



Atmospheric deposition of polycyclic aromatic compounds and associated sources in an urban and a rural area of Chongqing, China



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HIGHLIGHTS

- Atmospheric deposition of PPAHs, NPAHs and OPAHs were measured in Chongqing.
- OPAHs occurred in all samples with deposition flux 2 times lower than PPAHs.
- Both secondary formation and combustions had impacts on PACs in Chongqing.
- High relative humidity may be important for the high OPAHs deposition in winter.

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ABSTRACT

Monthly bulk (dry + wet) deposition samples were collected at an urban and a rural site in Chongqing, southwestern China during May 2014 to April 2015 for analyzing the contents of parent polycyclic aromatic hydrocarbons (PPAHs) and three types of substituted PAHs (SPAHS) including oxygenated PAHs (OPAHS), nitrated PAHs (NPAHS) and methyl PAHs (MPAHs). Annual average (\pm standard deviation) deposition fluxes of Σ PPAHs, Σ OPAHS, and Σ MPAHs were 536 ± 216 ; 221 ± 118 , and 131 ± 41.9 ng/m²/d, respectively, in the urban area, and 347 ± 185 , 160 ± 112 , and 85.2 ± 32.0 ng/m²/d, respectively in the rural area. Deposition of Σ NPAHS (6.01 ± 3.93 and 3.91 ± 4.84 ng/m²/d) were about two orders of magnitude lower than those of Σ PPAHs. In the urban area, temporal variations of PPAHs and MPAHs fluxes were positively correlated with particle deposition, while the trends of OPAHs and NPAHs were probably controlled by secondary formation. In the rural area, SPAHs and PPAHs deposition fluxes had similar temporal trends but differed from particle deposition. High relative humidity in Chongqing likely played an important role in facilitating the partitioning of OPAHs to atmospheric aerosols and resulting in the relatively high OPAHs level in winter. Principle component analysis identified secondary formation (21.7%) and combustion emission (52.7%) as two important contributors to polycyclic aromatic compounds (PACs) deposition fluxes in urban area.

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1. Introduction

Polycyclic aromatic compounds (PACs) have been causing great

concerns because of their widespread occurrence and potential toxic effects to ecosystem and human health (Ross and Nesnow, 1999; Wang et al., 2015). Sixteen polycyclic aromatic hydrocarbons (PAHs) have been designated as priority pollutants and regulated by the United States Environmental Protection Agency due to their potential teratogenicity, mutagenicity, and carcinogenicity (Ross and Nesnow, 1999). Recently, more and more attentions have been paid to the PAHs derivatives, such as alkylated PAHs, nitrated PAHs (NPAHs) and oxygenated PAHs (OPAHS) (Tidwell

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et al., 2016; Tomaz et al., 2017; Zhang et al., 2015a, 2015b). These derivatives have been ubiquitously detected in the environment and some of them are generally more toxic than their parent PAHs (PPAHs) (Alves et al., 2017; Durant et al., 1996; Lin et al., 2015b; Umbuzeiro et al., 2008; Wei et al., 2015). For instance, the direct acting mutagenicity of airborne particles induced by OPAHs and NPAHs was 2 times higher than the indirect mutagenicity caused by PPAHs in an urban environment in Beijing, although the sum of OPAHs and NPAHs concentrations was only 8% of the PPAHs concentrations (Wang et al., 2011a). Moreover, OPAHs are end products of many biological and chemical degradation pathways, making them more persistent in the environment (Lundstedt et al., 2007).

OPAHs and NPAHs are released together with PPAHs into the atmosphere as they all are the products of incomplete combustion, such as coal combustion and vehicle engine exhaust (Huang et al., 2014b; Kawanaka et al., 2007; Wang et al., 2016b, 2016c). They can also be formed from PPAHs in post-emission processes through chemical oxidation, photooxidation, and microbiological processes (Cerniglia, 1993; Haritash and Kaushik, 2009; Kojima et al., 2010; Yu, 2002; Zhang et al., 2011). Many studies have been carried out to investigate the vapour phase and heterogeneous reactions of PPAHs with atmospheric oxidants such as OH, NO₃, and O₃ (Atkinson and Arey, 2007; Jariyasopit et al., 2014). However, the relative contributions of direct combustion emissions and secondary reactions to the observed atmospheric burden of the PAHs derivatives were not well understood (Keyte et al., 2013), despite many existing efforts using various approaches (Eiguren-Fernandez et al., 2008; Kojima et al., 2010; Lin et al., 2015a; Ma et al., 2016; Zhuo et al., 2017). For example, Eiguren-Fernandez et al. (2008) and Kojima et al. (2010) quantified secondary contributions of OPAHs in Los Angeles and Tokyo, respectively, based on the same model developed by Eiguren-Fernandez et al. (2008). Positive Matrix Factorization and Principal Component Analysis-Multiple Linear Regression were also adopted to evaluate the relative contribution of secondary formation to NPAHs and OPAHs in Beijing (Lin et al., 2015a), Hong Kong (Ma et al., 2016), and Nanjing (Zhuo et al., 2017).

The majority of the existing studies worldwide focused on the levels, sources, gas-particle partitioning, and atmospheric reactions of PPAHs and related derivatives, with only limited efforts attributed to transport and fate of these pollutants. Atmospheric deposition is the major process removing these chemicals from the atmosphere and also an important pathway delivering contaminants to various ecosystems (Bidleman, 1988; Wang et al., 2011b; Xing et al., 2016). Existing studies on atmospheric deposition were mostly focused on PPAHs (Moon et al., 2006; Pekey et al., 2007; Poor et al., 2004; Tsapakis et al., 2006; Wang et al., 2011b), and very limited efforts concerned PAHs derivatives (Murahashi et al., 2001). Due to the different chemical and physical properties between PPAHs and their derivatives, their deposition rates are likely different (Zhang et al., 2015b). For instance, OPAHs have higher water solubility than their PPAHs and thus are expected to be removed faster from the atmosphere through both wet and dry deposition processes (Albinet et al., 2008; Bidleman, 1988). Apart from the properties of the contaminants, atmospheric concentration, meteorological condition, topography, and emission density would influence the atmospheric deposition of PACs as well (Xing et al., 2016), leading large spatial and temporal variabilities in their deposition amounts.

About eighty percent area of the Three Gorges Reservoir Area (TGRA) is within the jurisdiction of Chongqing, southwest China. Atmospheric emissions of PPAHs in Chongqing reached 684 tons in 2003 (Xu et al., 2006) and were even higher in recent years, which may exert potential threat to the ecosystems in TGRA and the downstream regions of the Yangtze River. The atmospheric PPAHs

levels in Chongqing have been reported to be higher than in many other Chinese cities (Chow et al., 2006; Xu et al., 2006; Zhu et al., 2014), partly due to the unique topographic condition and frequent stagnant air conditions (Tian et al., 2017; Wang et al., 2017). To gain some knowledge of the spatiotemporal patterns of atmospheric deposition of PPAHs and their derivatives and related sources and possible transformation processes, bulk (dry + wet) deposition samples were collected at two different types of locations, one at an urban site and another at a rural site, both in Chongqing. Annual and seasonal deposition fluxes were first quantified, chemical transformation mechanisms were explored using OPAHs to PPAHs ratio approach, and potential sources were identified using principle component analysis. Knowledge gained from the present study will be useful for assessing potential ecological impacts of this group of pollutants and for making emission control policies where needed.

2. Methodology

2.1. Sampling sites and sample collection

The urban site is located in Wanzhou in the northeastern part of Chongqing (Fig. 1). The atmospheric bulk deposition sampler was deployed on the rooftop of a 27 m tall experimental building in the campus of Chongqing Three Gorges University located in the central urban area of Wanzhou. The site represents a mixed residential and commercial urban environment (Wang et al., 2016a; Zhang et al., 2015c).

The rural site is situated in the rural area of Kaizhou distinct. The sampling instrument was set up at the roof of a two-story building, about 10 m above the ground. It's an agricultural area 41 km northeast of the urban site and with low residential density and no industrial activities nearby.

Monthly samples were collected from May 2014 to April 2015 using round stainless steel buckets 60 cm tall and with an inner diameter of 80 cm. The samplers were placed 1.2 m above the rooftop at both sites. Different amounts of purified water (depending on season) and copper sulfate were added to each sampler to reduce the resuspension loss of deposited particles and to prevent algal growth. The average temperature and relative humidity (RH) during sampling period was about 19 °C and 77%, respectively. Rainfall between April and September (wet season) accounted for over 80% of the sampling period.

2.2. Sample preparation and instrument analyses

The deposition samples were filtered through glass fiber filters (Whatman GFFs, 47 mm) that were baked at 450 °C for 4 h and weighed before use. After filtration, GFFs were dried and then

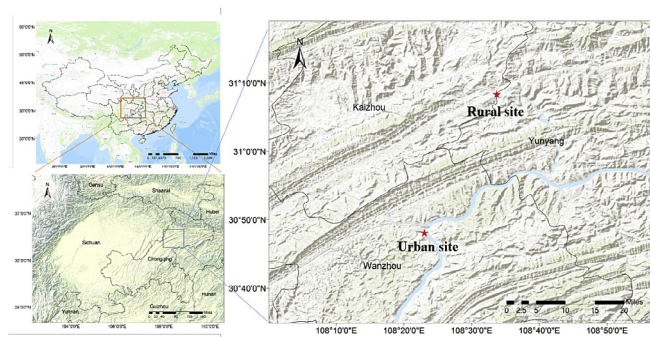


Fig. 1. Locations of sampling sites in Chongqing.

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