



Concentrations of polybrominated diphenyl ethers and alternative flame retardants in surface soils and river sediments from an electronic waste-processing area in northern Vietnam, 2012–2014



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HIGHLIGHTS

- PBDE and alternative FR concentrations were determined in the environment for 3 years.
- Contamination by PBDEs remains high in the study area.
- Concentration of DPs and oligomeric PFRs increased during the study period.
- Contaminating constituents might have changed owing to the international regulations.

ARTICLE INFO

Article history:

Received 16 August 2016

Received in revised form

27 September 2016

Accepted 28 September 2016

Handling Editor: Myrto Petreas

Keywords:

Electronic waste processing

PBDEs

Alternative flame retardants

Surface soil

River sediment

Vietnam

ABSTRACT

We investigated the concentrations of polybrominated diphenyl ethers (PBDEs) and alternative flame retardants (FRs) in environmental samples collected in January 2012, 2013, and 2014 from an electronic waste-processing area in northern Vietnam. During the study period, PBDE and alternative FR concentrations in soils around the electronic waste-processing workshops ranged from 37 to 9200 ng g⁻¹ dry weight (dw) and from 35 to 24,000 ng g⁻¹ dw; the concentrations in soils around the open-burning sites ranged from 1.6 to 62 ng g⁻¹ dw and from <4 to 1900 ng g⁻¹ dw; and the concentrations in river sediments around the workshops ranged from 100 to 3800 ng g⁻¹ dw and from 23 to 6800 ng g⁻¹ dw, respectively. Over the course of study period, we observed significant decreases in concentrations of PBDEs and significant increases in concentrations of alternative FRs, particularly Dechlorane Plus isomers and oligomeric organophosphorus FRs (*o*-PFRs) in both soils and sediments around the workshops. We also report information on concentrations and environmental emissions of *o*-PFRs and their low-molecular-weight impurities in the same soils and sediments. The detection of *o*-PFR impurities around the workshops and the open-burning sites highlights an enhanced breakdown of *o*-PFRs probably due to weathering during open storage and high temperature attained during the burning of electronic wastes.

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1. Introduction

Global production of electronic wastes (e-wastes) in 2014 was estimated at approximately 41.8 million tons, and it is expected to increase to 49.8 million tons by 2018, with an annual growth rate of

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4–5% (Baldé et al., 2015). For both environmental and economic reasons, there is considerable interest in the recovery of recyclable and reusable materials and the disposal of nonrecyclable materials from e-wastes (Widmer et al., 2005). However, concerns were raised about the environmental and human health risks posed by polybrominated diphenyl ether (PBDE) flame retardants (FRs) that are present in polymeric e-wastes (WHO, 1994) because of the persistence, bioaccumulation, and toxicity of these compounds (Ikonomou et al., 2002; Birnbaum and Staskal, 2004). Penta-BDE and Octa-BDE technical formulations were banned worldwide (UNEP, 2009), and Deca-BDE technical formulations were gradually phased out in many countries in the last decade (Dodson et al., 2012).

Since the 2000s, uncontrolled dumping and primitive processing of e-wastes in economically developing countries in Asia have been recognized as causing extreme localized environmental contamination and human exposure levels, as well as more widespread contamination due to subsequent diffusion of PBDEs into indoor and outdoor environments and into the food chain (Robinson, 2009; Sepúlveda et al., 2010). In fact, PBDEs were widespread in surface soils, river sediments, and indoor dusts collected from the informal e-waste-processing areas in northern Vietnam (Tue et al., 2013; Matsukami et al., 2015a). Furthermore, a broad array of alternative FRs – specifically other brominated FRs (BFRs), Dechlorane Plus isomers (DPs), monomeric organophosphorus FRs (*m*-PFRs), and oligomeric organophosphorus FRs (*o*-PFRs) – were found in surface soils, river sediments, and indoor dusts from the same sampling sites in Vietnam (Tue et al., 2013; Matsukami et al., 2015a; Someya et al., 2016); this indicates that these environmental media are sinks for both PBDEs and alternative FRs. The concentrations of PBDEs and alternative FRs in these media seem to depend on the amount of these compounds used, on how they are incorporated into polymeric materials (i.e., as reactants or additives), and on their physicochemical properties. In any case, the detection of these compounds underscores the potential for their emission into the environment because of e-waste processing.

Information about the types and concentrations of FRs in the environment is crucial for the assessment and prediction of the environmental and human health risks posed by FRs derived from e-waste processing. However, there is little definite information on the environmental concentrations and their trends of alternative FRs derived from e-waste processing. With regard to alternative FRs, we investigated the concentrations and potential environmental emissions of *o*-PFRs and low-molecular-weight impurities in technical *o*-PFR formulations (Ballesteros-Gómez et al., 2015; Matsukami et al., 2015b). The estimated vapor pressures and water solubilities suggest that the potential for environmental emission of low-molecular-weight impurities is higher than that of *o*-PFRs. These impurities of *o*-PFRs may also influence the total toxicity of *o*-PFR formulations. Ballesteros-Gómez et al. (2016) reported elevated concentrations of an *o*-PFR impurity in indoor dust samples on and around electronic gadgets and suggested that the impurity may migrate readily into the environment. Little information about the concentrations and environmental emission of *o*-PFR or their impurities because of e-waste processing is available.

The objective of the current study was to investigate the environmental contamination by PBDEs and alternative FRs in an e-waste-processing area in northern Vietnam from 2012 through 2014. Specifically, we used electron ionization (EI)–quadrupole mass spectrometry (QMS) combined with gas chromatography (GC) and electrospray ionization (ESI) tandem mass spectrometry (MSMS) combined with liquid chromatography (LC) to measure the concentrations of 13 PBDE congeners, 4 other BFRs, 2 DP isomers, 8 *m*-PFRs, and 3 *o*-PFRs in surface soil and river sediment samples

collected in January 2013 and January 2014 from sites at which samples had previously been collected in January 2012 as part of our earlier studies (Matsukami et al., 2015a; Someya et al., 2016). Furthermore, we used atmospheric pressure photoionization time-of-flight mass spectrometry (APPI-TOF-MS) combined with LC to measure the concentrations of two impurities found in technical formulations of *o*-PFRs in soils and river sediments. We then compared the peak abundance ratios for *o*-PFRs and their impurities in surface soil and river sediment samples from the e-waste-processing area in 2013 and 2014 with the ratios in the technical formulations to understand the relationship between the concentrations and environmental emissions of *o*-PFRs and their impurities because of e-waste processing.

2. Materials and methods

2.1. Sample collection

The study area was an informal e-waste-processing area in Bui Dau, Hung Yen Province, northern Vietnam (Fig. S1). A map of the surface soil and river sediment sampling sites is shown in Fig. S2. For our earlier studies (Matsukami et al., 2015a; Someya et al., 2016), we collected surface soil samples (depth 0–5 cm) from footpaths in rice paddy sites ($n = 19$), near the open-burning sites ($n = 3$), and near the e-waste-processing workshop sites ($n = 10$) in January 2012. During the same month, we also collected sediment samples from a river from upstream ($n = 1$), near the e-waste-processing workshop sites ($n = 3$), and downstream ($n = 4$). In the present study, we collected surface soil and river sediment samples from the same sampling points in January 2013 and January 2014. Thus, a total of 96 soil samples and 24 sediment samples were collected from January 2012 through January 2014.

Each sample was composed of five subsamples, which were collected with a stainless steel shovel from an area of approximately 10 m² and placed in polyethylene zipper storage bags. The samples were air-dried and manually homogenized with a wooden hammer after the removal of pebbles, weeds, and twigs. The homogenized samples were sieved to <2.0 mm prior to long-term storage at –20 °C and extraction.

2.2. Analytes

Thirteen PBDE congeners (BDE-28, 47, 99, 100, 153, 154, 183, 196, 197, 202, 206, 207, and 209), 4 other BFRs (TBBPA, HBB, BTBPE, and DBDPE; see Appendix A for a list of abbreviations), 2 DP isomers (syn-DP and anti-DP), 8 *m*-PFRs (TCEP, TCIPP, TDCIPP, TPHP, MPDPP, EHDPP, TMPP, and TDMPP), and 3 *o*-PFRs (PBDPP, BPA-BDPP, and PBDMPP) were selected as the target FRs on the basis of the results of our earlier studies (Matsukami et al., 2015a; Someya et al., 2016), our current understanding of potential replacements for PBDEs (Dodson et al., 2012), health concerns (Dodson et al., 2012), and the existence of appropriate analytical methods. In addition, HP-DPHP and BPA-DPHP, which are the impurities present in PBDPP and BPA-BDPP technical formulations (Ballesteros-Gómez et al., 2015, 2016; Matsukami et al., 2015b), were selected as the target *o*-PFR impurities. The CAS numbers, full names, abbreviations, molecular formulas, molecular weights, and physicochemical properties of all the analytes are listed in Table S1. Detailed information about the chemicals and reagents is given in Tables S1 and S2 in the “Chemicals and reagents” section of the supplementary material.

2.3. Analytical methods

The analytical methods used in our earlier studies (Matsukami et al., 2015a; Someya et al., 2016) were followed in this study.

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