Science of the Total Environment 647 (2019) 962–972

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Multimedia modeling of the PAH concentration and distribution in the Yangtze River Delta and human health risk assessment

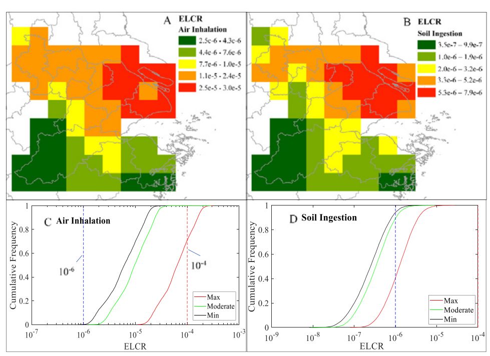
Ying Zhu [a,d,1], Shu Tao [a], Jianteng Sun [b], Xilong Wang [a], Xiangdong Li [c], Daniel C.W. Tsang [c], Lizhong Zhu [b], Guofeng Shen [a], Huijing Huang [a], Chuanyang Cai [a], Wenxin Liu [a,*]

[a] Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China
[b] Department of Environmental Sciences, Zhejiang University, Hangzhou, Zhejiang 310058, China
[c] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China
[d] Lancaster Environment Centre, Lancaster University, Lancaster LA1 4YQ, United Kingdom

HIGHLIGHTS

- ΣPAH16 concentrations were higher in northern regions than in southern regions.
- LMW PAHs were primarily in the air, while MMW PAHs were dominant in other compartments.
- Soil acted as the sink, and the net flux of ΣPAH16 was normally from air to soil.
- PAHs in vegetation mainly originated from the air.
- The probability of significant ELCR by air inhalation and soil ingestion was low.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 April 2018
Received in revised form 2 August 2018
Accepted 5 August 2018
Available online 07 August 2018

Editor: Wei Huang

Keywords:
PAHs
Multimedia modeling
Distribution
Exchange
Health risk assessment
The YRD

ABSTRACT

Emissions of polycyclic aromatic hydrocarbons (PAHs) in China remain at a high level compared to those in developed countries. The Yangtze River Delta (YRD) is an economic and industrial center in China with an extremely large population. The potentially high emissions and excess cancer risk from human exposure in this region cannot be neglected. This study applied a multimedia model to estimate the concentrations of 16 US EPA priority PAHs in the environment in the YRD with a well-developed PAH-emission inventory for 2014. The model predicted that the average concentrations of ΣPAHs were 274 ng/m^3 in the air, 255 ng/g in the soil, 15 ng/g in vegetation, 147 ng/L in freshwater and 144 ng/g in sediment, as well as 99 ng/L and 80 ng/g in seawater and sediment, respectively. Soil is the PAH sink in this region, and the net flux of the total PAHs is always from air to soil for each isomer. A deterministic assessment observed that the ELCR (excess lifetime cancer risk) ranged from 2.5×10^{-6} to 3.0×10^{-5} for exposure by air inhalation and from 3.5×10^{-7} to 7.9×10^{-6} for exposure by soil ingestion. The probabilistic results did not find any probability of ELCR $>10^{-4}$ by exposure via soil ingestion in the YRD. The probabilistic ELCR induced by inhalation exposure varied from 8.1×10^{-7} to 3.1×10^{-4} in the YRD. This study provided a comprehensive overview of PAHs occurrence in natural environments and of the relevant human health risks. The information presented in this study could help authorities to enact a strategy regarding emission reduction and pollution control relevant to PAHs.

* Corresponding author.
E-mail address: wxliu@urban.pku.edu.cn (W. Liu).
[1] Present address: State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China.

https://doi.org/10.1016/j.scitotenv.2018.08.075
0048-9697/

Capsule: Multimedia modeling predicted distributions and compositions of PAHs in different environmental compartments, and deterministic and probabilistic ELCRs induced by air inhalation and soil ingestion were also provided.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a type of carcinogens that come from the incomplete combustion of organic items and exist ubiquitously in the general and occupational environments (Kamal et al., 2015; Shen et al., 2013). Over 100 PAHs may be present in the environment as a mixture, and 16 of them are prioritized by the US EPA according to such factors as their toxicity, potential exposure, and carcinogenicity to human and animals (ATSDR, 2005; Mumtaz et al., 1996). Animal studies and epidemiological evidence have shown an association between exposure to PAHs and excesses of lung, bladder and skin cancers, especially for heavier PAHs (Armstrong et al., 2004; Boffetta et al., 1997; Mastrangelo et al., 1996). Ambient particulate PAHs are found to be linked to heritable mutations in mice (Somers et al., 2004; Somers et al., 2002). Several researchers have found that somatic mutation in human newborns is linked to transplacental exposure to PAHs (Perera et al., 2002). The extensive presence of PAHs possibly leads to frequent contact and a heavy exposure might increase the substantial risk of cancer (Boffetta et al., 1997). Therefore, it is essential to investigate PAH exposure levels and assess the relevant health risks by potential exposure via corresponding exposure routes.

The Yangtze River Delta (YRD) is an important economic and industrial center in China with an extremely high population density. Currently, the emission of PAHs in this area still remains high due to the demand of energy from fossil fuel combustion, such as motor vehicle emissions and biomass burning in rural areas (Shen et al., 2011; Shen et al., 2013; Zhang et al., 2009). Knowledge of the occurrence and environmental fate of PAHs in this region is vital for understanding the potential human health risks induced by exposure to PAHs. The concentrations of PAHs in soil, surface water and sediment in the YRD have been determined individually by different research groups for distinct objectives (Cai et al., 2017; Sun et al., 2017a; P. Wang et al., 2017; M.H. Wu et al., 2017; Zhang et al., 2014; Zhuo et al., 2017). However, these monitoring campaigns only focus on a single compartment or system, and the targeted area is very limited; thus, the entire region cannot be covered. A comprehensive and systematic understanding is required of PAH concentrations in multiple environmental compartments and of the partitioning and transport at the interface of those compartments. In addition, a large number of monitoring campaigns have concentrated on several large cities, such as Shanghai and Nanjing (Kang et al., 2017; Kong et al., 2015; Li et al., 2016; Liu et al., 2017; Zhuo et al., 2017). Multimedia models have been applied to predicting PAH concentrations in different areas in China, such as the Pearl River Delta, Bohai Bay, Hangzhou, Tianjin and some river or lake systems (Lang et al., 2009; Liu et al., 2014; Tao et al., 2003; Wang et al., 2012; Xu et al., 2013), while such models have not yet been applied to the YRD as a whole. Besides, a data gap exists for large regions in this area.

Many studies have estimated the excess cancer risk attributed to PAH exposure via different exposure routes in China. Zhang et al. estimated the lung cancer risk by inhalation exposure in China with predicted PAH concentrations (Zhang et al., 2009). Shen et al. conducted a more intensive estimation of the lung cancer risk globally accounting for individual susceptibility (Shen et al., 2014). However, these analyses are not refined for the YRD. The excess lifetime cancer risk (ELCR) by PAH exposure via soil ingestion has been assessed using measured concentrations in soil for the YRD (Cai et al., 2017). A more refined estimation of ELCR, induced through multiple exposure pathways focusing on the YRD, is essential to provide an extensive risk assessment for this area.

This study has applied a multimedia model to comprehensively estimate the concentration, spatial distribution, and environmental fate of the 16 PAHs in the YRD. Previous studies have applied the model nationally in China to a broad range of chemicals, including benzo[*a*]pyrene, and performed well (Zhu et al., 2018; Zhu et al., 2014; Zhu et al., 2015). The model has also been validated in this study on the YRD. The composition and partitioning of PAHs at steady state have been discussed to reveal the potential reservoir of PAHs in the environment. With predicted concentrations in air and soil, the ELCRs induced by PAH exposure through both air inhalation and soil ingestion have been estimated, respectively. A deterministic assessment has been conducted to show the distribution of the ELCRs. To account for the uncertainty of vital parameters, probabilistic assessment has been performed to investigate the probability of ELCR in the YRD. The results could reflect the complete information on PAH occurrence and storage in this area. The estimated ELCRs could offer a reference for authorities to formulate a strategy on emission reduction and pollution control.

2. Methods and materials

2.1. Study area and chemicals

The YRD covers Shanghai, Zhejiang and Jiangsu provinces in China and contributes >21% of China's GDP (NBSC, 2015). Agriculture and industry are highly developed in this region. The YRD is also one of the regions with the densest population (approx. 156 million) in China (CNSTATS, 2016). Approximately 4.6 million vehicles make emissions from transportation exhaust one of the major sources of PAHs in this region (NBSC, 2015). Burning fuel for heating and cooking is also an important source of PAHs, especially in rural areas. The target region of this study is located in the area between 29.0° and 32.61° N and between 118.33° and 122.83° E (Fig. 1), which is the core area of the YRD. The 16 US EPA priority PAHs were modeled in this study. The PAHs include naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benzo[*a*]anthracene (BaA), chrysene (CHR), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[123*cd*]pyrene (IcdP), dibenzo[*ah*]anthracene (DahA), and benzo[*ghi*]perylene (BghiP).

2.2. Model configuration and validation

SESAMe v3.3 (Sino Evaluative Simplebox-MAMI Model), a steady-state multimedia concentration model, was applied to the selected region of the YRD regionally (Zhu et al., 2016; Zhu et al., 2015). The model was developed to predict the concentration of chemicals in the air, freshwater and sediment, seawater and sediment, vegetation and soils across China with a spatial resolution of 50 km (see Fig. 1). The spatial resolution of the adopted model had to compromise on the coarsest resolution of available environmental parameters compiled for the model, which was the surface water flow rate across China. The model may not capture the hotspots due to the resolution compared with