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Characterization and source apportionment of particulate PAHs in the roadside environment in Beijing

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HIGHLIGHTS

- Temporal variations and major sources for roadside PAHs in Beijing were identified.
- The roadside PAH concentrations in Beijing are higher than those in other cities worldwide.
- Diesel vehicle, gasoline vehicle and coal combustion are three major sources for PAHs.
- Increase of trucks at night results in higher concentrations for Flu, Pyr, BaA and Chr.
- Temporary traffic control measures lead to much lower roadside PAHs during Olympics.

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ABSTRACT

The profiles of particulate polycyclic aromatic hydrocarbons (PAHs) near a major road and relative major sources were determined based on five 1-week intensive field campaigns in 2008 and 2009, and the impacts of temporary control measures on roadside PAHs during the Beijing Olympics are discussed. The annual average concentration of PAHs in the non-Olympic period was 42.3 ± 52.4 ng/m^3 and clear seasonal variation was present. Diesel vehicles, gasoline vehicles and coal combustion were identified as the three possible major sources of roadside PAHs using positive matrix factorization analysis. During the Olympics, the average total PAH concentration decreased to 4.8 ± 2.7 ng/m^3, which was attributed primarily to the reduction of local emissions. Temporary traffic control measures significantly changed the diurnal pattern of particulate PAHs at the roadside site. Diesel vehicle contribution, in particular, decreased to a negligible fraction because heavy-duty diesel vehicles were strictly banned.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are major toxic contaminants widely dispersed in the environment. Particulate PAHs, especially those with high-molecular-weight, are mainly found in fine particulate matter ($PM_{2.5}$) in the urban atmosphere (Allen et al., 1996). In the 1970s the US Environmental Protection Agency (US EPA) identified 16 PAH species as "priority pollutants". Some of the PAH species such as benzo(a)pyrene (BaP), chrysene (Chr) and indeno(1,2,3-cd)pyrene (InP) were reported to be mutagenic and carcinogenic (Nisbet and LaGoy, 1992). Taioli et al. (2007) discovered clear evidence of genotoxic effects and DNA damage associated with PAH exposure through molecular epidemiology studies in Europe. Limits for air concentrations of PAHs have been proposed by several countries and international organizations (WHO, 2000). In China, the limit for daily averaged BaP concentration in ambient air is 0.01 μg/m^3. Also, seven PAH species, including naphthalene (NaP), fluorine (Flu), benzo(b)fluoranthene (BbF), benzo(f)fluoranthene (BkF), BaP, InP and benzo(g,hi)perylene (BghiP), were identified as priority pollutants since 1990s.

Anthropogenic PAHs are primarily from the incomplete combustion and pyrolysis of organic materials such as fossil fuels, wood and plastics. PAHs derived from different sources have different profiles and tracers, which are generally used in the source apportionment of urban pollution (Chen et al., 2012). Researchers have identified vehicle exhaust emissions as one of the most important sources of PAHs in urban areas in China. For example, emissions from diesel and gasoline vehicles contributed the second largest fraction of

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particulate PAHs in Beijing after coal combustion (Duan, 2005; Zhang et al., 2007). PAHs derived from diesel and gasoline vehicles have specific profiles and size distributions (Rogge et al., 1993). However, most of the previous studies focused on urban-scale aerosols or direct tail-pipe particles with less attention given to the relationship between traffic conditions and the level of roadside ambient particulate PAHs.

It is well known that temporary traffic control measures were implemented from July 1 to September 20, 2008 during the Beijing Olympic Games to improve air quality and traffic conditions. For example, (1) 70% of government vehicles were ordered off the road; (2) trucks could only operate outside the 6th Ring Road, an expressway circling Beijing approximately 17 km from the city center, unless they were issued special passes; (3) high emitting vehicles with yellow environmental labels were banned from the roads throughout Beijing; and (4) private vehicles could only operate on odd or even days depending on the last digit on their license plate. These temporary traffic control measures changed the traffic pattern in the urban area and resulted in a significant reduction in vehicle emissions of PM, volatile organic compounds (VOC) and other pollutants in Beijing (Wu et al., 2011; Zhou et al., 2010). Seeing the substantial benefits in vehicle emission reduction due to those temporary traffic controls, similar actions were subsequently taken during other important events held in China (e.g., the Guangzhou Asian Games during 2010) (Zhang et al., 2013). In addition, other sources in Beijing and surrounding areas, such as coal-fired power plants, industrial boilers and construction activities, were under government restrictions. These control measures for mobile sources as well as other sources helped significantly improve air quality in the roadside environment (Song et al., 2012; Yang et al., 2011). Based on continuous PAH measurements from October 2007 to August 2008 in the North China Plain, Wang et al. (2011) found that low PAH concentrations during summer 2008 in Beijing's urban area might be associated with the control measures.

The primary objective of this study is to quantify and assess the characteristics of particulate PAHs in the atmosphere surrounding a major urban expressway in Beijing, including the seasonal variation, the diurnal profile and an Olympic-period profile. A factor analysis method is employed to identify the major sources and their contributions to particulate PAHs in the roadside micro-environment. We also discuss the effectiveness of the temporary control measures to reduce PAHs during the 2008 Olympic Games and indicate how this information will help policy-makers to develop future strategies for ensuring better air quality in the roadside micro-environment.

2. Material and methods

2.1. Sampling

Particulate PAH samples were collected on the south side of the North 4th Ring Road, an eight-lane urban expressway encircling Beijing approximately 8 km from the city center. The road is perpendicular to the prevailing wind direction in this area. A satellite map of the monitoring site and the surroundings is given in Fig. 1. Instruments were placed within 2 m of the curb and at 1.8–2.5 m above ground level at the roadside station. A high volume PM air sampler (GUV-15HBL-1, Thermo Inc.) with a $PM_{2.5}$ high volume size selective inlet (G1200-41, Thermo Inc.) was used for sample collection at a calibrated flow rate of 1.13 m^3/min. The continuous mass concentration of $PM_{2.5}$ was measured by a tapered element oscillating microbalance monitor (TEOM 1400a, R&P Co.). A video camera was set on an overpass near the roadside monitoring station and the local traffic flow was recorded for 20 min in an hour at 1–2 weekdays during each 1-week field campaign. The numbers of light-duty vehicles (LGVs, including cars and taxis), vans and light-duty trucks, buses and heavy-duty vehicles (HDVs) were counted separately from the recorded video. Meteorological data were taken simultaneously by an automatic weather station (Vantage Pro2™, Davis Inst.), which was located on the top of a five-story (20 m high) building about 200 m away from the monitoring site. The surrounding buildings nearby the sampling site are generally less than 10 m in height; therefore they would not result in strong channeling effects or other obstructions.

Sampling was conducted continuously for about one week in both the summer and winter of 2008 and 2009, including a special enhanced sampling period during the Beijing Olympic Games (specific sampling dates shown in Table 1). $PM_{2.5}$ samples were collected on pre-heated (550 °C for 12 h) quartz fiber filters (TISSUQUARTZ, Pall Co.) for a period of 12 h (07:00–19:00, 19:00–07:00) in order to evaluate the differences in PAH species in the daytime and nighttime, when the emission profiles from vehicle exhaust and other sources were different. Field blanks for every sampling period were taken in this study. For each sampling period, three blank filters were exposed during the whole sampling period at the roadside site, and then were treated in the same way as the samples. For the data analysis and discussion in this paper, samples collected during rainy periods (precipitation > 5 mm/h) were excluded to avoid the removal impacts of special meteorological conditions. The collected $PM_{2.5}$ samples were classified into three categories: the non-Olympic winter period (denoted by "Winter", December 2008 and December 2009); the non-Olympic summer period (denoted by "Summer", June 2008 and August 2009); and the Olympic

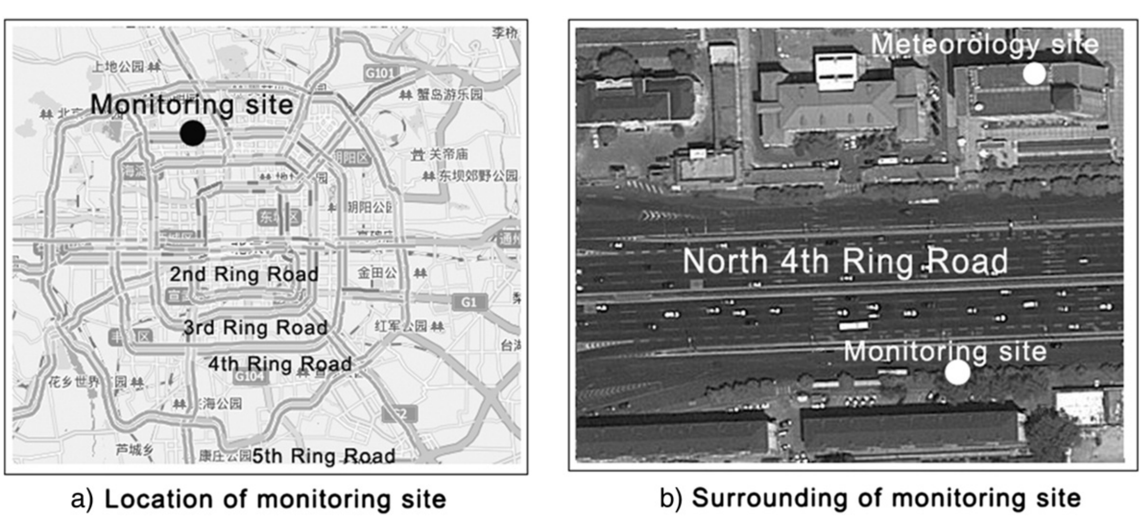

Fig. 1. Location and surroundings of the monitoring site and meteorology site.