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Altitudinal and spatial variations of polycyclic aromatic hydrocarbons in Nepal: Implications on source apportionment and risk assessment

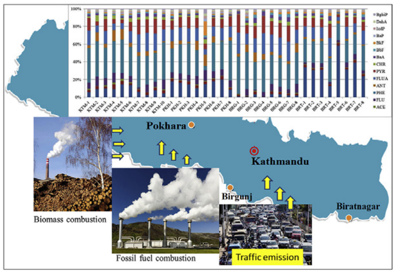
Ishwar Chandra Yadav [a], [b], [*], Ningombam Linthoingambi Devi [c], Jun Li [a], Gan Zhang [a]

[a] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, 510640, PR China
[b] Department of International Environmental and Agricultural Science (IEAS), Tokyo University of Agriculture and Technology (TUAT) 3-5-8, Saiwai-Cho, Fuchu, Tokyo, 1838509, Japan
[c] Centre for Environmental Sciences, Central University of South Bihar, BIT Campus Patna, 800014, Bihar, India

HIGHLIGHTS

- High level of $\sum$PAHs was detected in all four Nepalese cities.
- LMW-PAH concentrations were two folds higher than HMW-PAH concentrations.
- Biomass and coal combustions and vehicular emissions are the main source of PAHs.
- The estimated excess inhalation risk was greater in people living in plain areas than those in hilly areas.

GRAPHICAL ABSTRACT



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ABSTRACT

Although several global/regional studies have detailed the high level of polycyclic aromatic hydrocarbons in urban areas worldwide, unfortunately, Nepal has never been part of any global/regional regular monitoring plan. Despite few sporadic studies exist, the systematic monitoring and integrated concentration of PAHs in urban region of Nepal are lacking. In this study, the concentrations, sources, and health risk assessment of 16 PAHs in air ($n = 34$) were investigated in suspected source areas/more densely populated regions of Nepal. Four potential source areas in Nepal were focused as it was conjectured that urban centers in plain areas (Birgunj and Biratnagar) would possibly be more influenced by PAHs as a result of intense biomass/crop residue burning than those in hilly areas (Kathmandu and Pokhara). The overall concentrations of $\sum_{16}$PAHs ranged from 4.3 to 131 ng/m^3 (median 33.3 ng/m^3). $\sum_{16}$PAH concentrations in plain areas were two folds higher than those in hilly areas. PHE was the most abundant followed by FLUA, PYR, and NAP, which accounted for 36%, 15%, 12%, and 9% of $\sum_{16}$PAHs, respectively. Principal component analysis confirmed that PAHs in highly urbanized areas (Kathmandu and Pokhara) were related to diesel exhausts and coal combustion, while PAHs in less urbanized regions (Birgunj and Biratnagar) originated from biomass and domestic wood combustions. Furthermore, in the urban areas of Nepal, vehicular emission could also influence atmospheric PAHs. The lifetime cancer risk per million populations due to PAH exposures was estimated to be higher for plain areas than that for hilly areas, suggesting a relatively greater risk of cancer in people living in plain areas.

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[*] Corresponding author. Department of International Environmental and Agricultural Science (IEAS), Tokyo University of Agriculture and Technology (TUAT) 3-5-8, Saiwaicho, Fuchu Tokyo 1838509, Japan.
E-mail address: icyadav.bhu@gmail.com (I.C. Yadav).

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of semi-volatile organic compounds (SVOCs) (Ollivon et al., 1999; Wang et al., 2015; USEPA, 2017). Depending on the number of aromatic rings present in PAHs, they are commonly classified into low-molecular weight (LMW) PAHs (containing three or fewer aromatic rings) and high-molecular weight (HMW) PAHs (containing four or more aromatic rings) (CCME, 1999). PAHs are carcinogenic, genotoxic, and ubiquitous and are generated during the incomplete combustion of wood/grass/coal and fossil fuel and vehicular emission and industrial coking (Baek et al., 1991; Zhang and Tao, 2009; Shen et al., 2013; Mugica Alvarez et al., 2015). Sixteen PAHs have been categorized as priority pollutants by the United States Environmental Protection Agency (USEPA) on the basis of their toxicity, potential for human exposure, frequency of occurrence at hazardous waste sites, and extent of available information (ATSDR, 2005; NTP, 2005). The incomplete combustions of coal, biomass burning, and motor vehicle emissions are the primary reasons for the universal occurrence of PAHs in air (Zhang et al., 2009). Because PAHs are the most suspected carcinogenic pollutants, exposure to these compounds in air has been broadly studied (IARC, 1987; WHO/IPCS, 1998; Bostrom et al., 2002). In 2007, it was estimated that the worldwide emission of 16 PAHs was approximately 500 Gg, of which 60.5% originated from residential/commercial biomass burning, while 12.8% was released from motor vehicles (Shen et al., 2013).

The passive air sampling (PAS) proposed by Shoeib and Harner (2002) is an inexpensive alternative method to the regular active air sampling (AAS) for the measurement and monitoring of atmospheric pollutants including PAHs on a large scale, where electricity is inaccessible (Jaward et al., 2004; Zhang et al., 2008). In recent decades, polyurethane foam (PUF) disks mounted in PAS have been widely utilized to supplement the AAS under different regional and global air monitoring programs (Jaward et al., 2004; Klanova et al., 2008; Hung et al., 2010; Devi et al., 2011, 2014; Yadav et al., 2017a, b). This capability of PUF-PAS makes it an appropriate tool for sampling airborne pollutants in a remote area with high spatial resolution (Hayward et al., 2010). PUF-PAS is a reliable method and can accomplish a semi-quantitative comparison of atmospheric levels and patterns of persistent organic pollutants (POPs) on a large scale. Additionally, the PUF-PAS is useful in distinguishing source/sink regions together with temporal trends by long-term monitoring (Jaward et al., 2004). The disadvantage of PAS is that it is difficult to determine the volume of air tested at each site, particularly when the sample volume varies with wind speed and the octanol-air partition coefficient (Zhang et al., 2013). However, PAS allows the instantaneous and integrated sampling of environmental pollutants at many locations by deploying from few days to months. This benefits the analysis of fluxes, transport, sinks/sources, and trends in atmospheric pollutants. In addition, by using multiple PAS samplers, one can identify the hot spots and potential sources of air pollutants.

The atmospheric PAH concentrations have been extensively studied in urban areas worldwide because of the high population density, abundance of motor vehicles, and substantial industrial activities present therein. Evidence suggests that elevated levels of PAHs are usually seen in densely populated urban regions (Hafner et al., 2005), thereby posing adverse effects on human health (Cotham and Bidleman, 1995; Motelay-Massei et al., 2005; Ravindra et al., 2008; Peverly et al., 2015). Urban areas tend to be the focal point of several industrialization activities, which emit different chemicals into the atmosphere that are transported and then deposited downwind in potentially sensitive environments, for instance, soil and sediments (Totten et al., 2004). Because of the high toxicological impacts of PAHs and their well-known carcinogenic and mutagenic properties, incredible endeavors have been made in the recent years to investigate atmospheric PAHs in urban areas.

A few global and regional air monitoring programs, for instance, Global Monitoring Plan (GMP), European Monitoring and Evaluation Programme (EMEP), and Asian Soil and Air Monitoring Program (Asia-SAMP), have been established to monitor the progress of regulatory measures to reduce or eliminate the emissions of SVOCs, under the protection of the Stockholm Convention (SC) on POPs and the Aarhus Protocol on POPs under the Convention on Long-Range Transboundary Air Pollutants (UN/ECE, 2010; UNEP, 2011; Hong et al., 2016). Unfortunately, Nepal has never been part of any global or regional consistent monitoring plan, and no systematic monitoring campaign for organic pollutants has been conducted so far. Although few previous studies reported elevated concentrations of distinctive POPs including PAHs (Yadav et al., 2017a, b; Kishida et al., 2009; Chen et al., 2015, 2016; 2017), detailed information regarding the environmental fate and the sources of PAH emission in the urban areas of Nepal is either obscure or little is known. Therefore, understanding the contamination level of PAHs in Nepalese air and identification of their sources of emission are required to highlight the potential contribution of organic compounds to the territorial and worldwide PAH levels. In this study, an aggregate of 34 PUF-PAS samplers were used in four noteworthy urban areas of Nepal (Kathmandu, Pokhara, Birgunj, and Biratnagar) to investigate the environmental concentration, fate, and sources of PAH emission in Nepalese air. Furthermore, human inhalation exposures to PAHs were assessed to anticipate the potential impact on general population through inhalation of PAHs.

2. Materials and methods

2.1. Passive air sampling

PAS was conducted in four major cities of Nepal (Kathmandu, Pokhara, Birgunj, and Biratnagar) covering residential, commercial, public places, office premises, industrial area, high traffic area, occupational area, and nearby airport (Table S1). The details about geographical location and sampling sites are discussed elsewhere (Yadav et al., 2017a). Before deployment, PUF disks (diameter 14 cm, thickness 1.3 cm, and density 0.017 g/cm^3) were cleaned by soxhlet extraction using a mixture of acetone and dichloromethane (DCM) (1:1 V/V) at 100 °C and 1500 psi for 15 min. The PUF disks were then wrapped in an aluminum foil, sealed in a Teflon bag, and transported to the sampling sites for deployment. PUF-PAS was deployed at a preselected location in each city following the standard protocol proposed by Zhang et al. (2008). After 60 days of deployment, all the PUF disks were retrieved, wrapped in an aluminum foil, sealed, transported back to wet chemistry laboratory of Guangzhou Institute of Geochemistry, and stored at −20 °C until analysis.

2.2. Extraction and cleanup

All the PUF disks were soxhlet extracted with DCM for 24 h. Before extraction, the PUF disks were spiked with 5 μL (200 μg/L) of deuterated PAHs (acenapthene-d10, phenanthrene-d10, chrysene-d12, and perylene-d12) as recovery standard. Small granules of activated copper were added to the collection flask to remove the elemental sulfur. The copper granules were prewashed and activated with hydrochloric acid before adding into the collection flask. The extract was then concentrated by rotary evaporation and was solvent exchanged to hexane. The extract was purified by an