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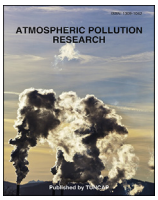

Particle dry deposition of polycyclic aromatic hydrocarbons and its risk assessment in a typical coal-polluted and basin city, northern China

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ABSTRACT

Atmospheric dry deposition is a major pathway of pollutants removal from the atmosphere to the water and soil ecosystem. Polycyclic aromatic hydrocarbons (PAHs) in Taiyuan, China, are one class of the most hazardous air pollutants due to the high emission intensity from coal consumption. To better understand the PAHs removing from the atmosphere, 16 USEPA priority PAHs (except for naphthalene) in the particle dry deposition samples were detected from August 2013 to November 2014 at an urban site in Taiyuan. Dry deposition fluxes of particles were estimated as 157.44–358.17 mg/(m^2·d), with the highest value in spring and lowest in autumn. Particle dry deposition fluxes of ∑15-PAHs ranged from 2436.20 to 14,967.36 ng/(m^2·d), and was the highest in winter and lowest in summer. Higher PAHs particle dry deposition fluxes were related to higher PAHs emission, lower air temperature, less precipitation as well as wind speed. Overall dry deposition velocities of 15-PAHs were 0.056 ± 0.027 cm/s in winter and 0.42 ± 0.050 cm/s in summer, respectively, and high molecular weight PAHs showed low dry deposition velocities. Positive matrix factorization model's results indicated that coking, vehicle exhaust, coal combustion were the primary PAHs sources in particle dry deposition samples, accounting for 18.64%, 26.54% and 54.82% of total PAHs, respectively. The toxic equivalent concentrations (BaPeq) of ∑15-PAHs in particle dry deposition samples was in the range of 0.69–2.94 μg/g, which might be a good explanation to high soil PAHs concentrations reported in Shanxi province, China. Furthermore, this work could prove the significance of coal combustion, and related pollution control works are needed to conduct to alleviate the PAHs pollution situation.

1. Introduction

Interest in atmospheric deposition has increased over the past several decades. Atmospheric deposition is considered as an important removal pathway of hazardous pollutants from atmosphere into aquatic and terrestrial ecosystem, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), trace metals, etc. (Franz et al., 1998; Tasdemir et al., 2004; Terzi and Samara, 2005; Gonzalez-Gaya et al., 2014; Pan and Wang, 2015). Dry and wet depositions are two main mechanisms of atmospheric deposition. Dry deposition refers to the transfer of both gaseous and particulate pollutants from air to nature surface in no-precipitation (Odabasi et al., 1999; Terzi and Samara, 2005; Tasdemir and Esen., 2007; Zhang et al., 2008). In contrast to wet deposition, dry deposition is a continuous and dependable process and the predominant removal mechanism on an annual basis, especially in regions with low precipitation, such as Northern

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China, Mediterranean climate area, and other reported areas (Muezzinoglu and Cizmecioglu, 2006; Škrdlíková et al., 2011; Pan and Wang, 2015; Zhang et al., 2014; Pekey et al., 2007; Odabasi et al., 2002; Tsitouridou and Anatolaki., 2007; Wang et al., 2016).

PAHs are a class of the most serious contaminants due to their ubiquitous occurrence and potential risk to ecosystem and human health. Dry deposition fluxes of atmospheric PAHs are often conducted to assess its ecological load, which are highly influenced by PAHs emissions strength and prevailing meteorological conditions. In recent years, China has become the biggest PAHs emitter in the world due to the rapid industrialization and economic development, contributing 10^6 Gg 16 priority PAHs in 2007 (Shen et al., 2013). Several previous studies showed that PAHs dry deposition fluxes in the northern China (Beijing, Tianjin, etc) (Wu et al., 2005; Zhang et al., 2008; Wang et al., 2014) were higher than those in the southern (Guangzhou, Hangzhou, etc) (Li et al., 2009; Chen and Zhu, 2010; Zhang et al., 2012; Guo et al., 2014). This situation was related to more PAHs emissions due to higher coal consumption and less rainfall regime in northern China (CSB, 2007-2014). PAHs dry deposition fluxes were also higher in industrial (more industrial activities) and urban areas than those in rural sites (Bae et al., 2002; Fang et al., 2004; Terzi and Samara, 2005). Because of increasing residential heating in winter, Dry deposition fluxes for ∑PAHs were generally higher than those in other seasons (Bae et al., 2002; Terzi and Samara, 2005; Pekey et al., 2007). Furthermore, weather condition (e.g. wind speed, temperature) also would affect the PAHs dry deposition fluxes (Bae et al., 2002; Terzi and Samara, 2005; Tasdemir and Esen., 2007).

Atmospheric PAHs are distributed in the gas and particulate phases, which is a key factor influencing the removal of PAHs from the atmosphere by dry deposition. In general, surrogate surfaces like greased smooth deposition plate, automated wet/dry deposition collectors and water surface, etc, have been used to collect dry deposition by directly simulating natural surfaces (Bae et al., 2002; Shannigrahi et al., 2005; Terzi and Samara, 2005; Tasdemir and Esen., 2007; Birgül et al., 2011). Since the surrogate surfaces seldom completely simulated natural surfaces and gaseous and particulate PAHs might not deposit in an equal measure, the measured results could not fully extrapolate the natural deposition (Terzi and Samara, 2005; Zhang et al., 2012). For these reasons, PAHs dry deposition fluxes in the particulate and gas phases could also be estimated by the measured ambient concentrations multiplying by an assumed or modeled deposition velocities (V_d) in the inferential method (Sheu et al., 1996; Fang et al., 2004; Terzi and Samara, 2005; Zhang et al., 2012). In previous studies, V_d for the gas and particle PAHs both varied largely with 0.001–0.01 and 0.01–6.7 cm/s among different areas, respectively (Sheu et al., 1996; Odabasi et al., 1999; Vardar et al., 2002; Terzi and Samara, 2005; Bozlaker et al., 2008; Demircioglu et al., 2011). Meanwhile, V_d of particles based on model estimation were generally underestimated due to lack of larger particles (>10 μm) or lower V_d than field studies investigating some trace species for 0.1–1 μm (Hoff et al., 1996; Kaupp and McLahlan, 1999; Odabasi et al., 1999; Zhang et al., 2001). And large uncertainties would be introduced when a previous reported or molded V_d was used to estimate the PAHs dry deposition fluxes (Zhang et al., 2012).

Taiyuan, a basin city in northern China, is faced with serious environmental problems in the last few decades because of high coal consumption (Xia et al., 2013; Wang et al., 2013). It has been reported that the PAHs emission in Taiyuan reached 332.10 t in 2010, and PAHs emission intensity was 3.72 times higher than national mean level (Jiang et al., 2013). PAHs have become one of the primary pollutants in Taiyuan and posed a severe threat to the human health and local ecosystem (Xia et al., 2013; Li et al., 2016). Furthermore, wet deposition fluxes of ∑16-PAHs in dissolved phase in Taiyuan reached 2342.80 ng/(m^2·d) and further proved the worsening air pollution situation (Zhang et al., 2014). However, there were few literature on the dry deposition of PAHs in Taiyuan. In this context, particle dry deposition samples were collected by a half-month period from August 2013 to November 2014 in Taiyuan. The main objectives of this study were to: (1) measure the particle dry deposition fluxes and their dry deposition velocities of PAHs in Taiyuan, (2) elucidate the influence of meteorological parameters (e.g. temperature, relative humidity (RH), precipitation) on the atmospheric PAHs particle dry deposition, (3) evaluate the potential risk of particle PAHs dry deposition, (4) identify PAHs potential sources and their respective contributions. These results might not only gain a better understanding of PAHs removal mechanism by dry deposition but also provide some references to pollution control and risk assessment of PAHs.

2. Materials and methods

2.1. Regional description

Taiyuan, the provincial capital of Shanxi, has a population of 4.2 million, and is in the north of Taiyuan Basin which limits the diffusion of air pollutants (Figs. S1b and c). Located in the continental interior, Taiyuan belongs to the warm temperate zone continental monsoon climate, and the annual average temperature was 11.2 °C, with the lowest in December and January (−5.1 °C) and the highest in June and July (23.6 °C). The weather is extremely dry and prone to dust storms in spring (from March to May), hot and rainy in summer (from June and August), and dry and cold in winter (from December to February). Annual average precipitation is about 487.3 mm and over 60% of the annual precipitation occurs in the summer (TSB, 2014).

There are a lot of industrial plants such as Taiyuan Power Plants, the Iron and Steel Group Company and Heavy Machinery Making Group Company in Taiyuan Basin (Fig. S1c). As the primary energy, the annual coal consumption in Taiyuan reached 6.6×10^8 tons in 2013, accounting for more than 90% of the total energy consumption (TSB, 2014). Furthermore, the main coking and coal industries in Shanxi province are located in the Taiyuan Basin, such as Qingxu, Jiexiu, Xiaoyi and Fenyang counties. Influenced by the industrial activities and special basin topography in the Taiyuan basin, Taiyuan is facing serious PAHs pollution (Xia et al., 2013; Jiang et al., 2013; Li et al., 2016).

2.2. Sampling procedures

Half-monthly particle dry deposition samples were collected by a bucket (diameter 200 mm, height 400 mm) on an automatic dry and wet deposition collector (UNS 130/E, Eigenbrodt, Germany) on the rooftop of a 16–story building in the campus of Taiyuan University of Science and Technology (about 50 m above ground) in Taiyuan city (Fig. 1). A quartz microfiber filter (diameter 200 mm, GF/A, Whatman, England) was set at the bottom of the bucket to collect particle dry deposition. The filters were prebaked in a muffle furnace at 450 °C for 4 h, then equilibrated at constant temperature and humidity (25 °C, 50%) for 24 h and weighed by electronic microbalance (AB204-S, Mettler Toledo, Switzerland). After sampling, the bottom and inner surface of the bucket were also wiped with pre-clean and weighed cotton. The cotton and filter were sealed with aluminum foil, weighted after equilibrium, and stored in a freezer at −10 °C for further analysis. The mass of dry deposition sample for particles was calculated by the difference before and after sampling of the filter and the cotton. A total of 30 particle dry deposition samples were collected during the period of August 2013 to November 2014. At the same time, total suspended

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