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Organophosphate esters in house dust: A comparative study between Canada, Turkey and Egypt.

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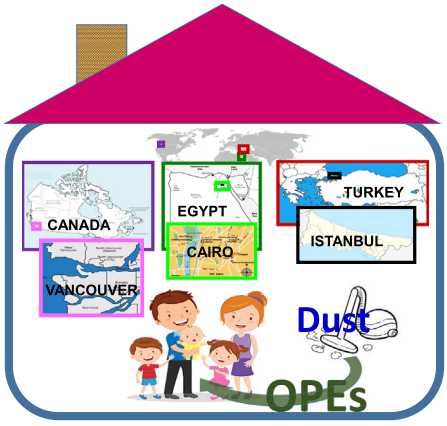
HIGHLIGHTS

- Twelve OPEs were investigated in house dust from Canada, Turkey and Egypt.
- OPEs were ~2 orders of magnitude higher than PBDEs and NFRs in the same dust samples.
- Tris (2-butoxyethyl) phosphate was the dominant OPE in dust in the three countries.
- TBEP exposure via dust was 80% of the RfD for highly exposed children in Vancouver.

GRAPHICAL ABSTRACT



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ABSTRACT

Organophosphate esters (OPEs) are commonly used as flame retardants (FRs) and plasticizers. The usage of OPEs has increased recently due to the ban of several brominated flame retardants, but information on levels in the environment, including the indoor environment is still limited. We investigated the occurrence and distribution of 12 OPEs in urban house dust from Vancouver, Canada; Istanbul, Turkey; and Cairo, Egypt. The median ∑OPE concentration was 41.4 μg/g in the Vancouver samples while median levels in Istanbul and Cairo were significantly lower. The median composition profiles of OPEs in Vancouver and Cairo were dominated by tris (2-butoxyethyl) phosphate (TBOEP), accounting for 56 and 92% of total OPEs respectively while it showed a detection frequency of only 14% in Istanbul. Tris (2-chloropropyl) phosphate (TCPP) was the most abundant chlorinated OPE representing 20 and 36% of the total OPEs in Vancouver and Istanbul respectively, but was below the detection limit in the Cairo dust samples. Consistent with other studies, ΣOPE concentrations were ~1 to 2 orders of magnitude higher than PBDEs and currently used flame retardants in the same dust samples. The mean estimated daily intakes (EDI) of ΣOPE from dust were 115, 38 and 9 ng/kg/bw/day in Vancouver, Cairo and Istanbul respectively for toddlers where adults were ~10 times lower. The total toddler OPE intake

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ranged from 115 to 2900, 38 to 845 and from 9 to 240 ng/kg bw/day across the three cities. TBOEP had the largest contribution to the EDI in both toddler and adults, where toddler TBOEP exposures via dust represented 4% to 80%, 2% to 44% and 0.1% to 6% of the Reference Doses (RfD) in the mean and high intake scenarios for toddlers in Vancouver, Cairo and Istanbul respectively.

1. Introduction

Organophosphate esters (OPEs) are high-production-volume chemicals used as plasticizers and flame retardants in many products such as spray foam insulation, polyurethane foam, furniture, plastics, electronic equipment and textiles (van der Veen and de Boer, 2012). Additionally, they are used as hydraulic fluids and in floor waxes. The recent phase-out of the Penta and Octa polybrominated diphenyl ethers (PBDEs) formulations has led to the increased use of alternative flame retardants (FRs), including OPEs, to meet flammability standards. The annual market demand for FRs has reached 2.2 metric tons in 2018 with the transportation and the construction industries expected to experience the strongest percentage growth for the use of FRs until 2024 (Ceresana Market Research, 2018). In 2013, the market volume of OPEs used as flame retardants reached 562,000 metric tons accounting for 30% of the global total flame retardant demand (China Market Research Reports, 2014–2016). The annual production of two OPEs, tri-phenyl phosphate (TPHP) and tris(1,3-dichloro-2-propyl) phosphate (TDCiPP) in the United States has been estimated to be between 5000 and 23,000 metric tons per year (US EPA, 2006) and the total consumption of FRs in Europe in 2006 was 465,000 metric tons with OPEs constituting 20% of this value (van der Veen and de Boer, 2012).

Similar to PBDEs, OPEs are 'additive', as opposed to 'reactive' FRs, allowing them to migrate out of the material into the environment via volatilization, abrasion, and dissolution (Kemmlein et al., 2003; Marklund et al., 2003) and from direct contact with dust (Rauert et al., 2015). Measurements of OPEs in environmental samples have confirmed their widespread presence throughout the environment at levels that generally exceed those of PBDEs (Cao et al., 2017; Stapleton et al., 2009; Van den Eede et al., 2011). For example, OPEs have been reported in surface fresh water (Regnery and Püttmann, 2010), sediment (Ma et al., 2017; S. Cao et al., 2012; Cristale and Lacorte, 2013), indoor air and dust (Tokumura et al., 2017; Cequier et al., 2014; Hoffman et al., 2015; Tajima et al., 2014), remote arctic air (Sühring et al., 2016; Salamova et al., 2014), fish and biota (Chen et al., 2012), as well as in human milk (Sundkvist et al., 2010; Kim et al., 2014).

Exposure to OPEs may pose a potential threat to human health, even though they are less persistent than PBDEs. OPE metabolites are excreted through urine (Feng et al., 2016; Alves et al., 2014; Van den Eede et al., 2013, 2015). Several OPEs, including TCPP and TDCiPP are thought to be carcinogenic (Freudenthal and Henrich, 2000; van der Veen and de Boer, 2012, Ni et al., 2007). TPHP and TDCiPP were associated with altered hormone levels and decreased semen quality in men (Meeker and Stapleton, 2010) and were inversely correlated with the production of the thyroid hormone free thyroxine (free T4) (Meeker and Stapleton, 2010). Evidence has also appeared regarding the endocrine disruptive effects of TPHP (Kojima et al., 2013; Fang et al., 2003; Liu et al., 2012), whereas tri-n-butyl phosphate (TNBP) was significantly associated with the prevalence of asthma and allergic rhinitis (Araki et al., 2014) and TDCiPP is thought to be a Neurotoxin (Dishaw et al., 2011; Zhang et al., 2014; Wang et al., 2015a, 2015b; Yuan et al., 2016). Chronic exposure to ethylhexyldiphenyl phosphate (EHDPP) has been shown to lead to potential developmental toxicity instigating its listing as a biomonitoring priority chemical in California (Meeting of the Scientific Guidance Panel Potential Priority Chemicals: California, 2013, April 11, 2013). Positive significant correlations have also been found between OPE levels in dust and human hair (Kucharska et al., 2015), suggesting that environmental OPEs contribute to human exposure.

Since OPEs are additive flame retardants which are used in a wide array of household consumer products, their presence in indoor dust is expected. OPEs have been detected in household and office dust at levels up to several mg/kg (Ali et al., 2011; Bergh et al., 2011; Dodson et al., 2012; Tajima et al., 2014; Faiz et al., 2016). Studies point out that indoor dust serves as an important exposure pathway for humans, especially for toddlers who ingest larger quantities of dust compared to adults because of increased hand-to-mouth contact and related behaviors (Larson et al., 2018; Al-Omran and Harrad, 2016; Sugeng et al., 2017).

The objectives of this study were to investigate the occurrence and distribution of OPEs in house dust from 3 different countries with different usage, flammability regulations and typical housing characteristics including the type of construction and insulation. The dust samples analyzed here for OPEs have been previously investigated for PBDEs and currently used flame retardants (Shoeib et al., 2016; Hassan and Shoeib, 2015; Kurt-Karakus et al., 2017; Shoeib et al., 2012) which provides the opportunity to compare the concentrations of PBDEs and currently used flame retardants to OPE levels reported here. This study also aims to compare OPE levels and distributions in dust from the three countries investigated with those reported around the world; and finally, to assess the relative human intake of OPEs and compare these intakes to available reference doses.

2. Experimental

2.1. Chemicals

OPE compounds and isotopically labelled surrogates were purchased from Sigma Aldrich, Accustandard, Wellington Laboratories and Cambridge Isotope Laboratories. Table S1 lists the organophosphate Esters (OPEs), their abbreviations (Bergman et al., 2012) used in the manuscript and corresponding CAS numbers.

2.2. Sample collection

Dust was collected by obtaining whole vacuum cleaner bags (Al-Omran and Harrad, 2017), or by subsampling the contents of canisters from bag-less or central vacuums from houses in three cities from 3 different countries: Vancouver, Canada (n = 92, sampled during 2007–2008), Istanbul, Turkey (n = 39, sampled during February–March 2012) and Cairo, Egypt (n = 17, sampled during December 2013 and January 2014). Blanks were assessed using sodium sulfate that was transported back and forth to the field, exposed to air in the houses. These travel blanks (n = 12) were sieved, stored, and processed/analyzed in the same way as the samples.

2.3. Sample extractions and analysis

Details on sample storage and treatment are provided elsewhere (Shoeib et al., 2016; Hassan and Shoeib, 2015; Kurt-Karakus et al., 2017; Shoeib et al., 2012). Briefly, around 0.1 g of dust sample was homogenized and sieved with mesh size of 150 μm. Sieving removes larger pieces, hair, etc., and makes the dust fraction obtained in the size range that is more likely to be inhaled, stick to skin and/or be