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A wintertime study of $PM_{2.5}$-bound polycyclic aromatic hydrocarbons in Taiyuan during 2009–2013: Assessment of pollution control strategy in a typical basin region

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HIGHLIGHTS

- $PM_{2.5}$-bound PAH pollution in Taiyuan was still serious despite being controlled.
- Coal combustion and coking activities dominated the sources of PAHs in $PM_{2.5}$.
- Coal/coking-related PAHs emitted in Taiyuan basin could be transported to Taiyuan.
- Increasing local traffic caused increasing health risk through enriching HMW PAHs.
- Controlling residential coal combustion was effective to abate PAHs pollution.

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ABSTRACT

Taiyuan city in Shanxi province, China has been one of the top heavily polluted cities in the world for a long time with large industrial emissions and high disease burden. Many pollution control strategies have been implemented forcefully by the government in recent years in Taiyuan. To better understand the effect of the strategies and related influence factors, we studied polycyclic aromatic hydrocarbons (PAHs) in fine particulate matter ($PM_{2.5}$) during heating seasons in Taiyuan from 2009 to 2013. The results showed that the concentrations of $PM_{2.5}$ (70.7–477.9 μg/m^3) and related total PAHs (T-PAHs, 128.7–1840.2 ng/m^3) far exceeded the air quality standards issued by the Ministry of Environmental Protection of China (MEP) and were higher than those in many domestic and foreign cities in spite of the pollution control. Source apportionment by the diagnostic ratio analysis and PMF model found that coal consumption contributed the most (52.1%) to the total $PM_{2.5}$-bound PAHs followed by the coking industry (27.3%) and traffic exhausts (20.6%). Significant decreases in $PM_{2.5}$ and PAHs levels were found in 2013, which was probably due to the large abatement of residential coal consumption and favorable meteorological factors. Being located in the north of Taiyuan basin, the pollution in Taiyuan could be aggravated by the regional transport of coal combustion- and coking-related pollutants from other industrial development zones in the south-western basin as found by the analysis of meteorological influence and back trajectory. Although the PAHs were the lowest in 2013, the BaP_{eq} or ILCR were the highest in that year. This should be related to the increasing vehicle numbers in Taiyuan, because vehicle exhaust tends to enrich in higher molecular weight and more toxic PAHs. Our results provided useful guidance for solving the air pollution problem for cities in a semi- or total-closed basin with coal as the major energy source such as Taiyuan.

1. Introduction

PAHs are a ubiquitous class of toxic pollutants in the urban

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atmosphere and have attracted increasing attention in recent decades. Particulate PAHs, especially $PM_{2.5}$-bound PAHs, are an important form of the atmospheric PAHs (Albuquerque et al., 2016). The synergy between $PM_{2.5}$ and PAHs could cause serious health effects because of the oxidative, pro-inflammatory, carcinogenic, and mutagenic damage from both of them (Yang et al., 2014; Brumsack et al., 1984). China is one of the largest PAH-emitting countries in the world with heavy $PM_{2.5}$ pollution (Inomata et al., 2012; Zhang and Tao, 2009; Zhang et al., 2011) due to large amounts of coal combustion and coking production especially in the northern area. High levels of $PM_{2.5}$-bound PAHs were detected in many cities in China including Beijing (407.6 ng/m^3), Zhengzhou (254.0 ng/m^3), and Qingdao (263.0 ng/m^3), with the levels of carcinogenic benzo[*a*]pyrene (BaP) far exceeding the guideline level of 2.5 ng/m^3 (http://www.sciencedirect.com/science/article/pii/S0269749113003321MEP, 2012) (Wang et al., 2008, 2014a; Guo et al., 2009). It was reported that 1.6% of the lung cancer morbidity in China was due to inhalation exposure to PAHs in the ambient air (Zhang et al., 2009). $PM_{2.5}$ could be persistently suspended in the atmosphere and cause serious damage to the environment and human health through long-range atmospheric transportation. Consequently, $PM_{2.5}$-bound PAHs emitted from China have also influenced surrounding countries like Japan and Korea (Inomata et al., 2012).

To reduce atmospheric pollution, efforts have been made to lower the emissions by implementing laws and regulations, by improving combustion technologies, by optimizing structure and layout of regional industry, and by employing cleaner fuel sources such as natural gas in China (Zhang et al., 2016). In terms of the Bulletins of Environmental State issued by MEP, the concentrations of SO_2, NO_x and PM have been reduced significantly year-by-year, and the air quality has met the Chinese Grade II standards in many regions (MEP, 2016). However, in many Chinese cities under pollution control, ambient PAHs had not showed a downward trend as expected. In Beijing 2005, there have been significant decreases in carcinogenic benz[*a*]anthracene (BaA) and chrysene (Chr) and increases in less harmful fluoranthene (Flu), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF) and indeno[1,2,3,-*cd*]pyrene (InP) in respirable particles. This is due to the reform of energy structure from coal to cleaner energy (Wang et al., 2008). In Beijing 2009, one year after the 29th Olympic Games when stringent and large-scale pollution controls were implemented, the concentration of gaseous PAHs decreased by more than 60% while PAHs in particulate phase kept almost unchanged compared to those before the Games (Ma et al., 2011). In Guangzhou from 2001 to 2009, a shift of dominant PAH sources from vehicle emission to coal combustion was believed to be the result of targeted control measures (Gao et al., 2012). However, higher PAH concentrations in $PM_{2.5}$ were found in the winter of 2012 than in 2009 (Gao et al., 2012; Liu et al., 2015). Long term observations in Zhengzhou, a middle city in China, indicated only a slight decrease of PAHs in $PM_{2.5}$ from 2011 to 2013 (Wang et al., 2015).

The unclear factors hampering the abatement of $PM_{2.5}$-bound PAHs are plaguing the governmental operation in many cities in China. Studies into the change of various atmospheric pollutants in Beijing during severe pollution episodes, world-scale sports games and high-level meetings indicated that regional transport also plays an important role in exacerbating local air pollution except local emissions. Regional joint control is ultimately needed to solve local pollution (Zhang et al., 2016; Wang et al., 2010, 2015). Due to the complexity of air pollution in nature under different meteorological and topographic conditions, the problems existing in the present pollution control strategies need to be clarified in different places before formulating reasonable and feasible joint-control strategies.

The Taiyuan-centered Taiyuan basin is the largest coking base in China and produces over half of the national coke. This makes it the main emitter of PAHs in China. Average PAHs emission per unit area reached up to 44.62 kg/km^2, which was 3 times the national level and 13 times American level (Jiang et al., 2013). As a typical valley-basin city, heavy local and regional emissions, as well as its special geographical condition (Fig. 1) lead to severe air pollution in Taiyuan. This ranks it among the most polluted cities in the world (Zhao et al., 2008) especially for PAHs (Zhang et al., 2007). Previous studies found over 150 ng/m^3 of $PM_{2.5}$-bound PAHs in the atmosphere of Taiyuan, and high cancer risk exceeded acceptable risk level (Xia et al., 2013). Many measures were implemented by the government to alleviate the air pollution during its 11 and 12 five year plans (FYPs) (Tang et al., 2014). Many heavy industries were reformed, reconstructed and moved out of Taiyuan to other less-developed regions in the basin (Wang et al., 2014b). However, the effectiveness is called into question because of the frequent occurrence of heavy haze featuring high levels of $PM_{2.5}$ in Taiyuan. A total of 203 days exceeded the Grade II standard of the National Ambient Air Quality Standard (NAAQS) (GB3095-2012) according to air quality monitoring data in 2013 (TSB, 2014). Except high health risk, PAHs also have tracer properties to resolve sources. Therefore, temporal variation of $PM_{2.5}$-bound PAHs is a primary issue and an important key to assessing the effectiveness of source reduction measures and regulatory controls that remain unclear (Albuquerque et al., 2016).

In this study, the long-term and elaborate analysis of $PM_{2.5}$-bound PAHs was undertaken in Taiyuan from 2009 to 2013 in the winter seasons. The samples were subjected to concentration measurements of $PM_{2.5}$ and associated PAHs, and then the main sources were identified and apportioned by employing the PMF model in conjunction with the hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model—both of these were well-adopted for source apportionment analysis in many studies (Wang et al., 2015; Gao et al., 2012). The objectives of this study are: (1) to know the levels and variation of $PM_{2.5}$-bound PAHs in Taiyuan in winter in recent five years, (2) to clarify the impact of meteorological and topographic factors on PAH variations, and (3) to assess the effects of emission-reduction measures. Our results may provide valuable insights for air pollution abatement in other energy-dependent cities—especially those in basins.

2. Materials and methods

2.1. Study area

The city of Taiyuan lies at the center of Shanxi province in China and in the northern end of Taiyuan basin with an East-West span of 144 km and a North-South span of 107 km (Fig. 1). The terrain is like a dustpan surrounded by mountains in its west, north and east directions, but with valley plain in the south. Located in the continental interior and far away from the coast, Taiyuan belongs to the warm temperate zone with continental monsoon climate. The annual average temperature in Taiyuan is 9.5 °C, with the lowest value in winter and the highest in summer.

The Taiyuan basin including Taiyuan city in Jinzhong Prefecture and other parts of Lvliang Prefecture is the major industrial center of Shanxi with many coking plants, steel companies and fire-powered electricity plants. Unlike other places in the north plain of China, Taiyuan basin is an independent area with high pollutant accumulation. This is because it is between two high mountains: Lvliang Mountain and Taihang Mountain (1000–3000 m ASL). Since 2011, dozens of heavily polluted enterprises were moved out of Taiyuan to other places in the basin Gujiao and Qingxu counties, relocated, or even shut down. With the movement of heavy industries, the industrial center was transferred gradually from