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Environmental concentration and atmospheric deposition of halogenated flame retardants in soil from Nepal: Source apportionment and soil-air partitioning[☆]

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ABSTRACT

While various investigations have been driven on polybrominated diphenyl ethers (PBDEs) and other flame retardants (FRs) in different framework around the world, information about contamination and fate of PBDEs and other FRs in developing countries especially in the Indian subcontinent is uncommon. Nepal being located in the Indian subcontinent, very little is known about contamination level of semi-volatile organic pollutants discharged into the environment. This motivated us to investigate the environmental fate of halogenated flame retardant (HFRs) in Nepalese condition. In this study, we investigated the concentration, fate, and sources of 9 PBDEs, 2 dechlorane plus isomers (DPs), and 6 novel brominated flame retardants (NBFRs). Moreover, air-soil exchange and soil-air partitioning were also evaluated to characterize the pattern of air-soil exchange and environmental fate. In general, the concentrations of NBFRs in soil were more prevalent than PBDEs and DPs, and accounted 95% of Σ HFRs. By and large, the concentrations of NBFRs and DPs were measured high in Kathmandu, while PBDEs level exceeded in Pokhara. Principal component analysis (PCA) study suggested contributions from commercial penta-, octa-, and deca-BDEs products and de-bromination of highly brominated PBDEs as the significant source of PBDEs. Likewise, low f_{anti} ratio suggested DPs in soil might have originated from long-range atmospheric transport from remote areas, while high levels of decabromodiphenyl ethane (DBDPE) in soil were linked with the use of wide varieties of consumer products. The estimated fugacity fraction (f) for individual HFR was quite lower (<0.05) than equilibrium value, suggesting that deposition and net transport from air to the soil is overwhelming. Soil-air partitioning study revealed neither octanol-air partition coefficient (K_{OA}) nor black carbon partition coefficient (K_{BC-A}) is an appropriate surrogate for soil organic matter (SOM), subsequently, absorption by SOM has no or little role in the partitioning of HFRs.

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1. Introduction

Halogenated flame retardants (HFRs) are semi-volatile organic compounds (SVOCs), which are widely utilized as part of many

domestic and industrial products to counteract fire (WHO, 1997; Tan et al., 2007; Lagalante et al., 2009). Some of their usages in consumer products include electronic casings, synthetic textiles, thermoplastics, foams, synthetic resins and glutinous materials (WHO, 1997; Covaci et al., 2003; Stapleton et al., 2012). Since 1970, PBDEs have been broadly utilized and recognized worldwide as critical contaminants (de Wit et al., 2006; Brits et al., 2016; Yu et al., 2016). PBDEs are likely to adhere to solids (e.g. soil, sediment and particles in the air) and may bio-accumulate after their discharge into the environment (Yun et al., 2008; Cetin, 2014).

Traditionally, PBDEs had been sold in three major commercial formulations known as penta-BDE, octa-BDE and deca-BDE, each of

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which contain a scope of the 209 mono-through deca-brominated congeners (La Guardia et al., 2006). Specific congeners have dominated within the commercial mixtures and are commonly investigated in environmental samples as a marker of the individual compound. More specifically, the tetra-brominated (BDE-47), penta-brominated (BDE-99 and -100), and hexa-brominated (BDE-153 and -154) congeners are representative of penta-BDE formulation, while octa-BDE products are shown by hepta-brominated (BDE-183) and deca-BDE products by the deca-brominated congener (BDE-209). The use of penta- and octa-BDE formulation have been prohibited in many jurisdictions (EU, 2003; NICNAS, 2007; Stapleton et al., 2012) while the most common compounds such as tetra-through hepta-brominated congeners are listed as persistent organic pollutants (POPs) under the United Nations Stockholm Convention 2009 (UNEP, 2009). Recently, considering the recommendation of the Persistent Organic Pollutants Review Committee, the 8th meeting of the Conference of the Parties to the Stockholm Convention held in Geneva, Switzerland during 24 May–5 May 2017 listed commercial deca-BDE too as POPs.

Toxicological reports suggested many adverse impacts on human and animal due to exposure to high concentration of PBDEs ranging from endocrine disruption to neurodevelopmental toxicity (Berghuis et al., 2015; Linares et al., 2015). The widespread environmental occurrence, as well as their dangerous health impact, prompted the worldwide ban on PBDEs and intentional withdrawals in many jurisdictions. The substitution of PBDEs by novel brominated flame retardants (NBFRs) has, be that as may be, displayed a large number of an indistinguishable poisonous attribute from PBDEs and seems to have similar environmental fate. Some of the most widely recognized NBFRs comprise deca-bromodiphenyl ethane (DBDPE), which replaced deca-BDE, bis(2,4,6-tribromophenoxy)ethane (BTBPE) was introduced in place of octa-BDE, and bis(2-ethylhexyl) tetrabromophthalate (TEBP) and 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (TBB) are substituents of penta-BDE (Covaci et al., 2011; Ezechias et al., 2014). Hexabromobenzene (HBB), and 2,3,4,5,6-pentabromoethylbenzene (PBEB) are broadly utilized flame retardants supplanting PBDEs in the wide range of polymers (Covaci et al., 2011; Ezechias et al., 2014). Dechlorane Plus (DP) is a chlorinated flame retardant that has been utilized as apart of coating electrical wires and cables, computer connector's fire prevention and numerous manufactured items, for example, nylon or plastic like polypropylene (Hoh et al., 2006; OxyChem, 2007). They are norbornylene derivatives that show flame-retardant and persistent properties similar to PBDEs. Syn- and anti-DP isomers are the significant constituents of technical DPs and have been used in replacement of mirex (Hoh et al., 2006).

Soil composed of organic and inorganic materials is an important environmental matrix for studying fate and behavior of POPs. It assumes an essential part in the dissemination and fate of POPs. Because of wide assimilation capacity of soil, it may serve as a significant supplier and sinks for POPs (Wang et al., 2012; Cetin, 2014). Likewise, it is also a secondary emission source for many SVOCs to groundwater, surface water, and the atmosphere (Tao et al., 2008). Soil quality has been known as a good indicator of environmental contamination and health hazard to human (Meijer et al., 2001). Soil contaminated by PBDEs and NBFRs likewise form potential exposure pathways for humans. While various investigations have been led on PBDEs and other flame retardants in different framework around the world, data, especially on the concentration of PBDEs and other BFRs in developing countries particularly in Indian subcontinent is rare (Eguchi et al., 2013; Zehra et al., 2015; Li et al., 2016a). Such data are extremely pertinent to the worldwide interest for information on the level of SVOCs discharged into the environment. Moreover, information about the

human health impact of HFRs in urban soil is lacking. Although, few past studies revealed serious contamination of brominated- and organophosphate-flame retardant in the air of Nepalese cities (Yadav et al., 2017a,b), very little is known about environmental fate and distribution trends of HFRs in the soil. It was hypothesized that the urban soil in Nepal might also been contaminated with high level of HFRs as they are not regulated in Nepal. This study aims to explore the contamination level, fate, spatial dissemination and sources of HFRs in urban soil of Nepal. Besides, air-soil exchange and soil-air partitioning were assessed to comprehend the influence of soil organic carbon (SOC) and black carbon (BC) in the distribution of HFRs.

2. Materials and methods

2.1. Soil sampling

About 50 g of surface soil was collected from 4 major cities (i.e. Kathmandu, Pokhara, Birgunj, and Biratnagar) of Nepal during October 2014. Details about study sites and sampling locations are well described in our previous study (Yadav et al., 2017c). Soil samples (vegetation removed) were collected at depth of 0–15 cm using stainless steel shovel designed for the purpose. Details about soil sampling and pretreatment process are discussed elsewhere (Yadav et al., 2016, 2017c). Briefly, a total of 72 surface soil samples were collected and mixed to make 24 representative samples. In this way, each sample was a composite of at least 3 subsamples collected in a different direction at distance of 5 m. After collection, subsamples were properly mixed, wrapped in aluminum foil and packed in a zipper bag. Later, all the soil samples were transported to Organic Geochemistry Laboratory of Guangzhou Institute of Geochemistry, Guangzhou, China where they were stored at -20 °C until analysis.

2.2. Determination of TOC and BC

The total organic carbon (TOC) in soil was determined as per method suggested by Ramu et al. (2010). Briefly, about 2–3 g of dried, sieved and homogenized soil was treated with 3 mL of 10% HCl to remove inorganic carbon, washed thrice with Milli-Q water and dried overnight in an oven at 45 °C. A portion of the soil was used to determine TOC. The TOC analysis was performed with Elemental Carbon-Hydrogen-Nitrogen Analyzer (Elementar VARIO EL III). The analysis of BC in soil sample was done following chemothermal oxidation (CTO-375) technique described elsewhere (Gustafsson et al., 2001; Elmquist et al., 2008). In brief, 2–3 g of dried, sieved and homogenized soil was burnt in a muffle furnace to thermal oxidation (375 °C, 18 h) under constant air flow. The soil was then treated with 1 N HCl to remove inorganic carbon. Later, the residual organic carbon in soil was analyzed as BC using Elemental Carbon-Hydrogen-Nitrogen Analyzer (Elementar VARIO EL III).

2.3. Extraction and GC-MS analysis

About 10 g of freeze-dried and properly homogenized soil was spiked with 20 ng of PCB 209 as recovery standard and soxhlet extracted for 24 h with dichloromethane (DCM). The extract was concentrated to a final approximate volume of 2–3 mL by rotary evaporator. The extract was then purified by multilayer silica gel/alumina column followed by anhydrous sodium sulfate. The column was pre-rinsed with 30 mL of hexane and eluted with around 50 mL mixture of DCM/hexane (1:1 vol). The eluent was then concentrated to 0.2 mL under the gentle nitrogen stream. Around 25 µL of isoctane was added to the GC vial as a solvent keeper.

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