Science of the Total Environment 556 (2016) 12–22

Contents lists available at ScienceDirect

Science of the Total Environment

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

Polycyclic aromatic hydrocarbons in soils from the Central-Himalaya region: Distribution, sources, and risks to humans and wildlife

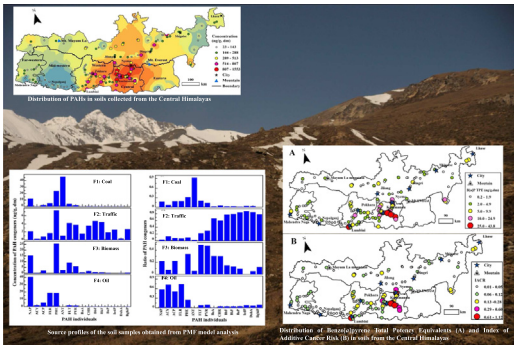
Xiang Bi [a,b,1], Wei Luo [a,*,1], Jiajia Gao [a,i], Lan Xu [a], Junming Guo [c], Qianggong Zhang [c,d], Kumar Y. Romesh [e], John P. Giesy [f], Shichang Kang [g,d], Jacob de Boer [h]

[a] State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
[b] Graduate University of Chinese Academy of Sciences, China
[c] Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences (CAS), Beijing 100085, China
[d] CAS Center for Excellence in Tibetan Plateau Earth Sciences, Chinese Academy of Sciences, Beijing 100101, China
[e] Department of Environmental Science and Engineering, School of Science, Kathmandu University, Kathmandu 6250, Nepal
[f] Department of Veterinary Biomedical Sciences and Toxicology Centre, University of Saskatchewan, Saskatoon, Canada
[g] State Key Laboratory of Cryospheric Sciences, Cold and Arid Regions Environmental and Engineering Research Institute, Chinese Academy of Sciences, Lanzhou 730000, China
[h] Institute for Environmental Studies, VU University, De Boelelaan 1087, 1081 HV Amsterdam, The Netherlands
[i] Tibet Climatic Center, Lhasa 850001, China

HIGHLIGHTS

- PAH concentrations in soils were presented in a unique area of the Himalayas.
- Altitude was a significant factor controlling PAHs in mountain soils.
- Sources of PAHs in soils were identified by positive matrix factorization analysis.
- 39% of soils had a slight risk to wildlife and environment of the Central Himalayas.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 29 November 2015
Received in revised form 2 March 2016
Accepted 2 March 2016
Available online xxxx

Editor: F. Riget

Keywords:
Tibetan Plateau
Nepal
China

ABSTRACT

The Central Himalayas are not only a natural boundary between China and Nepal but also a natural barrier for transport of air masses from South Asia. In this study, 99 samples of surface soil were collected from five regions of Nepal on the southern side of the Central Himalayas, and 65 samples of surface soil were obtained from the northern side on the edge of the Tibetan Plateau, China (TPC). Concentrations of polycyclic aromatic hydrocarbons (PAHs) in soils were measured to determine their distribution, potential for accumulation, and sources, as well as risks to humans and the environment. Mean concentrations of Σ_{16}PAHs were 2.4×10^2 and 3.3×10^2 ng/g dry mass (dm) in soils collected from the TPC and Nepal, respectively. Significant correlations between concentrations of lower molecular weight PAHs (LMW-PAHs) in soils and altitude were found. Total organic carbon (TOC) in soil was positively but weakly correlated with concentrations of PAHs in the study area, which suggested little role of TOC in adsorption of PAHs. The cities of Kathmandu and Pokhara in Nepal and Nyemo (especially Zhangmu Port), Shigatse, and Lhasa on the TPC, were areas with relatively great

* Corresponding author.
E-mail address: luow@rcees.ac.cn (W. Luo).
[1] Wei Luo and Xiang Bi contributed equally to this work.

http://dx.doi.org/10.1016/j.scitotenv.2016.03.006

PMF model
Toxicity assessment
Mountain

concentrations of PAHs in soils. The main sources of PAHs identified by positive matrix factorization were emissions from motor vehicles and combustion of coal and biomass in the Central Himalayas. Calculated total benzo[*a*]pyrene potency equivalents of 0.23–44 ng/g dm and index of additive cancer risk of 3.8×10^{-3}–9.2×10^{-1} indicated that PAHs in almost all soils investigated posed *de minimis* risk of additional cancer to residents via direct contact and had no significant risk of additional cancers through consumption of potable water. Mean risk quotient values indicated that 39% of soils had a slight risk to wildlife and the ambient environment of the Central Himalayas.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of pollutants that are widely distributed, persistent and toxic and have potential for bioaccumulation. They can migrate to remote alpine regions via long-range, atmospheric transport (LRAT) and be deposited in environments due to orographic, cold-trapping effects (Choi et al., 2009; Wania and Westgate, 2008). Accumulation of PAHs in cold environments might affect alpine ecosystems or pose risks to health of people relying on alpine ecosystems for food and water (Vives et al., 2004; Fernández et al., 2002).

In terrestrial environments, soils act as a sink for PAHs. More than 90% of the total mass of PAHs in the environment is stored in soils (Wild and Jones, 1995). In warmer climates, soil can contribute PAHs to the atmosphere (Dalla Valle et al., 2005). PAHs in soils might be further deposited on or accumulated into vegetables or other biota via food chains (Li et al., 2008). Further, leaching of PAHs from soils can contaminate groundwater (Bortey-Sam et al., 2014). Therefore, monitoring concentrations of PAHs in soils is important for assessing their potential effects on the environment or humans.

Investigation of patterns of accumulation of PAHs along altitudinal gradients can elucidate factors affecting their fates in mountainous environments. The Central Himalayas are a geographical zone separating India, Pakistan, and Nepal in South Asia from China. The Tibetan plateau, China (TPC) is at the northern side of the Central Himalayas, and it is also an important source of water for surrounding regions. Although local emissions of PAHs in the TPC are limited, it is surrounded by the most densely populated and rapidly industrializing countries such as India and China. Emission of ΣPAHs in China was estimated to be 25,300 tons in 2003. China and India had the greatest emissions of PAHs to the atmosphere in 2004 (Xu et al., 2006; Zhang and Tao, 2009). During the Indian monsoon, PAHs are transported and deposited to various environmental matrices, including soil, grass, snow and ice, and sediments on the TPC (Wang et al., 2007; Wang et al., 2008; Wang et al., 2010; Li et al., 2011). In recent years, emissions of PAHs in Nepal have increased due to growth of local population as well as development of highways and additional usage of motor vehicles. Furthermore, valleys of the Himalayas in Nepal are channels for transport of PAHs from the Indian subcontinent to the TPC (Yang et al., 1987). Thus, the Himalayas are considered to be an ideal region for investigating the LRAT of PAHs. Thus far, few studies have investigated emissions of PAHs from the Central Himalayas (Aichner et al., 2007; Guzzella et al., 2011; Chen et al., 2015).

Investigations of spatial distribution and sources of PAHs in soils from the Central-Himalaya region are important for understanding cross-border migration of PAHs between South Asia and the TPC, factors influencing accumulation of PAHs, and assessment of their potential effects on health people residing in this relatively pristine area. In this study, concentrations of 16 indicator PAHs specified by the US Environmental Protection Agency (EPA) were measured in 164 samples of soil collected from both sides of the Central Himalayas on the TPC and in Nepal. The information was used to determine the potential sources of PAHs, the factors influencing their accumulation, as well as risks for humans and the environment.

2. Materials and methods

2.1. Collection of samples

Distribution of PAHs in Nepal was determined by collecting samples of soil along a West-to-East transect. Soils were also collected from the TPC, where PAHs might be transported via monsoon and atmospheric circulations and (Fig. 1). Nine samples of soil from a subarea in far-western Nepal mountain area where, as suggested by Wang et al. (2014), sparse PAHs emission and human activities existed, were selected to investigate effects of altitude on accumulation of PAHs in soils (Table 1 and Fig. 1B). The climate in the studied regions is dominated alternatively by the Indian monsoon and westerly winds (Wang et al., 2013). In total, 164 soil samples (99 from Nepal and 65 from the TPC) were collected from remote sites (i.e., away from towns, roads, or other human activities) on the two sides of the Himalayas between 2013 and 2014, using a stainless steel spade (Fig. 1). Among soils from Nepal, 36, 27, 17, 17, and 2 samples of soil were collected from the central, western, mid-western, far-western, and eastern parts of Nepal, respectively. Newly fallen leaves were discarded before soils were collected. Three or four surface samples (0–5 cm) were obtained over an area of approximately 100 m^2 from each part, and then composited to form a representative sample of soil. Samples were wrapped in aluminum foil, kept in the dark, and transported to the laboratory and maintained at −20 °C.

2.2. Sample extraction and analysis

Soils were air-dried, sieved through 2-mm mesh, and spiked with chemical surrogates (four deuterated PAHs, i.e., naphthalene-d8 (NAP-d8), acenaphthene-d10 (ACP-d10), phenanthrene-d10 (PHE-d10), and chrysene-d12 (CHR-d12)) for calculating recoveries. Next, 16 PAHs, including naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACP), fluorene (FLR), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLT), pyrene (PYR), benz[*a*]anthracene (BaA), chrysene (CHR), benz[*b*]fluoranthene (BbF), benz[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[1,2,3-*cd*]pyrene (IcdP), dibenz[*a*,*h*]anthracene (DahA), and benzo[*ghi*]perylene (BghiP), were identified and quantified using an Agilent 6890 gas chromatograph equipped with a HP-5MS capillary column (30 m × 0.25 mm i.d × 0.25 μm film thickness) for separation and an Agilent 5975 mass spectrometer with an electron impact ionization source as a detector. Further information about the extraction and analysis methods is provided in Text *SI*-1. Samples of soils were treated with 1 mol/L HCl to remove inorganic carbon. Subsequently, they were cleaned with Milli-Q water until neutral pH (7 ± 0.2) and dried at 80 °C. The determination of total organic carbon (TOC) was performed by flash combustion at 1025 °C followed by thermal conductivity detection in a Vario TOC. The limit of detection was 0.1%.

2.3. Quality assurance/quality control

Interferences were checked by analyzing cross-contamination and stability of the instrument during the entire analytical process, procedural blanks, and spiked blank samples with each block of 10 samples.