Environment International 133 (2019) 105178

Contents lists available at ScienceDirect

Environment International

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

Organophosphate esters in indoor dust from 12 countries: Concentrations, composition profiles, and human exposure

Wenhui Li[a,b], Yu Wang[a,c], Alexandros G. Asimakopoulos[a,l], Adrian Covaci[d], Bondi Gevao[e], Boris Johnson-Restrepo[f], Taha A. Kumosani[g], Govindan Malarvannan[d], Hyo-Bang Moon[h], Haruhiko Nakata[i], Ravindra K. Sinha[j], Tri Manh Tran[k], Kurunthachalam Kannan[a,g,*]

[a] Wadsworth Center, New York State Department of Health, and Department of Environmental Health Sciences, School of Public Health, State University of New York at Albany, Empire State Plaza, P.O. Box 509, Albany, NY 12201-0509, United States
[b] School of Chemistry and Biological Engineering, University of Science and Technology Beijing, Beijing 100083, China
[c] MOE Key Laboratory of Pollution Processes and Environmental Criteria, College of Environmental Science and Engineering, Nankai University, Tianjin 300350, China
[d] Toxicological Center, University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Antwerp, Belgium
[e] Environmental Management Program, Environment and Life Sciences Center, Kuwait Institute for Scientific Research, P.O. Box 24885, Safat 13109, Kuwait
[f] Environmental and Chemistry Group, Sede San Pablo, University of Cartagena, Cartagena, Bolívar 130015, Colombia
[g] Biochemistry Department, Faculty of Science, Experimental Biochemistry Unit, King Fahd Medical Research Center and Bioactive Natural Products Research Group, King Abdulaziz University, Jeddah, Saudi Arabia
[h] Department of Marine Sciences and Convergent Technology, College of Science and Technology, Hanyang University, Ansan, South Korea
[i] Graduate School of Science and Technology, Kumamoto University, 2-39-1 Kurokami, Kumamoto 860-8555, Japan
[j] Nalanda Open University, Patna 800001, India
[k] Faculty of Chemistry, Hanoi University of Science, Vietnam National University, Hanoi, Le Thanh Tong, Hoan Kiem, Hanoi, Viet Nam
[l] Department of Chemistry, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway

ARTICLE INFO

Handling editor: Heather Stapleton

Keywords:
Organophosphorus flame retardants
Indoor dust
Human exposure
Global trend

ABSTRACT

A total of 20 organophosphate triesters (OPEs), including seven alkyl-OPEs, three chlorinated (Cl)-OPEs, seven aryl-OPEs, and three oligomeric-OPEs were measured in 341 house dust samples collected from 12 countries during the period 2010–2014. OPEs were ubiquitous in indoor dust, and the total concentrations of OPEs (ΣOPEs; sum of 20 OPEs) ranged from 49.4 to 249,000 ng/g dry weight (dw). Generally, Cl-OPEs were the predominant compounds (51% of total) in indoor dust samples, with a median concentration of 800 ng/g, followed by alkyl-OPEs (31%), aryl-OPEs (17%), and oligomeric-OPEs (1%), with median concentrations of 480, 270, and 21.9 ng/g, respectively. ΣOPE concentrations in indoor dust from more industrialized countries (South Korea: median, 31,300; Japan: 29,800; and the United States: 26,500 ng/g dw) were one or two orders of magnitude higher than those from less industrialized countries (Greece: 7140, Saudi Arabia: 5310, Kuwait: 4420, Romania: 4110, Vietnam: 1190, China: 1120, Colombia: 374, India: 276, and Pakistan: 138 ng/g dw). Statistically significant positive correlations ($0.114 < r < 0.748$, $p < 0.05$) were found among the concentrations of 16 OPEs in dust samples, indicating similar sources of these compounds. The median estimated daily intakes of ΣOPEs via dust ingestion for children and adults were in the ranges of 0.29–64.8 and 0.07–14.9 ng/kg bw/day, respectively.

1. Introduction

Organophosphate esters (OPEs) are used extensively worldwide as flame retardants, plasticizers, and anti-foaming agents in various commercial and industrial products, such as electronic equipment, paints, bedding, textiles, furniture, lacquers, floor polishes, and building materials (He et al., 2018). Due to the global restriction and phase-out of polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs), production and consumption of OPEs have sharply increased from 102,000 t in 1992 to 680,000 t in 2015 (Luo et al., 2016; Sun et al., 2018; Zheng et al., 2016). Because most OPEs are physically mixed rather than chemically bound to products, they can emit into the indoor environment from various consumer products via volatilization and abrasion (Cao et al., 2014; Dirtu et al., 2012; Rauert et al., 2014).

Indoor dust is a repository for OPEs released from consumer

* Corresponding author at: Wadsworth Center, Empire State Plaza, P.O. Box 509, Albany, NY 12201-0509, United States.
E-mail address: Kurunthachalam.Kannan@health.ny.gov (K. Kannan).

https://doi.org/10.1016/j.envint.2019.105178
Received 15 July 2019; Received in revised form 6 September 2019; Accepted 9 September 2019
Available online 21 October 2019

products (Kademoglou et al., 2017). Studies have reported on the concentrations of OPEs in indoor dust (Kademoglou et al., 2017; Shoeib et al., 2019; Wong et al., 2017; Yang et al., 2014), which are considerably higher than those found in sediment (Chen et al., 2018; Zhong et al., 2018), sludge (Gao et al., 2016; Wang et al., 2019), soil (Fries and Mihajlovic, 2011; Yadav et al., 2018) and food (Ding et al., 2018; Zhang et al., 2016). Dust ingestion can be an important OPE exposure pathway in humans. Toxicological studies have shown that some OPEs are neurotoxic, carcinogenic, and endocrine disruptors. For example, tris(2-chloroethyl) phosphate (TCEP), and tris(1-chloro-2-propyl) phosphate (TCIPP) elicited moderate acute toxicity in rats (Lu et al., 2017; Zhang et al., 2018a). TCEP was carcinogenic in both rats and mice (European Union, 2009). In vitro, tris (1,3-dichloro-2-propyl) phosphate (TDCIPP) was identified to possess neurotoxic properties on PC12 cells (Ospina et al., 2018). Tris(2-butoxyethyl) phosphate (TBOEP) caused developmental toxicity in zebrafish (Han et al., 2014). Tri-*n*-butyl phosphate (TNBP) and triphenyl phosphate (TPHP) have been reported to be neurotoxic to zebrafish larvae and rats (Behl et al., 2015; Shi et al., 2018). Thus, OPEs in indoor dust may pose a threat to human health, and studies on human exposure to these chemicals are needed (Khan et al., 2016; Zhang et al., 2018b).

Several studies have reported the occurrence of OPEs in geographically remote regions, such as the Arctic and Antarctic (Möller et al., 2012; Salamova et al., 2014). Indoor dust is an important source and pathway for OPEs found in the outdoor environment via dispersion (Marklund et al., 2003). Accordingly, indoor dust can play a role in the transportation of OPEs in the environment. A few multinational studies have reported the occurrence of OPEs in indoor dust. Wong et al. (2017) reported the concentrations of 13 OPEs in 24 dust samples collected from five countries (Australia, the United Kingdom [UK], Canada, Sweden, and China), Shoeib et al. (2019) reported the occurrence of 12 OPEs in 148 house dust collected from three countries (Canada, Egypt, and Turkey), and Kademoglou et al. (2017) reported the concentrations of 16 OPEs in 32 indoor dust samples collected from two European countries (the UK and Norway). In this study, we conducted a multinational survey of 20 OPEs, comprising seven alkyl-OPEs, three Cl-OPEs, seven aryl-OPEs, and three oligomeric-OPEs, in 341 indoor dust samples collected from 12 countries (China, Colombia, Greece, India, Japan, Kuwait, Pakistan, Romania, Saudi Arabia, South Korea, the United States [US], and Vietnam). Human exposure to OPEs via dust ingestion by children and adults was estimated based on the measured concentrations. This is the first study to report a broader geographic occurrence (including several Asian countries) and distribution of OPEs in indoor dust from 12 countries.

2. Materials and methods

2.1. Chemicals and reagents

Trimethyl phosphate (TMP), bisphenol A bis(diphenyl phosphate) (BDP), tetrakis(2-chloroethyl)dichloroisopentyl diphosphate (V6), and resorcinol bis(diphenyl phosphate) (RDP) were purchased from Toronto Research Chemicals (North York, ON, Canada). 2-Ethylhexyl diphenyl phosphate (EHDPP) and tri-iso-butyl phosphate (TIBP) were purchased from Sigma-Aldrich (St. Louis, MO). Triethyl phosphate (TEP), tripropyl phosphate (TPRP), TNBP, TBOEP, tris(2-ethylhexyl) phosphate (TEHP), TCEP, TCIPP, TDCIPP, TPHP, trimethylphenyl phosphate (TMPP), isodecyl diphenyl phosphate (IDDP), cresyl diphenyl phosphate (CDPP), *tert*-butylphenyl diphenyl phosphate (BPDP) and tris(*p-tert*-butylphenyl) phosphate (TBPHP) were purchased from AccuStandard (New Haven, CT). Nine isotopically labeled compounds were used as surrogate standards, which comprise, TEP-d15, purchased from Sigma-Aldrich; TMP-d9 and TEHP-d51, purchased from Toronto Research Chemicals; and TPRP-d21, TNBP-d27, TCEP-d12, TCIPP-d18, TDCIPP-d15, and TPHP-d15, purchased from Cambridge Isotope Laboratories (Tewksbury, MA). Detailed information on target compounds is listed in Table S1 (Supporting Information).

HPLC-grade methanol (99.99%) was purchased from Fisher Scientific (Fair Lawn, NJ). HPLC-grade water, acetonitrile (99.9%), ethyl acetate (99.6%), acetone (99.6%), pesticide grade *n*-hexane (95%), and anhydrous sodium sulfate were purchased from J. T. Baker (Phillipsburg, NJ).

2.2. Sample collection

In total, 341 indoor dust samples were collected from 12 countries, including China ($n = 50$), Colombia ($n = 45$), Greece ($n = 30$), India ($n = 36$), Japan ($n = 14$), Kuwait ($n = 24$), Pakistan ($n = 25$), Romania ($n = 23$), Saudi Arabia ($n = 31$), South Korea ($n = 30$), the US ($n = 15$), and Vietnam ($n = 18$) during the period 2010–2014. The details of the sampling locations are shown in Table S2 (Supporting Information). Sampling was performed by following a common protocol to reduce potential confounding factors. In this study, only house dust samples collected from the bedrooms and living rooms were analyzed. Settled dust samples from the floors in homes were collected by vacuuming for 10 min in the living areas using a household vacuum cleaner or with the use of a brush. After sampling, all dust samples were wrapped in aluminum foil, stored in sealed containers, and stored at −4 °C until they were transported to the laboratory. All samples were ground, sieved, and homogenized by passage through a 150-μm sieve, packed in clean aluminum foil, and stored at 4 °C in the darkroom until analysis.

2.3. Sample pretreatment

Target compounds were extracted from dust samples and purified by solid-phase extraction with Oasis HLB cartridges (6 mL, 200 mg; Waters, Milford, MA), as described in our previous study, with some modifications (Kim et al., 2019). One hundred milligrams of dust sample were weighed into a 15-mL polypropylene (PP) tube. After spiking with 10 ng of internal standards, dust samples were equilibrated for 30 min at room temperature and extracted twice by shaking in an orbital shaker (Eberbach Corp., Ann Arbor, MI) with 5 mL of methanol for 12 h each time. After shaking, samples were centrifuged at 4500 *g* for 10 min (Eppendorf Centrifuge 5804, Hamburg, Germany), and the supernatant was transferred into a 15-mL PP tube. The concentrated extracts were purified by passage through Oasis HLB cartridges (6 mL, 200 mg; Waters) that were preconditioned with 5 mL methanol. Cartridges were then eluted with 2 mL of methanol and 1 mL of ethyl acetate. The extracts were concentrated to near dryness and re-dissolved in 0.25 mL of water/methanol mixture (40,60, v/v), and microcentrifuged (10,000 *g*, 1 min) (0.2 μm nylon filter, Spin-X, Costar, Corning Inc., Corning, NY) before injection into HPLC-MS/MS.

2.4. Instrumental analysis

The concentrations of OPEs were determined by an Agilent 1100 HPLC system (Agilent Technologies, Santa Clara, CA) coupled with an API 2000 triple-quadrupole mass spectrometer (MS/MS), equipped with an electrospray ionization source (ESI) (Applied Biosystems, Foster City, CA). An analytical column (Betasil C_{18}, 2.1 mm × 100 mm i.d., 5.0 μm, Thermo, Waltham, MA), connected to a guard column (Betasil C_{18}, 2.1 mm × 20 mm i.d., 5 μm; Thermo) was used for LC separation. The mobile phase was methanol with 0.1% acetic acid (A) and HPLC-grade water with 0.1% acetic acid (B) maintained at a flow rate of 0.20 mL/min with a gradient program as follows: 0–2 min, 60% A; 2–7.5 min, 60–99% A; 7.5–17 min, 99% A; 17–17.5 min, 99–60% A; 17.5–25 min, 60% A. The positive ion multiple reaction monitoring mode was used for the identification and quantification of OPEs. Detailed information with regard to tandem mass spectrometry parameters is given in Table S3.

2.5. Quality assurance/quality control

All glassware and PP tubes were rinsed with hexane, acetone, methanol, and acetonitrile to avoid potential contamination during analysis. The internal standard method was used in the quantification of each target compound. An 11-point calibration curve was constructed with OPE standard solutions at a concentration range of 0.10–400 μg/L [0.1, 0.5, 1, 2, 5, 10, 25, 50, 100, 200, and 400 μg/L in water–methanol (4,6, v:v)], and the calibration curves exhibited excellent correlation coefficients (r^2 > 0.99). The instrumental limits of detection (LODs) and limits of quantitation (LOQs) were set at a signal-to-noise ratio of 3 and 10, respectively (Table S4). The method detection limits (MDLs) were estimated from LOQs by taking into account a sample amount of 100 mg and a dilution factor of 1:4 (v/v). Procedural blanks were analyzed to check for contamination that may occur during sample preparation and analysis. Trace levels of TNBP, TCIPP, TEHP, TPHP, TMPP, and IDDP (approximately 0.06–0.72 ng) were found in procedural blanks, and background subtraction was performed in the quantification of concentrations in samples. A 10-μg/L OPE standard mixture was injected after every 10 samples to ensure instrumental stability and calibration. Average recoveries of OPEs in spiked blanks and matrices ranged from 74.2 to 116% and from 68.1 to 137%, respectively. Detailed information about recoveries, correlation coefficients, and LODs of target compounds is listed in Tables S4 and S5 in the Supporting Information.

2.6. Statistical analysis

Statistical analyses were performed with SPSS/PASW Statistics 18.0 (SPSS Inc., 1993–2007). Concentrations of target compounds below the MDLs were assigned a value of 1/2 MDLs for data analysis. The total concentrations of seven alkyl-OPEs, three Cl-OPEs, seven aryl-OPEs, three oligomeric-OPEs, and the sum of 20 OPEs were denoted as ΣAlkyl-OPEs, ΣCl-OPEs, ΣAryl-OPEs, ΣOligo-OPEs, and ΣOPEs, respectively. A nonparametric Spearman rank correlation test (two-tailed, significance level at 0.05) was performed to assess the relationships among the concentrations of individual OPEs in indoor dust samples.

3. Results and discussion

3.1. Concentrations

The concentrations and detection frequencies of OPEs in indoor dust from 12 countries are presented in Tables 1 and 2. A total of 20 OPEs were detected in 341 indoor dust samples. Except for TMP, TPRP, TBPHP, RDP, and V6, other OPEs were found at detection frequencies higher than 73%, indicating their widespread usage in the indoor environment of several countries. The concentrations of ΣOPEs, ΣAlkyl-OPEs, ΣCl-OPEs, ΣAryl-OPE, and ΣOligo-OPE in indoor dust samples from 12 countries are shown in Table 2. The total concentration of OPEs (ΣOPEs) in indoor dust ranged from 49.4 to 249,000 ng/g dry weight (dw). Generally, Cl-OPEs were the predominant compounds (accounting for 51% of ΣOPEs) in indoor dust samples, with a median concentration of 800 ng/g, followed by alkyl-OPEs (31%), aryl-OPEs (17%), and oligo-OPEs (1%), with median concentrations of 480, 270, and 21.9 ng/g, respectively.

3.1.1. Cl-OPEs

Due to their high production, extensive usage and persistence in the environment (Guo et al., 2017; Kim et al., 2014; Reemtsma et al., 2008), Cl-OPEs were the dominant OPEs found in indoor dust. Among three Cl-OPEs measured, TCIPP was the most abundant compound, found at a median concentration of 222 ng/g (< MDL-66,700 ng/g), followed by TCEP and TDCIPP at 201 ng/g (< MDL-45,900 ng/g) and 79.6 ng/g (< MDL-16,6000 ng/g), respectively. The elevated concentration of TCIPP found in indoor dust was consistent with its high production volume. TCIPP accounted for about 80% of Cl-OPE usage in Europe and the US (Castro-Jimenez et al., 2014). It should be noted that TCIPP was found in only 76% of the samples, while TCEP, which has gradually been replaced by TCIPP in some regions (due to the carcinogenic and mutagenic properties of TCEP) (Green et al., 2008), was found in 99% of the samples. The relatively high concentrations and detection frequencies of TCEP suggest continuing usage of this chemical in Asia and North America (Ma et al., 2017). In addition, TCEP was found at concentrations of 14% (by weight) in V6 commercial mixtures (Fang et al., 2013). Thus, TCEP found in indoor dust also may originate from the use of V6 in consumer products, although V6 was found only in 51% of the samples. In comparison to the other two Cl-OPEs, TDCIPP was found at relatively low concentrations (median: 79.6 ng/g). This may be ascribed to its high cost, which is about twice that of TCIPP. Another reason is that TDCIPP is used mainly in the automotive industry (outdoor), while only a small fraction is used in furniture (indoor) (Danish Environmental Protection Agency, 2016).

Table 1
Overall concentrations of organophosphate esters in 341 indoor dust samples collected from 12 countries (ng/g dw).

Compounds	MDL[a]	Min	Median	Mean	95th	Max	df (%)[b]
TMP	0.32	< MDL	< MDL	1.40	1.16	332	11.4
TEP	1.2	< MDL	24.5	116	397	7340	82.7
TPRP	2.1	< MDL	< MDL	3.37	3.38	592	10.0
TNBP	0.95	< MDL	32.5	86.2	373	1690	98.2
TIBP	0.56	< MDL	9.64	37.1	117	1450	98.2
TBOEP	1.87	< MDL	190	7740	50,900	234,000	99.7
TEHP	2.02	< MDL	37.1	84.3	316	2930	93.8
TCEP	0.79	< MDL	201	1260	6880	45,900	99.1
TCIPP	2.33	< MDL	222	1310	5550	66,700	76.2
TDCIPP	0.90	< MDL	79.6	1440	4220	166,000	81.2
TPHP	0.40	< MDL	112	726	2680	25,200	97.4
TMPP	0.35	< MDL	16.5	125	323	7330	98.5
EHDPP	0.29	< MDL	27.3	96.5	396	1920	99.4
CDPP	1.0	< MDL	15.7	316	432	38,900	94. 4
IDDP	0.31	< MDL	8.21	171	260	13,600	97.7
BPDP	0.28	< MDL	3.88	17.3	62.5	716	90.9
TBPHP	1.56	< MDL	< MDL	22.3	23.9	5160	22.6
RDP	0.74	< MDL	< MDL	53.7	172	5480	46.9
BDP	0.52	< MDL	3.45	37.7	189	1390	73.3
V6	0.64	< MDL	0.75	124	210	23,500	51.3
ΣAlkyl-OPEs	–	17.0	480	8070	51,300	235,000	100
ΣCl-OPEs	–	2.01	800	4010	16,000	176,000	96.2
ΣAryl-OPEs	–	3.63	270	1470	4400	64,900	99.1
ΣOligo-OPEs	–	< MDL	21.9	215	703	23,600	79.5
ΣOPEs	–	49.4	2220	13,800	79,100	249,000	100

[a] Method detection limits.
[b] Detection frequency (%).

3.1.2. Alkyl-OPEs

All seven alkyl-OPEs were detected in house dust samples (Table 1). TBOEP was the dominant alkyl-OPE, found at a median concentration of 190 ng/g (< MDL-23,4000 ng/g, DF = 99.7%), accounting for 64.4% of the total alkyl-OPE concentrations. This may be related to its widespread use in the indoor environment, including floor polishes, synthetic rubber, plastics, and lacquers (Volkel et al., 2018) (Table S6). TEHP, TNBP, TEP, and TIBP also were frequently detected (DF > 93%) at median concentrations of 37.1, 32.5, 24.5, and 9.64 ng/g, respectively. In contrast, TMP and TPRP were found at low levels with detection frequencies below 12%. TMP is released primarily from industrial processes and seldom found in the indoor environment (Marklund, 2005). TPRP is commonly used as a flame retardant in combination with PBDEs and other OPEs (Van der Veen and de Boer, 2012).

3.1.3. Aryl-OPEs

All seven aryl-OPEs were detected in house dust samples. TPHP was

Table 2
Concentrations of organophosphate esters (OPEs) in indoor dust collected from 12 countries (ng/g dw).

Groups	Compounds		China (50[a])	Colombia (45)	Greece (30)	India (36)	Japan (14)	Korea (30)	Kuwait (24)	Pakistan (25)	Romania (23)	Saudi Arabia (31)	USA (15)	Vietnam (18)
Alkyl-OPEs	TMP	Median/df[b]	< MDL/16	< MDL/4	< MDL/0	< MDL/25	< MDL/14	< MDL/17	< MDL/8	< MDL/16	< MDL/0	< MDL/16	< MDL/13	< MDL/11
		range	< MDL-4.12	< MDL-1.34	< MDL	< MDL-0.57	< MDL-1.93	< MDL-2.67	< MDL-36.7	< MDL-2.76	< MDL	< MDL-7.77	< MDL-5.52	< MDL-332
	TEP	Median/df	13.7/92	17.0/78	39.6/93	5.69/53	34.9/93	141/97	18.2/88	< MDL/20	44.1/100	26.2/100	74.1/100	37.0/100
		Range	< MDL-236	< MDL-444	< MDL-2520	< MDL-3280	< MDL-902	< MDL-659	< MDL-695	< MDL-24.5	3.08–575	5.59–406	8.14–482	1.89–7340
	TPRP	Median/df	< MDL/10	< MDL/27	< MDL/7	< MDL/3	< MDL/21	< MDL/10	< MDL/21	< MDL/0	< MDL/0	< MDL/16	< MDL/13	< MDL/0
		Range	< MDL-24.5	< MDL-9.55	< MDL-27.7	< MDL-2.76	< MDL-592	< MDL-9.01	< MDL-26.9	< MDL	< MDL	< MDL-3.41	< MDL-21.0	< MDL
	TNBP	Median/df	32.2/100	4.82/100	66.6/100	7.39/100	78.8/100	130/90	33.6/92	6.56/100	84.0/100	42.3/97	89.2/100	16.6/100
		Range	1.71–306	1.35–57.8	19.7–448	1.23–144	13.0–507	< MDL-1690	< MDL-1050	2.16–28.7	44.4–963	< MDL-156	32.5–1320	2.60–12.6
	TIBP	Median/df	9.66/100	1.67/100	41.1/100	2.29/100	10.5/100	28.0/90	20.1/92	5.62/100	47.1/100	14.7/100	13.6/100	3.83/98
		Range	1.21–551	0.33–8.01	6.17–1060	0.42–32.5	1.62–45.4	< MDL-756	< MDL-1450	0.29–43.5	21.3–1130	2.61–49.1	1.23–238	< MDL-16.8
	TBOEP	Median/df	45.0/98	83.1/100	2870/100	27.9/100	19,400/100	23,300/100	196/100	17.9/100	712/100	629/100	12,200/100	103/100
		Range	< MDL-1200	8.58–1050	315/86300	4.91–1930	3240–91,000	982–234,000	7.44–7430	4.41–139	99.3–19,200	21.6–4400	575–51,100	52.3–197
	TEHP	Median/df	35.5/98	12.6/96	68.6/100	26.4/97	83.8/86	90.9/100	29.8/92	7.40/76	54.8/100	64.5/97	97.9/100	10.0/89
		Range	< MDL-922	< MDL-97.0	12.8–939	< MDL-380	< MDL-424	17.9–2930	< MDL-1320	< MDL-29.8	12.8–322	< MDL-173	29.0–585	< MDL-797
Cl-OPEs	TCEP	Median/df	298/100	16.9/100	335/100	15.7/100	960/93	2490/100	450/96	5.05/96	105/100	721/100	330/100	43.4/100
		Range	39.7–45,900	1.30–229	98.4–1520	2.40–647	< MDL-28000	192–15,400	< MDL-18500	< MDL-139	25.9–1370	153–6500	164–7530	6.23–2460
	TCIPP	Median/df	29.8/72	41.1/62	590/100	< MDL/42	1630/100	722/100	536/100	< MDL/0	745/100	1000/100	3070/100	40.5/89
		Range	< MDL-1610	< MDL-835	112–6480	< MDL-1100	47.1–34,300	19.7–8290	94.3–29,300	< MDL	7.18–6140	212–11,100	278–66,700	< MDL-2310
	TDCIPP	Median/df	37.6/72	10.6/71	667/100	26.7/81	1500/100	78.9/73	754/100	< MDL/40	81.0/96	899/100	1750/100	15.1/72
		Range	< MDL-4680	< MDL-691	188–13,100	< MDL-361	103–166,000	< MDL-20500	29.5–33,600	< MDL-33.0	< MDL-605	133–29,200	285–17,600	< MDL-3260
Aryl-OPEs	TPHP	Median/df	109/100	29.1/91	544/100	19.4/97	199/100	524/100	60.5/88	16.8/96	203/100	107/100	553/100	39.2/100
		Range	3.52–7620	< MDL-833	63.0–4660	< MDL-2240	50.8–833	66.8–25,200	< MDL-1770	< MDL-125	37.4–19,500	14.7–548	56.8–5780	9.28–21,800
	TMPP	Median/df	13.9/100	3.22/98	23.4/100	6.42/97	132/100	39.2/100	18.8/100	3.26/92	128/96	30.4/100	17.3/100	7.87/100
		Range	1.75–116	< MDL-215	6.34–145	< MDL-1150	30.5–1970	3.69–168	2.63–3970	< MDL-24.5	< MDL-7330	5.69–128	0.70–925	0.75–6880
	EHDPP	Median/df	14.7/100	12.8/100	90.0/100	6.12/100	77.4/100	132/100	23.8/100	2.04/92	46.7/100	113/100	70.6/100	16.6/100
		Range	1.26–361	0.83–832	13.4–1050	0.86–83.2	15.8–192	15.4–1490	3.58–400	< MDL-74.3	15.8–594	11.6–1200	5.53–530	0.54–294
	CDPP	Median/df	10.1/100	4.32/98	59.9/97	5.47/97	45.1/93	41.8/93	21.9/88	3.92/92	100/100	28.9/100	34.9/100	6.42/100
		Range	0.20–151	< MDL-202	< MDL-873	< MDL-866	< MDL-421	< MDL-3870	< MDL-433	< MDL-59.1	5.86–25,700	2.75–137	2.88–6700	1.01–38,900
	IDDP	Median/df	3.81/98	3.72/100	10.9/100	4.13/97	39.0/100	170/100	8.00/83	3.86/92	16.8/100	12.9/100	46.9/100	3.65/100
		Range	< MDL-12400	0.69–38.8	2.49–92.9	< MDL-262	11.9–156	6.69–13,600	< MDL-1810	< MDL-113	1.77–116	2.19–334	7.46–500	0.43–70.2
	BPDP	Median/df	2.12/88	2.73/98	10.1/100	0.98/78	4.67/100	6.70/100	3.38/79	1.70/84	13.6/100	13.6/97	37.3/100	0.82/89
		Range	< MDL-21.8	< MDL-79.6	2.22–165	< MDL-608	0.50–24.2	1.23–117	< MDL-54.7	< MDL-9.40	2.77–716	< MDL-78.7	0.87–565	< MDL-52.9
	TBPHP	Median/df	< MDL/10	< MDL/7	< MDL/40	< MDL/22	< MDL/14	< MDL/50	< MDL/25	< MDL/20	< MDL/26	< MDL/29	9.39/67	< MDL/28
		Range	< MDL-58.1	< MDL-5160	< MDL-24.2	< MDL-12.8	< MDL-8.37	< MDL-9.04	< MDL-10.3	< MDL-4.98	< MDL-5.73	< MDL-363	< MDL-358	< MDL-603
Oligo-OPEs	RDP	Median/df	< MDL/40	< MDL/31	< MDL/37	< MDL/44	< MDL/36	12.5/57	< MDL/54	2.73/76	< MDL/35	3.07/65	< MDL/40	22.0/89
		Range	< MDL-227	< MDL-59.1	< MDL-679	< MDL-912	< MDL-80.3	< MDL-326	2.18–667	< MDL-338	< MDL-67.7	< MDL-122	< MDL-95.6	< MDL-5480
	BDP	Median/df	2.00/82	< MDL/38	12.8/83	3.30/86	5.22/79	8.08/67	6.13/92	0.72/52	< MDL/43	13.0/94	4.95/87	52.6/100
		Range	< MDL-192	< MDL-101	< MDL-547	< MDL-330	< MDL-60.6	< MDL-1360	< MDL-830	< MDL-9.32	< MDL-26.2	< MDL-668	< MDL-163	6.80–1390
	V6	Median/df	< MDL/48	< MDL/11	9.80/83	< MDL/3	25.3/79	18.4/97	3.45/71	< MDL/16	< MDL/57	< MDL/97	65.7/100	< MDL/39
		Range	< MDL-114	< MDL-63	< MDL-260	< MDL-0.98	< MDL-23500	< MDL-2260	< MDL-4640	< MDL-2.50	< MDL-292	11.6–319	1.71–1900	< MDL-96.6
	ΣAlkyl-OPEs	Median/df	137/100	120/100	3090/100	70.9/100	19,600/100	23,700/100	299/100	39.3/100	943/100	778/100	12,500/100	171/100
		Range	6.70–3240	13.1–1670	356–91,300	9.39–5770	3260–93,500	1000–240,000	11.0–12,000	9.70–269	182–22,200	32.5–5200	647–53,800	59.3–8810
	ΣCl-OPEs	Median/df	580/100	94.4/100	1780/100	56.4/100	6700/100	6030/100	1990/100	7.91/96	1060/100	3420/100	8080/100	181/100
		Range	57.1–46,500	2.92–1470	544–21,100	6.64–2100	544–176,000	500–26,200	482–40,000	< MDL-153	88.8–6240	583–33,900	820–85,000	11.7–8030
	ΣAryl-OPEs	Median/df	161/100	63.3/100	1010/100	53.3/100	621/100	1060/100	213/100	33.0/100	582/100	384/100	1030/100	81.3/100
		Range	12.5–20,400	3.63–6580	271–4920	4.52–5200	296–2760	130–39,300	56.1–4330	6.11–346	75.6–53,400	64.3–1720	134–12,900	16.1–64,900
	ΣOligo-OPEs	Median/df	11.5/96	1.20/60	40.5/97	4.53/89	33.3/100	78.7/100	42.6/96	6.47/92	11.3/78	87.5/100	92.7/100	80.2/100
		Range	< MDL-265	< MDL-126	< MDL-1050	< MDL-1240	11.2–23,600	4.47–2280	< MDL-4680	< MDL-345	< MDL-360	2.10–794	7.95–1950	13.3–6020
	ΣOPEs	Median/df	1120/100	374/100	7140/100	276/100	29,800/100	31,300/100	4420/100	138/100	4110/100	5310/100	26,500/100	1190/100
		Range	149–47,400	54.6–8130	1690–90,200	52.5–9650	7720–238,000	3090–249,000	633–44,400	49.4–473	775–54,900	791–35,000	1930–101,000	228–79,600

[a] Sample number.
[b] Detection frequency (%).

the dominant aryl-OPE, found at a median concentration of 112 ng/g (< MDL-25,200 ng/g), accounting for 61% of the total aryl-OPE concentrations, which was followed by EHDPP, TMPP, CDPP, IDDP, and BPDP, found at median concentrations of 27.3, 16.5, 15.7, 8.21, and 3.88 ng/g, respectively. It was reported that TPHP was the most widely produced and used aryl-OPE in the US and European countries (Norway, Sweden, Denmark, and Finland) (Van der Veen and de Boer, 2012) and is used as a flame retardant in cellulose acetate, vinyl copolymers in vacuum cleaner parts, computer monitors, and television sets (Green et al., 2008; Saito et al., 2007). Besides, TPHP is an impurity in the commercial mixture of other aryl-OPEs, which can contribute to the increase in TPHP concentrations in indoor dust (Behl et al., 2016; Kademoglou et al., 2017). It should be noted that three novel aryl-OPEs, IDDP, CDPP, and BPDP, were frequently found in house dust samples (> 90%), indicating their widespread global usage. TBPHP, a minor component of commercial BPDP mixture (US EPA, 2008), was detected in only 23% of the house dust samples at a median concentration below the MDL.

3.1.4. Oligo-OPEs

In comparison to monomeric OPEs, oligo-OPEs have lower volatility and higher environmental stability (Van der Veen and de Boer, 2012). There is a general trend that oligo-OPEs may replace monomeric OPEs as flame retardants in the future (European Commission, 2011). For instance, V6 is an alternative to TCIPP and TDCIPP, while RDP and BDP are often used as substitutes of TPHP (Kademoglou et al., 2017). Generally, due to their limited current market share and low mobility (Fang et al., 2013; Liang et al., 2018), oligo-OPEs are rarely detected in the environment. BDP, which is used in furniture, housing for electric and electronic equipment, textiles, and flooring, was found in 73% of the indoor dust samples at a median concentration of 3.45 ng/g (< MDL-1390 ng/g). The other two oligo-OPEs, V6 (median: 0.75 ng/g) and RDP (median < MDL), were found in only 51% and 47% of the indoor dust samples, respectively. Limited studies, however, have reported high concentrations of oligo-OPEs in house dust from China (Tan et al., 2018), Norway, and the UK (Kademoglou et al., 2017). In addition, an increase in oligo-OPE concentration in river sediments has been reported during the period 2012–2014 in Vietnam (Matsukami et al., 2017).

3.2. Spatial distribution

Statistically significant differences ($p < 0.05$) in ΣOPE concentrations were found in indoor dust samples collected among the 12 countries from 2010 to 2014. The median concentrations of OPEs were the highest in dust samples from South Korea (31,300 ng/g), Japan (29,800 ng/g), and the US (26,500 ng/g). These values were one or two orders of magnitude higher than those in Greece (7140 ng/g), Saudi Arabia (5310 ng/g), Kuwait (4420 ng/g), Romania (4110 ng/g), Vietnam (1190 ng/g), China (1120 ng/g), Colombia (374 ng/g), India (276 ng/g) and Pakistan (138 ng/g). A strong and statistically significant correlation ($r = 0.862$, $p < 0.01$) was observed between median ΣOPE concentrations in house dust and per capita GDP of the 12 countries studied (Fig. S1). Earlier studies have reported higher concentrations of ΣOPEs in indoor dust from more industrialized countries (the US, Canada, Japan, Belgium, Spain, Sweden, Germany, and the Netherlands) than those from less industrialized nations (China, Egypt, Pakistan, Philippines, Saudi Arabia, and Kuwait) (Ali et al., 2017; Kim et al., 2013; Zhou et al., 2017). In general, the concentrations of OPEs in house dust reflect past and current consumption in different countries. The elevated OPE concentrations found in indoor dust from industrialized countries can be attributed to increasing use of these chemicals driven by the ban on PBDEs and requirement of higher flammability standards (He et al., 2018). It has been reported that approximately 60–70% of the global production and consumption of OPEs was in the US and Europe from 1995 to 2008 (Kim et al., 2017). In contrast, due to less stringent fire safety regulations in developing countries, the use of OPEs in these countries is currently low (Zhou et al., 2017).

3.3. Correlations and profiles

The Spearman correlations among individual OPEs with detection frequencies over 50% in indoor dust are shown in Table S7. All 16 OPEs showed statistically significantly positive correlations ($0.114 < r < 0.748$, $p < 0.05$) with one another in 341 indoor dust samples, indicating that these compounds may have similar sources of origin and have mixed application patterns (Cao et al., 2014). Most of the OPEs are used not only as flame retardants but also as plasticizers in consumer products, resulting in their concurrent occurrence in the indoor environment (Table S6). The strongest correlations were observed between TNBP and TIBP ($r = 0.748$, $p < 0.01$), probably due to their mixed application in household products. In addition, some commercial mixtures of OPEs contain impurities or byproducts (De Boer et al., 2016; Matsukami et al., 2015b). For instance, TCEP was strongly correlated with V6 ($r = 0.652$, $p < 0.01$), which can be attributed to its occurrence as an impurity in commercial V6 mixture (Fang et al., 2013). TPHP showed a moderate to strong correlation with BDP, IDDP, and BPDP ($0.259 < r < 0.602$, $p < 0.01$) because the former is a known impurity in the technical BDP, IDDP, and BPDP mixtures (Behl et al., 2016; Brandsma et al., 2013; Kademoglou et al., 2017).

The contribution of each of the target compounds (based on median concentrations) to ΣOPE concentrations in dust is presented in Fig. 1. ΣCl-OPEs accounted for 78% of ΣOPE concentrations in dust from Kuwait, followed by Saudi Arabia (73%) > China (65%) > Romania (41%) > the US (37%) > Vietnam (35%) > Colombia (34%) > India (31%) > Greece (30%) > Japan (25%) > South Korea (20%) > Pakistan (9%). ΣAlkyl-OPEs accounted for 77% of ΣOPE concentrations in dust from South Korea, followed by Japan (73%) > the US (58%) > Greece (52%) > Pakistan (45%) > Colombia (43%) > India (38%) > Romania (36%) > Vietnam (33%) > Saudi Arabia (17%) > China (15%) > Kuwait (12%). ΣAryl-OPEs accounted for 38% of ΣOPE concentrations in dust from Pakistan, followed by India (29%) > Colombia (23%) > Romania (22%) > China (18%) > Greece (17%) > Vietnam (16%) > Kuwait (9%) > Saudi Arabia (8%) > the US (5%) > South Korea (3%) > Japan (2%). ΣOligo-OPEs accounted for < 7% of ΣOPE concentrations in all countries except Vietnam (16%), which were consistent with relatively high concentrations of BDP and RDP reported in soil and sediment from Vietnam (Matsukami et al., 2015a; Matsukami et al., 2017).

Cl-OPEs are suspect carcinogens (Van der Veen and de Boer, 2012). TCEP has been listed as a “known carcinogen” by the Californian Environmental Protection Agency since 1992 (Stubbings et al., 2016) and classified as a Category 2 carcinogen by the European Union (European Union, 2009). Higher concentrations of TCEP found in Korea (median:2490 ng/g), Japan (960 ng/g) and Saudi Arabia (721 ng/g), require further attention with regard to potential impacts on human exposure and health. The profiles of three Cl-OPEs found in indoor dust exhibited a geographical difference among the 12 countries (Fig. 2). High contributions of TCEP to ΣCl-OPEs were found in dust from most East and South Asian countries, such as China (82%), Pakistan (76%), South Korea (76%), Vietnam (44%), and India (36%); followed by two Middle Eastern countries, Saudi Arabia (28%) and Kuwait (26%); one South American country, Colombia (25%); and Japan (23%), whereas this ratio was low in North America (the US: 6%), and Europe (Romania: 11%, and Greece: 21%). The collective contributions of TCIPP and TDCIPP, two alternatives of TCEP, presented an opposite trend. The composition profiles of OPEs in house dust reflect their consumption patterns globally. The low proportion of TCEP found in dust from the European Union and the US suggests a decline in the consumption of this compound in recent years. TCEP is no longer produced in the

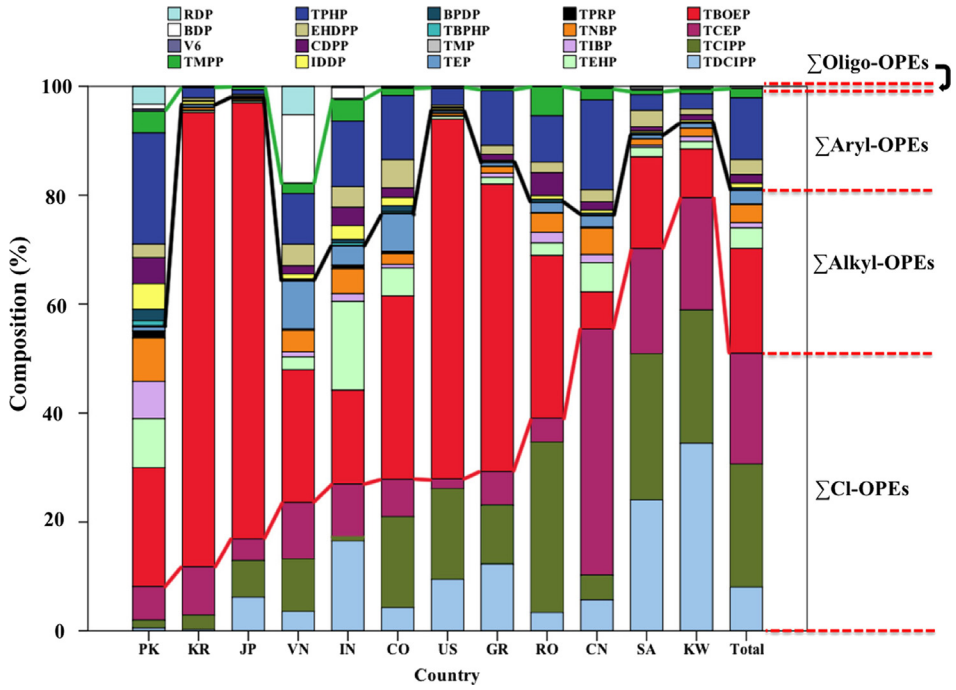

Fig. 1. Composition profiles of organophosphate esters (OPEs) in house dust from 12 countries. Median concentrations were used to calculate the composition profiles.

European Union, and consumption of this compound decreased from 10,500 t in 1992 to 1000 t in 2009 (European Union, 2009). Similarly, production of TCEP in the US dropped sharply from 227–454 t in 2006 to < 11.3 t in 2014 (Van der Veen and de Boer, 2012; US EPA, 2016). The high proportion of TCEP in Asian countries, however, indicates that this compound is still being used, where no regulations currently exist (Zhou et al., 2017). The high proportion of TCEP in house dust from Asian countries warrants further attention.

3.4. Exposure assessment

Previous studies have shown that dust ingestion was the dominant pathway of human exposure to OPEs, while dermal absorption and inhalation were minor contributors to the total daily exposures (Wei et al., 2015). In this study, we calculated the estimated daily intake (EDI) of OPEs via indoor dust ingestion for children and adults (Liao et al., 2012; Wang et al., 2015). The EDI was calculated as shown in Eq. (1).

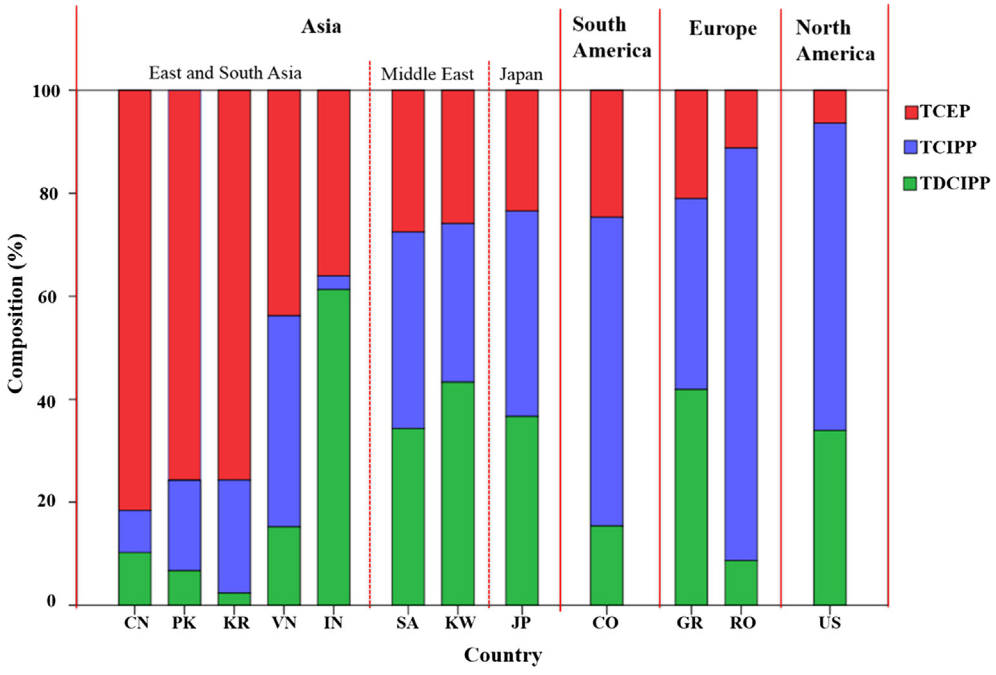

Fig. 2. Composition profiles of TCEP, TCIPP and TDCIPP in house dust from 12 countries. Median concentrations were used to calculate the composition profiles.

Table 3
Estimated daily intakes (EDIs, ng/kg-bw/day) of OPEs (for this with DF > 50%) via indoor dust ingestion for children and adults from 12 countries.

Country	Exposure scenario	TEP	TNBP	TIBP	TBOEP	TEHP	TCEP	TCIPP	TDCIPP	TPHP	TMPP	EHDPP	CDPP	IDDP	BPDP	BDP	V6	ΣOPEs
RfD[a]		13,000	10,000	–	15,000	100,000	7000	10,000	20,000	70,000	13,000	–	–	–	–	–	–	–
Pakistan																		
Children	Average[b]	0.00	0.01	0.01	0.04	0.02	0.01	0.00	0.00	0.03	0.01	0.00	0.01	0.01	0.00	0.00	0.00	0.29
	High[c]	0.03	0.03	0.04	0.17	0.05	0.13	0.00	0.06	0.19	0.04	0.04	0.05	0.08	0.02	0.02	0.00	0.92
Adult	Average	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.07
	High	0.01	0.01	0.01	0.04	0.01	0.03	0.00	0.01	0.04	0.01	0.01	0.01	0.02	0.04	0.00	0.00	0.21
India																		
Children	Average	0.01	0.02	0.00	0.06	0.05	0.03	0.00	0.06	0.04	0.01	0.01	0.01	0.01	0.00	0.01	0.00	0.57
	High	0.93	0.07	0.02	0.54	0.34	0.20	0.75	0.33	1.84	0.16	0.11	0.07	0.12	0.05	0.05	0.00	6.64
Adult	Average	0.00	0.00	0.00	0.01	0.01	0.01	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.13
	High	0.21	0.02	0.01	0.12	0.08	0.05	0.17	0.08	0.42	0.04	0.03	0.01	0.03	0.12	0.01	0.00	1.53
Colombia																		
Children	Average	0.03	0.01	0.00	0.16	0.02	0.03	0.08	0.02	0.05	0.01	0.02	0.01	0.01	0.01	0.00	0.00	0.70
	High	0.52	0.07	0.01	1.31	0.15	0.35	1.29	0.28	0.50	0.08	0.18	0.05	0.04	0.07	0.03	0.00	4.13
Adult	Average	0.01	0.00	0.00	0.03	0.00	0.01	0.02	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.14
	High	0.10	0.01	0.00	0.26	0.03	0.07	0.26	0.06	0.10	0.02	0.04	0.01	0.01	0.01	0.01	0.00	0.83
China																		
Children	Average	0.03	0.07	0.02	0.09	0.07	0.62	0.06	0.08	0.23	0.03	0.03	0.02	0.01	0.00	0.00	0.00	2.32
	High	0.19	0.42	0.46	0.82	0.44	15.0	0.91	1.92	2.28	0.09	0.11	0.10	0.03	0.02	0.14	0.04	24.6
Adult	Average	0.01	0.02	0.00	0.02	0.02	0.14	0.01	0.02	0.05	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.53
	High	0.04	0.10	0.10	0.19	0.10	3.44	0.21	0.44	0.52	0.02	0.03	0.02	0.01	0.02	0.03	0.01	5.67
Vietnam																		
Children	Average	0.08	0.03	0.01	0.21	0.02	0.09	0.08	0.03	0.08	0.02	0.03	0.01	0.01	0.00	0.11	0.00	2.46
	High	4.45	0.10	0.03	0.35	0.40	1.50	2.86	1.81	11.5	8.34	0.18	14.6	0.08	0.07	1.20	0.04	52.8
Adult	Average	0.02	0.01	0.00	0.05	0.00	0.02	0.02	0.01	0.02	0.00	0.01	0.00	0.00	0.00	0.03	0.00	0.57
	High	1.02	0.02	0.01	0.08	0.09	0.35	0.66	0.42	2.65	1.92	0.04	3.36	0.02	1.07	0.28	0.01	12.1
Romania																		
Children	Average	0.08	0.16	0.09	1.34	0.10	0.20	1.40	0.15	0.38	0.24	0.09	0.19	0.03	0.03	0.00	0.00	7.71
	High	0.34	0.67	0.34	26.4	0.32	1.49	10.3	0.43	33.6	2.44	0.77	14.3	0.18	0.24	0.04	0.08	60.0
Adult	Average	0.02	0.03	0.02	0.27	0.02	0.04	0.28	0.03	0.08	0.05	0.02	0.04	0.01	0.01	0.00	0.00	1.54
	High	0.07	0.13	0.07	5.29	0.06	0.30	2.05	0.09	6.71	0.49	0.15	2.87	0.04	0.01	0.01	0.02	12.0
Kuwait																		
Children	Average	0.04	0.07	0.04	0.41	0.06	0.93	1.11	1.56	0.13	0.04	0.05	0.05	0.02	0.01	0.01	0.01	9.14
	High	0.55	1.96	0.28	10.1	0.69	31.9	39.5	7.22	1.41	1.65	0.48	0.74	0.25	0.07	1.35	0.43	86.5
Adult	Average	0.01	0.02	0.01	0.09	0.01	0.21	0.26	0.36	0.03	0.01	0.01	0.01	0.00	0.00	0.00	0.00	2.10
	High	0.13	0.45	0.07	2.33	0.16	7.33	9.10	1.66	0.32	0.38	0.11	0.17	0.06	0.08	0.31	0.10	19.9
Saudi Arabia																		
Children	Average	0.05	0.09	0.03	1.30	0.13	1.49	2.07	1.86	0.22	0.06	0.23	0.06	0.03	0.03	0.03	0.02	11.0
	High	0.25	0.31	0.05	5.98	0.24	10.4	6.48	17.8	0.77	0.24	1.44	0.23	0.41	0.11	0.91	0.25	31.2
Adult	Average	0.01	0.02	0.01	0.30	0.03	0.34	0.48	0.43	0.05	0.01	0.05	0.01	0.01	0.01	0.01	0.01	2.53
	High	0.06	0.07	0.01	1.38	0.06	2.39	1.49	4.09	0.18	0.06	0.33	0.05	0.10	0.03	0.21	0.06	7.19
Greece																		
Children	Average	0.07	0.12	0.08	5.38	0.13	0.63	1.11	1.25	1.02	0.04	0.17	0.11	0.02	0.02	0.02	0.02	13.4
	High	0.39	0.52	0.43	83.8	0.45	2.04	4.05	7.82	5.53	0.22	1.22	0.40	0.15	0.11	0.57	0.30	103
Adult	Average	0.01	0.02	0.02	1.08	0.03	0.13	0.22	0.25	0.20	0.01	0.03	0.02	0.00	0.00	0.00	0.00	2.68
	High	0.08	0.10	0.09	16.8	0.09	0.41	0.81	1.56	1.11	0.04	0.24	0.08	0.03	0.04	0.11	0.06	20.7
the United States																		
Children	Average	0.14	0.17	0.03	22.9	0.18	0.62	5.76	3.28	1.04	0.03	0.13	0.07	0.09	0.07	0.01	0.12	49.7
	High	0.49	1.57	0.21	70.3	0.86	15.9	55.9	17.8	7.07	0.99	0.68	3.96	0.49	0.52	0.16	2.18	138
Adult	Average	0.03	0.03	0.01	4.58	0.04	0.12	1.15	0.66	0.21	0.01	0.03	0.01	0.02	0.01	0.00	0.02	9.94
	High	0.10	0.31	0.04	14.1	0.17	3.18	11.2	3.56	1.41	0.20	0.14	0.79	0.10	0.02	0.03	0.44	27.6
Japan																		
Children	Average	0.07	0.16	0.02	40.1	0.17	1.99	3.37	3.10	0.41	0.27	0.16	0.09	0.08	0.01	0.01	0.05	61.7
	High	1.17	0.70	0.07	172	0.54	28.8	35.6	146	1.31	1.96	1.76	0.85	0.28	0.05	0.07	18.7	317
Adult	Average	0.02	0.04	0.01	9.24	0.04	0.46	0.78	0.71	0.09	0.06	0.04	0.02	0.02	0.00	0.00	0.01	14.2
	High	0.27	0.16	0.02	39.7	0.12	6.62	8.19	33.7	0.30	0.45	0.41	0.19	0.07	0.02	0.02	4.31	72.9
South Korea																		
Children	Average	0.29	0.27	0.06	48.2	0.19	5.15	1.49	0.16	1.08	0.08	0.27	0.09	0.35	0.01	0.02	0.04	64.8
	High	1.28	1.60	0.25	381	0.70	23.2	7.43	6.87	35.4	0.32	2.23	1.91	14.0	0.16	0.27	0.22	406
Adult	Average	0.07	0.06	0.01	11.1	0.04	1.19	0.34	0.04	0.25	0.02	0.06	0.02	0.08	0.00	0.00	0.01	14.9
	High	0.29	0.37	0.06	87.6	0.16	5.33	1.71	1.58	8.14	0.07	0.51	0.44	3.22	0.14	0.06	0.05	93.3

[a] Values are from Li et al. (2018).
[b] Median concentrations were used for calculating exposure dose.
[c] 95th percentile (P95) concentrations were used for calculating exposure dose.

$$EDI = \frac{C \times DIR}{BW} \quad (1)$$

where C is the median and 95th percentile concentrations of OPEs measured in indoor dust (ng/g), to estimate average and high exposure scenarios, respectively; DIR is the dust ingestion rate (g/day), and the average values used in this study were 60 mg/day for children and 30 mg/day for adults; and BW is the body weight (kg). The respective average body weights for children and adults in Western countries (the US, Greece, Romania, and Colombia) were 32 and 80 kg, whereas the corresponding values for Asian countries (China, Japan, South Korea, India, Pakistan, Vietnam, Kuwait, and Saudi Arabia) were 29 and 63 kg (Liao et al., 2012).

The EDIs of OPEs via indoor dust ingestion from the 12 countries are summarized in Table 3. The median exposure doses (based on median concentrations) of ΣOPEs in children and adults were in the ranges of 0.29–64.8 and 0.07–14.9 ng/kg bw/day, respectively. Owing to their lower body weights and more frequent hand-to-mouth contact (Hou et al., 2016), children are more highly exposed to OPEs than are adults via dust ingestion. The highest average exposure doses of ΣOPEs through dust ingestion were found in South Korea (adults: 14.9, children: 64.8 ng/kg bw/day), whereas the lowest EDIs were found in Pakistan (adults: 0.07, children: 0.29 ng/kg bw/day). Such regional difference may be related to consumption patterns and the regulation of OPEs among these countries (Hou et al., 2016; Wei et al., 2015).

The highest EDIs found in this study were much higher than those reported from Egypt (adults: 1.14, children: 14.8 ng/kg bw/day) (Abdallah and Covaci, 2014) and Pakistan (adults: 0.78, children: 15.0 ng/kg bw/day) (Ali et al., 2013). However, these EDI values were in the same order of magnitude or lower than those reported in New Zealand (adults: 11.1, children: 259 ng/kg bw/day) (Ali et al., 2013), Belgium (adults: 28.1, children: 548 ng/kg bw/day) (Van de Eede et al., 2011) and Germany (adults: 32.0, children: 160 ng/kg bw/day) (Brommer et al., 2012).

The EDIs of individual OPEs were 3–5 orders of magnitude below the corresponding reference doses (RfDs, Table 3). However, the average and high exposure doses of ΣOPEs in adults were found up to 14.9 and 93.3 ng/kg bw/day, respectively, which were much higher than those reported from drinking water consumption (average: 0.22, high: 1.25 ng/kg bw/day for adults) (Kim and Kannan, 2018), inhalation of particulate matter (average: 0.01–0.59 ng/kg bw/day for adults) (Yang et al., 2014), inhalation of air (high: 34 ng/kg bw/day for adults), dermal exposure from hand wipes (average: 0.32 ng/kg bw/day for adults) (Xu et al., 2016), and daily intake via fish consumption (average: 5.45 ng/kg bw/day for adults) (Kim et al., 2011) and accounted for 2.7–11% of total dietary intake through beverages, dairy products, grains, vegetables, meats, and fresh fruits (average: 539, high: 881 ng/kg bw/day for adults) (Zhang et al., 2016), indicating that indoor dust ingestion is a significant route for human exposure to OPEs.

Despite the fact that we report OPE concentrations and profiles in dust from 12 countries for the first time, this study has some limitations. First, only dust samples from living rooms and bedrooms were collected, whereas dust samples from other indoor microenvironments, such as offices, school buildings, stores, and hospitals, were not included in this study. Second, the sample size for individual countries was small. Nevertheless, we believe that the similar sampling techniques and analytical methods used in this study make direct comparisons of OPE concentrations in dust from various countries more reliable. Information regarding the global distribution of OPEs in the indoor environment is sparse and sporadic, and this study establishes baseline data for comparison with future studies.

4. Conclusions

In summary, this is the first study to report global occurrence and distribution of 20 OPEs in indoor dust from 12 countries. Cl-OPEs were the predominant OPEs found in indoor dust samples. The variations in ΣOPE concentrations and profiles indicated various consumption patterns and different regulations in regard to the use of OPEs among different countries. Significant correlations among several OPEs in indoor dust samples indicated their similar sources and concurrent applications in consumer products. The median EDI of ΣOPEs via dust ingestion in children and adults was considerably lower than the reference doses, suggesting that the current exposures to OPEs through indoor dust ingestion are not likely to pose risks to human health.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.105178.

References

Abdallah, M.A., Covaci, A., 2014. Organophosphate flame retardants in indoor dust from Egypt: implications for human exposure. Environ. Sci. Technol. 48, 4782–4789.

Ali, N., Ali, L., Mehdi, T., Dirtu, A.C., Al-Shammari, F., Neels, H., Covaci, A., 2013. Levels and profiles of organochlorines and flame retardants in car and house dust from Kuwait and Pakistan: implication for human exposure via dust ingestion. Environ. Int. 55, 62–70.

Ali, N., Shahzad, K., Rashid, M.I., Shen, H.Q., Ismail, I.M.I., Eqani, S., 2017. Currently used organophosphate and brominated flame retardants in the environment of China and other developing countries (2000-2016). Environ. Sci. Pollut. Res. 24, 18721–18741.

Behl, M., Hsieh, J.H., Shafer, T.J., Mundy, W.R., Rice, J.R., Boyd, W.A., Freedman, J.H., Hunter 3rd, E.S., Jarema, K.A., Padilla, S., Tice, R.R., 2015. Use of alternative assays to identify and prioritize organophosphorus flame retardants for potential developmental and neurotoxicity. Neurotoxicol. Teratol. 52, 181–193.

Behl, M., Rice, J.R., Smith, M.V., Co, C.A., Bridge, M.F., Hsieh, J.H., Freedman, J.H., Boyd, W.A., 2016. Comparative toxicity of organophosphate flame retardants and polybrominated diphenyl ethers to caenorhabditis elegans. Toxicol. Sci. 154, 241–252.

Brandsma, S.H., Sellstrom, U., de Wit, C.A., de Boer, J., Leonards, P.E., 2013. Dust measurement of two organophosphorus flame retardants, resorcinol bis(diphenylphosphate) (RBDPP) and bisphenol A bis(diphenylphosphate) (BPA-BDPP), used as alternatives for BDE-209. Environ. Sci. Technol. 47, 14434–14441.

Brommer, S., Harrad, S., Van den Eede, N., Covaci, A., 2012. Concentrations of. organophosphate esters and brominated flame retardants in German indoor dust samples. J. Environ. Monit. 14, 2482–2487.

Cao, Z., Xu, F., Covaci, A., Wu, M., Wang, H., Yu, G., Wang, B., Deng, S., Huang, J., Wang, X., 2014. Distribution patterns of brominated, chlorinated, and phosphorus flame retardants with particle size in indoor and outdoor dust and implications for human exposure. Environ. Sci. Technol. 48, 8839–8846.

Castro-Jimenez, J., Berrojalbiz, N., Pizarro, M., Dachs, J., 2014. Organophosphate ester (OPE) flame retardants and plasticizers in the open Mediterranean and black seas atmosphere. Environ. Sci. Technol. 48, 3203–3209.

Chen, M.H., Liu, Y.H., Guo, R.X., Xu, H.Z., Song, N.H., Han, Z.H., Chen, N.N., Zhang, S.H., Chen, J.Q., 2018. Spatiotemporal distribution and risk assessment of organophosphate esters in sediment from Taihu Lake, China. Environ. Sci. Pollut. Res. 25, 13787–13795.

Danish Environmental Protection Agency, 2016. Chlorinated phosphorous-based flame retardants in children's articles containing foam available: https://www2.mst.dk/Udgiv/publications/2016/05/978-87-93435-69-8.pdf, Accessed date: 10 July 2019.

De Boer, J., Ballesteros-Gomez, A., Leslie, H.A., Brandsma, S.H., Leonards, P.E.G., 2016. Flame retardants: dust - and not food - might be the risk. Chemosphere 150, 461–464.

Ding, J.J., Deng, T.Q., Xu, M.M., Wang, S., Yang, F.X., 2018. Residuals of organophosphate esters in foodstuffs and implication for human exposure. Environ. Pollut. 233, 986–991.

Dirtu, A.C., Ali, N., Van den Eede, N., Neels, H., Covaci, A., 2012. Country specific comparison for profile of chlorinated, brominated and phosphate organic contaminants in indoor dust. Case study for Eastern Romania, 2010. Environ. Int. 49, 1–8.

European Commission, 2011. Identification and Evaluation of Data on Flame Retardants in Consumer Products. available. https://www.ifv.nl/kennisplein/Documents/20110426-ec-arcadis-ebrc-evaluation-of-data-on-flame-retardants-in-consumer-products.pdf, Accessed date: 10 July 2019.

European Union, 2009. European Union. European union risk assessment report: Tris(2-chloroethyl) phosphate, TCEP. available. https://echa.Europa.Eu/documents/10162/2663989d-1795-44a1-8f50-153a81133258 (accessed Jul 10, 2019).

Fang, M., Webster, T.F., Gooden, D., Cooper, E.M., McClean, M.D., Carignan, C., Makey,

C., Stapleton, H.M., 2013. Investigating a novel flame retardant known as V6: measurements in baby products, house dust, and car dust. Environ. Sci. Technol. 47, 4449–4454.
Fries, E., Mihajlovic, I., 2011. Pollution of soils with organophosphorus flame retardants and plasticizers. J. Environ. Monitor. 13, 2692–2694.
Gao, L., Shi, Y., Li, W., Liu, J., Cai, Y., 2016. Occurrence and distribution of organophosphate triesters and diesters in sludge from sewage treatment plants of Beijing, China. Sci. Total Environ. 544, 143–149.
Green, N., Schlabach, M., Bakke, T., Brevik, E.M., Dye, C., Herzke, D., Huber, S., Plosz, B., Remberger, M., Schoyen, M., Uggerud, H.T., Vogelsang, C., 2008. Screening of Selected Metals and New Organic Contaminants 2007. Norwegian Pollution Control Agency available: http://www.miljodirektoratet.no/old/klif/publikasjoner/2367/ta2367.pdf, Accessed date: 10 July 2019.
Guo, J., Venier, M., Salamova, A., Hites, R.A., 2017. Bioaccumulation of Dechloranes, organophosphate esters, and other flame retardants in Great Lakes fish. Sci. Total Environ. 583, 1–9.
Han, Z., Wang, Q., Fu, J., Chen, H., Zhao, Y., Zhou, B., Gong, Z., Wei, S., Li, J., Liu, H., Zhang, X., Liu, C., Yu, H., 2014. Multiple bio-analytical methods to reveal possible molecular mechanisms of developmental toxicity in zebrafish embryos/larvae exposed to tris(2-butoxyethyl) phosphate. Aquat. Toxicol. 150, 175–181.
He, M.J., Lu, J.F., Ma, J.Y., Wang, H., Du, X.F., 2018. Organophosphate esters and phthalate esters in human hair from rural and urban areas, Chongqing, China: concentrations, composition profiles and sources in comparison to street dust. Environ. Pollut. 237, 143–153.
Hou, R., Xu, Y., Wang, Z., 2016. Review of OPFRs in animals and humans: absorption, bioaccumulation, metabolism, and internal exposure research. Chemosphere 153, 78–90.
Kademoglou, K., Xu, F., Padilla-Sanchez, J.A., Haug, L.S., Covaci, A., Collins, C.D., 2017. Legacy and alternative flame retardants in Norwegian and UK indoor environment: implications of human exposure via dust ingestion. Environ. Int. 102, 48–56.
Khan, M.U., Li, J., Zhang, G., Malik, R.N., 2016. New insight into the levels, distribution and health risk diagnosis of indoor and outdoor dust-bound FRs in colder, rural and industrial zones of Pakistan. Environ. Pollut. 216, 662–674.
Kim, U.J., Kannan, K., 2018. Occurrence and distribution of organophosphate flame retardants/plasticizers in surface waters, tap water, and rainwater: implications for human exposure. Environ. Sci. Technol. 52, 5625–5633.
Kim, J.W., Isobe, T., Chang, K.H., Amano, A., Maneja, R.H., Zamora, P.B., Siringan, F.P., Tanabe, S., 2011. Levels and distribution of organophosphorus flame retardants and plasticizers in fishes from Manila Bay, the Philippines. Environ. Pollut. 159, 3653–3659.
Kim, J.W., Isobe, T., Sudaryanto, A., Malarvannan, G., Chang, K.H., Muto, M., Prudente, M., Tanabe, S., 2013. Organophosphorus flame retardants in house dust from the Philippines: occurrence and assessment of human exposure. Environ. Sci. Pollut. Res. 20, 812–822.
Kim, J.W., Isobe, T., Muto, M., Tue, N.M., Katsura, K., Malarvannan, G., Sudaryanto, A., Chang, K.H., Prudente, M., Viet, P.H., Takahashi, S., Tanabe, S., 2014. Organophosphorus flame retardants (PFRs) in human breast milk from several Asian countries. Chemosphere 116, 91–97.
Kim, U.J., Oh, J.K., Kannan, K., 2017. Occurrence, removal, and environmental emission of organophosphate flame retardants/plasticizers in a wastewater treatment Plant in new York State. Environ. Sci. Technol. 51, 7872–7880.
Kim, U.J., Wang, Y., Li, W., Kannan, K., 2019. Occurrence of and human exposure to organophosphate flame retardants/plasticizers in indoor air and dust from various microenvironments in the United States. Environ. Int. 125, 342–349.
Li, J.F., Zhang, Z.Z., Ma, L.Y., Zhang, Y., Niu, Z.G., 2018. Implementation of USEPA RfD and SFO for improved risk assessment of organophosphate esters (organophosphate flame retardants and plasticizers). Environ. Int. 114, 21–26.
Liang, K., Shi, F.Q., Liu, J.F., 2018. Occurrence and distribution of oligomeric organophosphorus flame retardants in different treatment stages of a sewage treatment plant. Environ. Pollut. 232, 229–235.
Liao, C.Y., Liu, F., Guo, Y., Moon, H.B., Nakata, H., Wu, Q., Kannan, K., 2012. Occurrence of eight bisphenol analogues in indoor dust from the United States and several Asian countries: implications for human exposure. Environ. Sci. Technol. 46, 9138–9145.
Lu, S.Y., Li, Y.X., Zhang, T., Cai, D., Ruan, J.J., Huang, M.Z., Wang, L., Zhang, J.Q., Qiu, R.L., 2017. Effect of E-waste recycling on urinary metabolites of organophosphate flame retardants and plasticizers and their association with oxidative stress. Environ. Sci. Technol. 51, 2427–2437.
Luo, P., Bao, L.J., Guo, Y., Li, S.M., Zeng, E.Y., 2016. Size-dependent atmospheric deposition and inhalation exposure of particle-bound organophosphate flame retardants. J. Hazard. Mater. 301, 504–511.
Ma, Y., Xie, Z., Lohmann, R., Mi, W., Gao, G., 2017. Organophosphate Ester flame retardants and plasticizers in ocean sediments from the North Pacific to the Arctic Ocean. Environ. Sci. Technol. 51, 3809–3815.
Marklund, A., 2005. Levels and Sources of Organophosphorus Flame Retardants and Plasticizers in Indoor and Outdoor Environment. Umea University. available. https://www.diva-portal.org/smash/get/diva2:144103/FULLTEXT01.pdf (accessed Jul 10, 2019).
Marklund, A., Andersson, B., Haglund, P., 2003. Screening of organophosphorus compounds and their distribution in various indoor environments. Chemosphere 53, 1137–1146.
Matsukami, H., Nguyen Minh, T., Suzuki, G., Someya, M., Le Huu, T., Pham Hung, V., Takahashi, S., Tanabe, S., Takigami, H., 2015a. Flame retardant emission from e-waste recycling operation in northern Vietnam: environmental occurrence of emerging organophosphorus esters used as alternatives for PBDEs. Sci. Total Environ. 514, 492–499.
Matsukami, H., Suzuki, G., Takigami, H., 2015b. Compositional analysis of commercial oligomeric organophosphorus flame retardants used as alternatives for PBDEs: concentrations and potential environmental emissions of oligomers and impurities. Environ. Sci. Technol. 49, 12913–12921.
Matsukami, H., Suzuki, G., Someya, M., Uchida, N., Nguyen Minh, T., Le Huu, T., Pham Hung, V., Takahashi, S., Tanabe, S., Takigami, H., 2017. 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. Chemosphere 167, 291–299.
Möller, A., Sturm, R., Xie, Z., Cai, M., He, J., Ebinghaus, R., 2012. Organophosphorus flame retardants and plasticizers in airborne particles over the Northern Pacific and Indian Ocean toward the Polar Regions: evidence for global occurrence. Environ. Sci. Technol. 46, 3127–3134.
Ospina, M., Jayatilaka, N.K., Wong, L.Y., Restrepo, P., Calafat, A.M., 2018. Exposure to organophosphate flame retardant chemicals in the US general population: data from the 2013-2014 National Health and Nutrition Examination Survey. Environ. Int. 110, 32–41.
Rauert, C., Lazarov, B., Harrad, S., Covaci, A., Stranger, M., 2014. A review of chamber experiments for determining specific emission rates and investigating migration pathways of flame retardants. Atmos. Environ. 82, 44–55.
Reemtsma, T., Quintana, J.B., Rodil, R., Garcia-Lopez, M., Rodriguez, I., 2008. Organophosphorus flame retardants and plasticizers in water and air I. Occurrence and fate. Trac-Trend Anal. Chem. 27, 727–737.
Saito, I., Onuki, A., Seto, H., 2007. Indoor organophosphate and polybrominated flame retardants in Tokyo. Indoor Air 17, 28–36.
Salamova, A., Hermanson, M.H., Hites, R.A., 2014. Organophosphate and halogenated flame retardants in atmospheric particles from a European Arctic site. Environ. Sci. Technol. 48, 6133–6140.
Shi, Q.P., Wang, M., Shi, F.Q., Yang, L.H., Guo, Y.Y., Feng, C.L., Liu, J.F., Zhou, B.S., 2018. Developmental neurotoxicity of triphenyl phosphate in zebrafish larvae. Aquat. Toxicol. 203, 80–87.
Shoeib, T., Webster, G.M., Hassan, Y., Tepe, S., Yalcin, M., Turgut, C., Kurt-Karakus, P.B., Jantunen, L., 2019. Organophosphate esters in house dust: a comparative study between Canada, Turkey and Egypt. Sci. Total Environ. 650, 193–201.
Stubbings, W.A., Drage, D.S., Harrad, S., 2016. Chlorinated organophosphate and "legacy" brominated flame retardants in UK waste soft furnishings: a preliminary study. Emerg. Contam. 2, 185–190.
Sun, Y., Gong, X., Lin, W., Liu, Y., Wang, Y., Wu, M., Kannan, K., Ma, J., 2018. Metabolites of organophosphate ester flame retardants in urine from Shanghai, China. Environ. Res. 164, 507–515.
Tan, H., Chen, D., Peng, C., Liu, X., Wu, Y., Li, X., Du, R., Wang, B., Guo, Y., Zeng, E.Y., 2018. Novel and traditional organophosphate esters in house dust from South China: association with hand wipes and exposure estimation. Environ. Sci. Technol. 52, 11017–11026.
US EPA, 2008. Supporting Documents for Initial Risk-Based Prioritization of High Production Volume Chemicals Butylated Triphenyl Phosphate (CAS No. 220352-35-2) (9th CI Name: Phenol, tert-Butyl Derivatives, Phosphates (3:1)). Environmental Protection Agency, U.S. Available at: http://www.epa.gov/hpvis/rbp/220352352_Butylated%20Triphenyl%20Phosphate_Web_RBPSuppDocs.July2008.pdf, Accessed date: 10 July 2019.
US EPA, 2016. Chemical Data Access Tool (CDAT). Chemical Data Reporting Information. https://www.epa.gov/chemical-data-reporting/2016-chemical-data-reporting-results (accessed Jul 10, 2019).
Van de Eede, N., Dirtu, A., Neels, H., Covaci, A., 2011. Analytical developments and preliminary assessment of human exposure to organophosphate flame retardants from indoor dust. Environ. Int. 37, 454–461.
Van der Veen, I., de Boer, J., 2012. Phosphorus flame retardants: properties, production, environmental occurrence, toxicity and analysis. Chemosphere 88, 1119–1153.
Volkel, W., Fuchs, V., Wockner, M., Fromme, H., 2018. Toxicokinetic of tris(2-butoxyethyl) phosphate (TBOEP) in humans following single oral administration. Arch. Toxicol. 92, 651–660.
Wang, W., Abualnaja, K.O., Asimakopoulos, A.G., Covaci, A., Gevao, B., Johnson-Restrepo, B., Kumosani, T.A., Malarvannan, G., Minh, T.B., Moon, H.B., Nakata, H., Sinha, R.K., Kannan, K., 2015. A comparative assessment of human exposure to tetrabromobisphenol A and eight bisphenols including bisphenol A via indoor dust ingestion in twelve countries. Environ. Int. 83, 183–191.
Wang, Y., Kannan, P., Halden, R.U., Kannan, K., 2019. A nationwide survey of 31 organophosphate esters in sewage sludge from the United States. Sci. Total Environ. 655, 446–453.
Wei, G.L., Li, D.Q., Zhuo, M.N., Liao, Y.S., Xie, Z.Y., Guo, T.L., Li, J.J., Zhang, S.Y., Liang, Z.Q., 2015. Organophosphorus flame retardants and plasticizers: sources, occurrence, toxicity and human exposure. Environ. Pollut. 196, 29–46.
Wong, F., Suzuki, G., Michinaka, C., Yuan, B., Takigami, H., de Wit, C.A., 2017. Dioxin-like activities, halogenated flame retardants, organophosphate esters and chlorinated paraffins in dust from Australia, the United Kingdom, Canada, Sweden and China. Chemosphere 168, 1248–1256.
Xu, F., Giovanoulis, G., van Waes, S., Padilla-Sanchez, J.A., Papadopoulou, E., Magner, J., Haug, L.S., Neels, H., Covaci, A., 2016. Comprehensive study of human external exposure to organophosphate flame retardants via air, dust, and hand wipes: the importance of sampling and assessment strategy. Environ. Sci. Technol. 50, 7752–7760.
Yadav, I.C., Devi, N.L., Li, J., Zhang, G., 2018. Organophosphate ester flame retardants in Nepalese soil: spatial distribution, source apportionment and air-soil exchange assessment. Chemosphere 190, 114–123.
Yang, F., Ding, J., Huang, W., Xie, W., Liu, W., 2014. Particle size-specific distributions and preliminary exposure assessments of organophosphate flame retardants in office air particulate matter. Environ. Sci. Technol. 48, 63–70.

Zhang, X., Zou, W., Mu, L., Chen, Y., Ren, C., Hu, X., Zhou, Q., 2016. Rice ingestion is a major pathway for human exposure to organophosphate flame retardants (OPFRs) in China. J. Hazard. Mater. 318, 686–693.

Zhang, B., Lu, S., Huang, M., Zhou, M., Zhou, Z., Zheng, H., Jiang, Y., Bai, X., Zhang, T., 2018a. Urinary metabolites of organophosphate flame retardants in 0-5-year-old children: potential exposure risk for inpatients and home-stay infants. Environ. Pollut. 243, 318–325.

Zhang, T., Bai, X.Y., Lu, S.Y., Zhang, B., Xie, L., Zheng, H.C., Jiang, Y.C., Zhou, M.Z., Zhou, Z.Q., Song, S.M., He, Y., Gui, M.W., Ouyang, J.P., Huang, H.B., Kannan, K., 2018b. Urinary metabolites of organophosphate flame retardants in China: health risk from tris(2-chloroethyl) phosphate (TCEP) exposure. Environ. Int. 121, 1363–1371.

Zheng, C.L., Feng, S.S., Liu, P.P., Fries, E., Wang, Q.R., Shen, Z.X., Liu, H.X., Zhang, T.C., 2016. Sorption of organophosphate flame retardants on Pahokee peat soil. Clean-Soil Air Water 44, 1163–1173.

Zhong, M.Y., Wu, H.F., Mi, W.Y., Li, F., Ji, C.L., Ebinghaus, R., Tang, J.H., Xie, Z.Y., 2018. Occurrences and distribution characteristics of organophosphate ester flame retardants and plasticizers in the sediments of the Bohai and Yellow Seas, China. Sci. Total Environ. 615, 1305–1311.

Zhou, L., Hiltscher, M., Puttmann, W., 2017. Occurrence and human exposure assessment of organophosphate flame retardants in indoor dust from various microenvironments of the Rhine/Main region, Germany. Indoor Air 27, 1113–1127.