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Concentration and spatial distribution of organophosphate esters in the soil-sediment profile of Kathmandu Valley, Nepal: Implication for risk assessment



Ishwar Chandra Yadav ^{a,d,*}, Ningombam Linthoingambi Devi ^b, Jun Li ^a, Gan Zhang ^a, Adrian Covaci ^c

^a State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, PR China

^b Centre for Environmental Sciences, Central University of South Bihar, BIT Campus Patna, 800014, Bihar, India

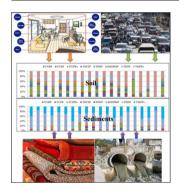
^c Toxicological Centre, University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Belgium

^d Department of International Environmental and Agricultural Science (IEAS), Tokyo University of Agriculture and Technology (TUAT) 3-5-8, Saiwai-Cho, Fuchu-Shi, Tokyo 1838509, Japan

HIGHLIGHTS

- Elevated level of \sum OPEs was detected both in sediments and soil samples.
- TMPP was most abundant OPE in soil while TEHP exceed in sediments.
- High level of OPEs in soil released from consumer materials and intensive traffic areas
- Domestic sewage discharge and effluent from carpet industry was recognized as sources of OPE in sediments.
- Dermal absorption of OPEs via soil is the primary pathway of human exposure.

G R A P H I C A L A B S T R A C T



A R T I C L E I N F O

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ABSTRACT

Despite the fact that soil and sediments, which act as a sink or potential source of organic pollutants, have been polluted with organophosphate esters (OPEs) around the globe, extremely constrained data is accessible on environmental concentration and fate of OPEs in solid matrices in whole of the South Asia particularly if there should be an occurrence in Nepal. In this study, surface soil (N = 19) and sediments samples (N = 20) were analyzed for eight different OPE in Kathmandu Valley during October 2014. The concentration of \sum_{8} OPE measured in sediments samples was 12 times higher than soil and ranged 983-7460 ng/g dw (median 2210 ng/g dw) and 65-27,500 ng/g dw (186 ng/g dw), respectively. TMPP was most abundant in soil followed by TCIPP, TEHP and EHDPHP and ranged 17-25,300 ng/g dw (41.3 ng/g dw), 11.2-911 ng/g dw (31.7 ng/g dw), 8.52-858 ng/g dw (26.1 ng/g dw) and 10.2-114 ng/g dw (25.6 ng/g dw), respectively. TEHP was most prevalent in sediments followed by TMPP and EHDPHP and were in the range of 657–3020 ng/g dw (median 1140 ng/g dw), 267–2630 ng/g dw (median 815 g/g dw), 34-418 ng/g (median 131 ng/g dw), respectively. The sources of the high level of OPEs in soil was related to the end point use of consumer materials, traffic emission, and close proximity to commercial and industrial areas; while domestic sewage discharges and effluents from carpet industry were identified as the possible entry of OPE in sediments. Total organic carbon (TOC) and black carbon (BC) content in soil were moderately and positively correlated with \sum_8 OPE indicating more or less influence of soil organic carbon. The health risk assessment suggested dermal absorption of OPEs via soil is the primary pathway of human exposure to the general

* Corresponding author.

E-mail address: icyadav.bhu@gmail.com (I.C. Yadav).

population. The significantly high-risk quotient (RQ) estimated for \sum_{8} OPEs especially TMPP and TPHP suggested significant potential adverse risk for aquatic organisms.

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1. Introduction

Organophosphate esters (OPEs) are a group of important chemicals normally utilized as flame retardants (FRs) and plasticizers in different types of consumer products such as varnishes, polyurethane foam, hardware gear, upholstery, and textile materials (USEPA, 1976, 2005; van der Veen and de Boer, 2012). For the most part, chlorinated OPEs such as tris (2-chloroethyl) phosphate (TCEP), tris (2-chloro-isopropyl) phosphates (TCPPs), and tris (1,3-dichloro-2-propyl) phosphate (TDCIPP), are specially utilized as flame retardants, while non-chlorinated OPEs such as tributyl phosphate (TNBP), and triphenyl phosphate (TPHP), are used as plasticizer in added substances. Moreover, they are also utilized as uttermost pressure supplement and anti-wear agent in hydraulic fluid, lubricator, transmission liquids, engine oils, and other comparable items (Solbu et al., 2007; Regnery et al., 2011; Westerdahl et al., 2011).

Due to ban and limitation on the use of penta- and octa-brominated diphenyl ethers (PBDEs), the utilization of OPEs as flame retardants have been increased since to meet merchant needs. In recent year, OPEs are recognized as high-end product chemicals (Reemtsma et al., 2008; Stapleton et al., 2011; Mihajlovic et al., 2011). Generally, the OPEs are specifically mixed into the materials and are not chemically attached; hence, they can be easily released or drained into the surrounding environment during production, application, and/or transmission of OPE-comprised materials (Marklund et al., 2003). Because of low water solubility, high adsorption potential and persistent behavior, some of the OPEs specifically chlorinated phosphorus chemicals gets accumulated in different environmental matrices, for instance, soil and sediments (Marklund et al., 2005; Chen and Bester, 2009; Chung and Ding, 2009; García-López et al., 2009a, 2009b; Cao et al., 2012). The low water soluble-OPEs such as tricresvl phosphate (TMPP) and tri (2-ethylhexvl) phosphate (TEHP) can strongly be adsorbed on the organic particulate matter, precipitate and/or may be gathered together with suspended solids in the bottom of the sediments (Martínez-Carballo et al., 2007). Likewise, TPHP may also be adsorbed onto sediments quickly as soon as released into water bodies (WHO, 1997; Chung and Ding, 2009; García-López et al., 2009a; Martínez-Carballo et al., 2007). Furthermore, OPE chemicals are more stable and resistant to biodegradation; particularly the chlorinated OPEs are not biodegradable and are believed to be a persistent organic pollutant (Kawagoshi et al., 2002; Meyer and Bester, 2004; Bollmann et al., 2012).

Soil comprised mostly of organic matter and mineral content is the major terrestrial ecological reservoir and one of the major sink for the different class of organic chemicals including OPEs. OPEs may find their way into the soil after the use of wastewater for irrigation, sewage sludge application, land dissipation of flame retardant plastics, leakage of hydraulic fluid, dry and wet deposition of aerosolized and semi-volatile phosphorus compounds, and flash flood of river (Muir, 1984; Stachel et al., 2007; Passuello et al., 2010). In addition, precipitation may also favor the deposition of some group of phosphorus chemicals in soil and sediments from the air through water wash mechanism (Laniewski et al., 1998; Fries and Puttmann, 2003; Mihajlovic et al., 2011). Low water-soluble potential together with the relatively high octanol-water partition coefficient ($\log K_{ow}$) of OPE chemicals make them accumulate in soil and sediments (USEPA, 2003). Toxicological studies of OPEs indicated the majority of them are highly toxic (Reemtsma et al., 2008). For instance, TPHP and TNBP most significantly inhibit the function of human blood monocyte carboxylesterase (Saboori et al., 1991) and have neurotoxic impacts (Reemtsma et al., 2008). TPHP and TNBP, are known to be neurotoxic (WHO, 1991), while TCEP, TDCIPP, and TCIPP are carcinogenic (European Commission, 2001; Reemtsma et al., 2008). Likewise, the chlorinated-OPEs, such as TCEP and TCIPP are known to cause cancer in human beings (Reemtsma et al., 2008).

Worldwide application of these highly persistent phosphorus chemicals led to widespread environmental contamination, accumulation, and global distribution in different matrices of the environment including soil and sediments. Following this, the European Commission categorized them under the list of aquatic priority pollutants (European Commission, 2002). In recent year, the monitoring of phosphorus chemicals in the different ecological matrix has become widely debated environmental research (Regnery and Püttmann, 2010; Bollmann et al., 2012; Moller et al., 2012). As of now, OPE has been documented worldwide in varieties of environmental samples, including indoor air and dust (Stapleton et al., 2009; Takigami et al., 2009; Tollback et al., 2010; Van de Eede et al., 2011; Yadav et al., 2017), wastewater (Martínez-Carballo et al., 2007; Chen and Bester, 2009), surface water (Reemtsma et al., 2008; Martínez-Carballo et al., 2007; Bacaloni et al., 2008; Regnery and Püttmann, 2010), groundwater (Regnery et al., 2011), and living materials (Meyer and Bester, 2004; Shah et al., 2006; Schindler et al., 2009). However, little is known regarding contamination and sources of OPE in soil (Mihajlovic et al., 2011; Wan et al., 2016; Cui et al., 2017a, 2017b; Zheng et al., 2017), and sediments environment (Martínez-Carballo et al., 2007; Chung and Ding, 2009; García-López et al., 2009a, 2009b; Cristale et al., 2013; Cao et al., 2017).

Despite the fact that soil and sediments, which act as a sink or potential source of organic pollutants, have been observed to be polluted with OPEs in different area around the world, very limited information is available on environmental contamination, fate and sources of OPEs in whole of South Asia and more specifically in case of Indian subcontinent including Nepal. Hence, this study aims to investigate the environmental concentration, sources, and fate of 8 OPEs in soil-sediment matrices. Additionally, risks of OPEs exposure to the general population through different pathways and ecological risk were estimated to predict the intake of OPEs. This study will provide preliminary information about the contamination level and fate of OPEs in the soil-sediment matrices of Nepal, located between world's two most populous countries India and China.

2. Materials and methods

Detailed descriptions of the materials and methods are given in Supporting Information part.

2.1. Soil sampling

A total of 19 surface soils from 19 different locations in Kathmandu Valley, the capital city of Nepal (Table S1, Supporting information) were collected in October 2014. The details about sampling locations and the study site are given in SI. Each soil sample was a composite of 3 sub-samples collected in the radius of 5 m in the different direction. The soil samples were then wrapped in aluminum foil, packed into sealed polythene bags and transported to laboratory keeping in the ice bag. The soil samples were freeze dried, ground to powder, homogenized, and sieved through 500 μ m sieves, and stored at -20 °C until analysis.

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