010; Dockery et al., 1993; Englert, 2004). Ambient PM in Beijing is generally from primary sources such as motor vehicles and coal combustion. Re-suspended soil and secondary particles are also important (Sun et al., 2004; He et al., 2001). PM from different sources tend to have different size distributions. For example, coarse PM observed in Beijing is often from the long-range transport of dust storms (Shi et al., 2003) and fugitive sources (Huang et al., 2010). Fine particles usually originate from combustion and secondary formation (Tan et al., 2014). A previous study showed that the annual mean mass fractions of $PM_{2.5}$, $PM_{2.5-10}$, and $PM_{>10}$ in Beijing were 31.8, 21.8, and 46.4%, respectively (Zhang et al., 2010). The contributions from different particle size fractions varied significantly among seasons, with relatively high $PM_{>10}$ (55.2%) during spring and high $PM_{2.5}$ (37.9%) during winter (Zhang et al., 2010).

Ambient PM in Beijing varies diurnally, daily, and seasonally because of the mixture of particle sources and varying meteorological conditions (Wang et al., 2005; Huang et al., 2010). It was suggested that 30% of $PM_{2.5}$ at an urban background site could be attributed to sources outside Beijing, particularly the highly contaminated regions to the south. Therefore, daily concentrations of PM in Beijing are strongly influenced by wind direction (Streets et al., 2007). According to results of a study conducted in 2002, primary $PM_{2.5}$ concentrations in Beijing were higher during winter than summer, mainly because of intensive emissions from coal

* Corresponding author.
E-mail address: taos@pku.edu.cn (S. Tao).

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burning for domestic heating (Sun et al., 2004). Concentrations of ions associated with secondary particles were high during summer, indicating secondary particle formation often occurred under the conditions of high humidity and strong solar radiation (Sun et al., 2004; Wang et al., 2005).

Emissions of primary PM and precursor gases for secondary PM formation in Beijing and the surrounding areas have changed rapidly. Household coal cooking stoves were previously popular in urban areas, but have gradually been replaced with piped natural gas and liquefied petroleum gas. Residential and commercial heating demands are now predominantly provided by centralized heating systems (National Bureau of Statistics of China, 2002–2013). As a result, emissions of primary PM from coal combustion have decreased significantly (Tan et al., 2014). Despite these improvements, the number of motor vehicles has increased from 1.63 million in 2003 to 5.37 million, which has increased both the primary particle and secondary particle precursor gas emissions from the transportation sector (Editorial Board of China Auto Market Almanac (2012)).

The objectives of this study were to collect data on the daily concentrations of size-segregated PM over one year to address: 1) the concentrations and size distributions of PM in ambient air and to compare the results with those reported previously; 2) the major factors affecting daily variations in PM concentrations and size distributions; and 3) the relationship between indoor and outdoor PM concentrations using data from only one household as a preliminary survey.

2. Materials and methods

2.1. Sample collection

PM samples were collected on the roof of a seven-storey building at the Peking University main campus. The campus is located within the urban area of Beijing (39.991°N/116.308°E). Daily (24-h) samples were collected on quartz fiber filters (25 mm in diameter), typically beginning at 6:00 pm, from March 5, 2012 to April 12, 2013. Because of student vacations, equipment failures, and power outages, samples were not collected every day. A total of 312 samples were obtained in this study. Low-volume Leland Legacy sampling pumps (SKC, USA) coupled with Sioutas Cascade Impactors (SKC) were used at a flow rate of 9.0 L/min. The five fractions collected by the impactors were those smaller than 0.25 μm ($PM_{0.25}$), between 0.5 and 0.25 μm ($PM_{0.25-0.5}$), between 0.5 and 1.0 μm ($PM_{0.5-1.0}$), between 1.0 and 2.5 μm ($PM_{1.0-2.5}$), and larger than 2.5 μm ($PM_{>2.5}$).

In addition to the ambient samples, 17 indoor air samples (April 1, 9, 16; May 10, 26; June 21, 30; July 19; Sept. 5, 9, 18, 23; Nov. 11, 12; and Dec. 22, 23, 25, 2012) were also collected for 24 h in a non-smoking, non-cooking residence flat (15th floor, located at the northeast corner of an apartment building several hundreds meters from the ambient air sampling building). The indoor samples were collected to investigate the relationship between PM concentrations indoors and outdoors for this particular household. The samples were collected using the same equipment and procedures. The indoor air sampling days were chosen to cover different seasons and a wide range of ambient PM concentrations.

Table 1
Concentrations and statistical parameters for different PM size fractions (μg/m^3).

	Mean	Std. dev.	P5	P25	P50	P75	P95
TSP	147	96	23	73	130	196	331
$PM_{2.5}$	102	72	16	47	87	144	232
$PM_{1.0}$	83	61	9.9	35	72	118	194
$PM_{0.5}$	68	49	11	30	61	98	153
$PM_{0.25}$	33	26	5.5	17	29	42	78

2.2. Sample analysis

The filters were stored in desiccators for at least 24 h prior to weighing, and then weighed using a five-digit balance (XS-105, Mettler Toledo, Switzerland; Accuracy 0.01 mg) in a temperature-controlled room. At least three readings were obtained for reporting an average value. After sampling, the filters were stored in desiccators for at least 24 h and weighed under the same conditions as the unloaded filters. The gravimetric masses were calculated based on their pre- and post-weights.

Duplicate sampling and gravimetric weighing were conducted randomly and a total of 18 ambient sample pairs and four indoor sample pairs were analyzed. The average coefficients of variations were 7.2, 8.1, 8.5, 5.7, and 9.4% for the ambient duplicate samples and 10, 7.3, 3.6, 6.3, and 5.7% for the indoor samples.

2.3. Meteorological conditions

Meteorological parameters were collected using a weather station (DT80, Rainroot, China) set up on the top of the same building as the sampler. Readings of temperature (*T*), relative humidity (*RH*), wind direction, wind speed (*WS*) and precipitation (*R*) were recorded at 10-min intervals. Daily averages were calculated according to the exact sampling periods. For each 24-h sampling period, the calculated daily mean wind speeds were broken into eight direction components: north (WS_N), northeast (WS_{NE}), east (WS_E), southeast (WS_{SE}), south (WS_S), southwest (WS_{SW}), west (WS_W), and northwest (WS_{NW}), for further data analysis.

2.4. Trajectory calculation

Backward air mass trajectories over 120 h were calculated in 6-h intervals using the NOAA hybrid single-particle Lagrangian integrated trajectory model, driven by meteorological variables from global NOAA-NCEP/NCAR pressure level reanalysis data (Draxler and Rolph, 2003). A probabilistic function was derived and normalized based on the geographical locations of the trajectories.

2.5. Data analysis

The five size fractions were converted into TSP (total suspended solids), $PM_{2.5}$ (<2.5 μm), $PM_{1.0}$ (<1.0 μm), $PM_{0.5}$ (<0.5 μm) and $PM_{0.25}$. Statistica (StatSoft, USA) was used for statistical analysis. A significance level of 0.05 was used. For multiple regression analysis, a forward stepwise method was applied.

3. Results and discussion

3.1. Concentrations

The measured concentrations of the five size fractions are presented in Table 1 as annual means, standard deviations, and several percentiles based on daily concentrations. The measured $PM_{2.5}$ concentration range is similar to those reported previously. For instance, He et al. (2001) measured $PM_{2.5}$ at two stations in Beijing in 1999 and 2000 and the annual mean concentration at one station located approximately 1 km east of our site was 127 μg/m^3. An annual mean $PM_{2.5}$ concentration slightly higher than 100 μg/m^3 has been reported (Zheng et al., 2005). The $PM_{2.5}$ concentration was also very close to that (100 ± 80.3 μg/m^3) measured at Wanliu Station during the same period by the Beijing Municipal Monitoring Center (Beijing Municipal Environmental Monitoring Center, 2014),