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DOI:
10.1016/j.chemosphere.2022.134730

Document Version
Peer reviewed version

Citation for published version (Harvard):
Waiyarat, S, Boontanon, SK, Boontanon, N, Fujii, S, Harrad, S, Drage, DS & Abdallah, MA-E 2022, 'Exposure, risk and predictors of hexabromocyclododecane and Tetrabromobisphenol-A in house dust from urban, rural and E-waste dismantling sites in Thailand', *Chemosphere*, vol. 302, 134730.
https://doi.org/10.1016/j.chemosphere.2022.134730

Exposure, Risk and Predictors of Hexabromocyclododecane and Tetrabromobisphenol-A in House Dust from Urban, Rural and E-Waste Dismantling Sites in Thailand

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Abbreviations [1]

[1] Hexabromocyclododecane; HBCDD, tetrabromobisphenol-A; TBBPA, estimated daily intake; EDI, brominated flame retardants; BFRs, Persistent Organic Pollutants; POPs, limits of quantification; LOQ

HIGHLIGHTS

- First report of HBCDD and TBBPA concentrations in house dust in Thailand
- TBBPA detected in all house dust samples and at higher concentrations than HBCDD
- TBBPA concentrations highest in dust from houses located in an e-waste dismantling site
- TBBPA originates mainly from various electronic appliances
- Thai toddlers exposed to higher TBBPA and HBCDD levels than children and adults

Abstract

In this study, for the first time, we determined concentrations of hexabromocyclododecane (HBCDD) and tetrabromobisphenol-A (TBBPA) in house dust and estimated human exposure to these substances in houses located in an e-waste dismantling site and in urban and rural residential areas of Thailand. The median HBCDD concentration in urban residential houses (2.10 ng g^{-1}) was similar to that in houses in an e-waste dismantling site (2.05 ng g^{-1}, $p>0.05$) and slightly higher than that in rural residential houses (1.11 ng g^{-1}, $p>0.05$). In contrast, significantly higher TBBPA concentrations were present in house dust from an e-waste dismantling site (median = 720 ng g^{-1}; range = 44–2,300 ng g^{-1}) compared to those in urban (68.6 ng g^{-1}; 3.5–300 ng g^{-1}, $p<0.001$) and rural residential areas (17 ng g^{-1}; 2.0–201 ng g^{-1}, $p<0.001$). TBBPA concentrations increased with the increasing presence of electronic devices and a decreasing distance to the e-waste dismantling site. These results suggest that e-waste dismantling activities may contribute to TBBPA contamination of house dust. The median estimated daily intake (EDI) of HBCDD and TBBPA through dust ingestion for toddlers exceeded that for children and adults. However, EDI values for HBCDD and TBBPA from all age groups were below the oral reference dose guideline value suggested by the US National Research Council and National Toxicology Program (NTP).

Keywords: Hexabromocyclododecane, tetrabromobisphenol-A, house dust, e-waste, human exposure

1. Introduction

Indoor pollution with brominated flame retardants (BFRs) is concerning because exposure to these substances is associated with significant health risks, particularly when exposure is high for sensitive age groups (Liagkouridis et al., 2015; Kweon et al., 2018; Bastiaensen et al., 2019). BFRs such as hexabromocyclododecane (HBCDD) and tetrabromobisphenol-A (TBBPA), have been widely used in many materials and consumer products (Abdallah et al., 2016; Kweon et al., 2018). HBCDD is mainly added to expanded and extruded polystyrene foams for thermal insulation of buildings, in addition to other minor applications in the back coating of fabrics, and high-impact polystyrene used in electronic equipment enclosures. It is toxic and characterised by long-range atmospheric transport, environmental persistence, and high bioaccumulation potential (Drage et al., 2015; Abdallah et al., 2016; Jo et al., 2017). Because HBCDD does not chemically bond to foam (and is thus termed an additive), it can be released and enter the environment during product manufacturing, consumer use, and waste disposal (Hassan and Shoeib, 2015). Once released, it may bioaccumulate in the fatty tissues of living organisms. It acts as an endocrine disruptor and can negatively affect the development of the nervous and reproductive systems (Fromme et al., 2014; Li et al., 2016). Although HBCDD was listed in Annex A of the Stockholm Convention on Persistent Organic Pollutants (POPs) in 2013, its production and use in expanded and extruded polystyrene building insulation foam was exempted, provided that such foam is clearly labelled and/or identifiable as containing HBCDD throughout its lifetime (UNEP, 2014; Sharkey et al., 2020). Thus, the potential for HBCDD contamination of the environment remains.

TBBPA is primarily used as a reactive flame retardant covalently bound to epoxy and polycarbonate resins. In addition, it has been applied as an additive flame retardant in acrylonitrile-butadiene-styrene plastic and high-impact polystyrene (Liu et al., 2016). TBBPA

may leach out from such products where it is used as an additive, causing widespread pollution of the environment (Malkoske et al., 2016; Liu et al., 2016: Wu et al., 2016). This is of concern, as it may have adverse health effects, such as: immunotoxicity, neurotoxicity, and disruption of the endocrine system, as demonstrated in laboratory animals (Zhou et al., 2014). Despite this, there are no regulatory restrictions on TBBPA production in Asia (Barghi et al., 2017).

The general population is exposed to HBCDD and TBBPA through a combination of inhalation (indoor and outdoor air), ingestion (dust and diet), and dermal contact (dust and consumer products). House dust plays an important role in human exposure to chemical pollutants and poses a potential risk to human health in indoor environments (Fromme et al., 2014; Barghi et al., 2017; Wang et al., 2018; Yadav et al., 2019). Chemicals associated with dust enter the human body through ingestion after hand-to-mouth contact, inhalation of resuspended dust, or direct absorption through the skin. Furthermore, toddlers are at a higher risk of exposure to chemicals associated with in house dust than adults because their hands more often come in contact with their mouth and they spend most of their time at home (Fromme et al., 2014; Bastiaensen et al., 2019: Gwon et al., 2021). Previous studies have reported the occurrence of HBCDD and TBBPA in house dust worldwide in residential areas (Abdallah et al., 2008; Dodson et al., 2012; Abdallah et al., 2016; Allgood et al., 2017; Barghi et al., 2017; Peng et al., 2017; Gwon et al., 2021), rural areas (Fromme et al., 2014), and houses in electronic waste (e-waste) dismantling sites (Wu et al., 2016). The latter location often contains elevated concentrations of flame retardants. Recently, Thailand has become one of the largest e-waste dismantling sites globally since China banned the import of plastic and e-waste in January, 2018 (Olafisoye et al., 2013; Zheng et al., 2013). Thus, e-waste dismantling sites in Thailand may be particularly polluted with HBCDD and TBBPA, and this problem requires greater attention.

Although HBCDD has been gradually phased out since 2013, recent studies have shown in house dust contamination with HBCDD at average concentrations of 3,700 ng g^{-1} and 46,000 ng g^{-1} in Turkey (Kurt-Karakus et al., 2017) and the UK (Drage et al., 2020), respectively. However, while there is one study in Thailand reporting concentrations of derivatives of TBBPA (TBBPA-DBPE) in indoor dust in e-waste recycling facilities in Ayutthaya and Nonthaburi province (Ali et al., 2011), data on concentrations of HBCDD and TBBPA in house dust in Thailand is currently lacking. Thus, in this study, we determined HBCDD and TBBPA concentrations in indoor dust samples collected from residential houses in an e-waste dismantling site, urban residential houses, and rural residential houses in Thailand. We identified potential factors associated with HBCDD and TBBPA concentrations in house dust. Finally, the exposure of general population groups (toddlers, children, and adults) to these compounds through dust ingestion was estimated and the risk arising from such exposure was assessed.

2. Materials and methods

2.1. Chemicals and reagents

Standard solutions of TBBPA, α-, β-, and γ-HBCDD, as well as isotopically labelled HBCDD standards, specifically: $^{13}C_{12}$-α-HBCDD, $^{13}C_{12}$-β-HBCDD, $^{13}C_{12}$-γ-HBCDD, and $^{13}C_{12}$-TBBPA (each with purity of ≥ 98%) were purchased from Cambridge Isotope Laboratories (Andover, MA, USA). d_{18}-γ-HBCDD was purchased from Wellington Laboratories (Guelph, ON, Canada). Solvents used for extraction and clean-up processes and analysis (e.g., hexane, dichloromethane, and methanol) were all HPLC grade chemicals obtained from Merck (Darmstadt, Germany). Concentrated sulfuric acid (98% purity) and silica gel 60 (0.063–0.200 mm) were supplied by Merck (Darmstadt, Germany). Indoor dust reference material SRM 2585

was obtained from the U.S. National Institute of Standards and Technology (NIST, Gaithersburg, MD, USA).

2.2. Sample collection

Indoor dust samples (n = 48) were collected between November 2019 and January 2021 from the living rooms of 13 houses in an e-waste dismantling site (northeastern region of Thailand), of 23 homes located in a typical urban area (Bangkok Metropolitan Region, Thailand), and of 12 houses in a rural area (Nakhon Si Thammarat Province, Thailand) (Fig. 1) The northeastern region of Thailand was selected as it has one of the networks of e-waste dismantling sites in Thailand. The network is characterised as informal and risky. Most people operate their houses as e-waste dismantling workshops and separate household appliances and electronic equipment. Recycling methods include the uncontrolled dismantling of common facility households, open burning, and dumping at unsafe e-waste landfill sites. The Bangkok Metropolitan Region was selected as the urban site as it is the centre of economic growth in Thailand and is characterised by rapid urbanisation and a large population. Many electrical devices, household appliances, and building materials are used in the daily lives of residents living in this urban area. For rural area, Nakhon Si Thammarat Province located in southern Thailand was selected. The area is surrounded by mountains and has a high standard of air quality. The main occupation in the province is fruit growing. There are many orchards, and the people maintain a rural lifestyle with usage of fewer electronic devices and pieces of furniture in their homes.

Before and after sampling in each house, the sampling instrument was cleaned thoroughly with water and an isopropanol-impregnated disposable wipe to minimise contamination. The samples were collected using a nylon sock with 25 μm pore size inserted into the nozzle of a

portable vacuum cleaner (Abdallah et al., 2008; Harrad et al., 2010; Abdallah et al., 2016; Drage et al., 2020). In brief, 1 m^2 of carpet was sampled for 2 min or if the room was not carpeted, a 4 m^2 area of the bare floor was vacuumed for 4 min. After sampling, the sock was closed, wrapped with aluminium foil, and sealed in a plastic bag. In the laboratory, each dust sample was passed through a pre-cleaned 250 μm mesh to remove coarse particles and then stored at −20 °C until analysis. During collection time, the number of electronic devices, pieces of furniture, and building characteristics in each sampled house were recorded using an observation checklist.

2.3. Sample extraction and clean-up

House dust samples were extracted following a previously described procedure (Abdallah et al., 2008; Harrad et al., 2010; Drage et al., 2020). The complete details of extraction and clean-up procedures are described in the supplementary information (SI1). Briefly, 100 mg of the sieved indoor dust sample was spiked with 25 ng of each of $^{13}C_{12}$-α-HBCDD, $^{13}C_{12}$-β-HBCDD, $^{13}C_{12}$-γ-HBCDD, and $^{13}C_{12}$-TBBPA used as internal (surrogate) standards. 7 mL of a hexane:dichloromethane mixture (1:1, v/v) was added, and the sample was vortexed for 5 min, sonicated at 20 °C for 30 min and centrifuged for 5 min at 3,500 rpm. The extraction procedure was repeated for another two cycles. The concentrated sulfuric acid was used to concentrate and treat the extracts. Then, the clear extracts from the top layer were purified through an SPE cartridge packed with 4 g of pre-cleaned acidified silica (44% concentrated sulfuric acid, w/w) with 3 x 1 mL hexane rinses. The extracts were eluted with 25 mL of a hexane:dichloromethane mixture (1:1, v/v). The eluents were evaporated and reconstituted in 200 μL of methanol containing 25 pg/μL d_{18}-γ-HBCDD as a recovery determination (or syringe) standard before LC-MS/MS analysis.

2.4. Instrumental analysis

Instrumental analysis was conducted using an Agilent 1200SL HPLC system coupled with an Agilent 6400 tandem mass spectrometer. An Agilent Pursuit XRS3 C18 reversed-phase analytical column (150 mm × 2.0 mm i.d., 3 μm particle size) was used to separate HBCDD isomers (α-, β-, and γ-HBCDD) and TBBPA. The column temperature and injection volume were maintained at 40 °C and 10 μL. The mobile phase was comprised of (A) 1:1 methanol/water and (B) methanol flowing at 0.15 mL/min to elute target compounds. The mass spectrometry was conducted in the electrospray ionisation negative-ion multiple reaction monitoring mode. More information on instrument analysis is presented in the supplementary information (SI2). The monitoring transition for the measurement of target compounds is summarised in Table S1 of supplementary information.

2.5. Quality assurance/quality control

All glassware was cleaned and rinsed with hexane and acetone and then heated at 400 °C for 5 h before use. One procedural blank was used to run every ten samples of the extraction batch and to check for contamination during the experimental processes. Field blanks (n=5) were also analysed, comprising sodium sulphate collected using a vacuum cleaner and sock in accordance with the procedures used to collect samples. No target compounds were detected in any procedural or field blanks. The accuracy and precision of our analytical procedure were evaluated by analysis of certified reference material SRM 2585 (n=5) from NIST (Table S2). A signal-to-noise ratio of 10:1 was assumed when calculating the limits of quantification (LOQs). The LOQs were 0.7 ng g^{-1} for HBCDD and 0.1 ng g^{-1} for TBBPA. The recovery values of internal standards for HBCDD and TBBPA in dust samples were between 83–124% and 75–110%, respectively.

2.6. Estimation of daily exposure

HBCDD and TBBPA concentrations in dust ingested by toddlers (12–24 months), children (5–11 years), and adults (>20 years) were determined under the "median" and "high-end" exposure scenarios. The estimated daily intake (EDI, ng kg^{-1} bw day^{-1}) of HBCDD and TBBPA through dust ingestion was calculated using the following equations (Hassan and Shoeib, 2015; Abafe and Martincigh, 2016; Peng et al., 2017):

$$EDI_{ingestion} = \Sigma \frac{C_{dust} \times EF \times IR}{BW} \quad (1)$$

Where C_{dust} is the concentration of the target contaminant in dust samples (ng g^{-1}), IR is the daily ingestion rate (g day^{-1}), EF is the estimated fraction of time spent within the house each day and BW is the body weight (kg). A standard body weight was used for toddlers (12 kg), children (31.8 kg) and adults (63 kg) (USEPA, 2011; Abdallah et al., 2016; Peng et al., 2017; Yu et al., 2012). We assumed 100% absorption of contaminants for dust ingested orally. The U.S. EPA guideline (U.S. EPA, 2011) recommends the same dust ingestion rate for everyone aged 1 to <21 years. Moreover, previous studies used the same ingestion rate for toddlers and children (Abdallah et al., 2016; Peng et al., 2017; Gwon et al., 2021). Thus, in this study, the same dust ingestion rate for toddlers and children was applied in the calculations. Median and high exposure scenarios were considered, which assumed dust intake rates of 0.05 and 0.2 g day^{-1} for toddlers, 0.05 and 0.2 g day^{-1} for children, and 0.02 and 0.05 g day^{-1} for adults (Abdallah et al., 2016; Peng et al., 2017; Corsolini et al., 2021; Gwon et al., 2021). Based on previous research, it was assumed that toddlers spent an average of 86.1% of their time in the house, children 79.2% and adults 63.8% (Abdallah et al., 2008; Yu et al., 2012; Abdallah et al., 2016; Peng et al., 2017)

2.7. Statistical analyses

Descriptive statistics, including minimum, maximum, mean, median, and standard deviation, were calculated using Microsoft Office Excel 2016. Statistical comparisons were performed using IBM SPSS version 21. Due to the small sample size from each site in this study, preliminary testing for normality data of HBCDD and TBBPA concentration was performed with a Shapiro-Wilk test with a significance level of 0.05. TBBPA concentrations were normally distributed after log-transformation, and a one-way ANOVA was conducted to compare TBBPA concentrations from the three sampled areas. HBCDD concentrations had a non-normal distribution before and after log-transformation, so the non-parametric Kruskal-Wallis test was used to assess the differences of HBCDD concentrations between the three sample areas. A p-value < 0.05 was considered statistically significant. A multiple linear regression model was used to determine factors associated with HBCDD and TBBPA concentrations in house dust. In all cases, the concentration below the LOQ was replaced by zero to conduct the statistical analysis.

3. Results and discussion

3.1. HBCDD and TBBPA concentrations in house dust

HBCDD and TBBPA concentrations in house dust from houses located in e-waste dismantling site, and in urban and rural residential areas are presented in Table 1. HBCDD and TBBPA concentrations in house dust samples in our study were non-normally distributed data. Thus, median concentration was used to report and compare our results with those of other studies. Briefly, HBCDD was detected in 15 out of 45 samples, whereas TBBPA was detected in all samples. The median concentration of TBBPA exceeded that of HBCDD in all sample areas. Median concentrations of TBBPA and HBCDD were 720 and 2.05 ng g^{-1}, respectively, in houses located in e-waste dismantling site, 68.6 and 2.10 ng g^{-1}, respectively, in urban residential houses, and 16.5 and 1.11 ng g^{-1}, respectively, in rural residential houses. It can be

clearly observed that TBBPA concentration in house dust was higher than that of HBCDD concentration in our study. This contrasted with observations in previous studies from other locations around the world that reported an opposite trend, that is, HBCDD concentration in house dust was higher than that of TBBPA concentration (Abdallah et al., 2008; Fromme et al., 2014; Stapleton et al., 2014; Abdallah et al., 2016; Allgood et al., 2017). It is possible that in some of those studies, in house dust was sampled before HBCDD was listed under Annex A of the Stockholm Convention on POPs in 2013, when HBCDD was used in larger volumes globally (Fromme et al., 2014), which could explain why HBCDD concentrations in house dust exceeded those of TBBPA. In contrast, in our study, we collected samples after HBCDD use had been banned for almost six years. Moreover, a recent report (MTEC & NSTDA, 2021) revealed that HBCDD is only used in small quantities in polystyrene building insulation foam, such as sandwich panels, in applications for cold storage and clean rooms. In addition, Thailand is a tropical country and heat insulating building material is not frequently used in houses in Thailand. Furthermore, based on available data, HBCDD has not been imported to Thailand since 2017 (MTEC & NSTDA, 2021). These reasons may explain why HBCDD concentrations in house dusts were low in this study.

3.1.1. TBBPA

Descriptive statistics of TBBPA concentrations in house dust from houses in an e-waste dismantling site, urban residential houses, and rural residential houses, as well as a global comparison are given in Table 1. TBBPA was detected at concentrations above the LOQ in all samples. A one-way ANOVA test showed that TBBPA concentrations in house dust samples collected from houses in the e-waste dismantling site (ranging from 44.0 to 2,300 ng g^{-1}, median 720 ng g^{-1}) were 10 to 30 times higher than in samples from the urban residential houses (median 68.6 ng g^{-1}; $P < 0.001$) and rural residential houses (median 16.5 ng g^{-1}; $P <$

0.001; Fig. 2 b). There was a significant difference between concentrations of TBBPA in samples from urban and rural residential houses (P = 0.009). 70–90% of TBBPA produced is used as a reactive BFR in epoxy, polycarbonate and phenolic resins on printed circuit boards, with 10%–20% used as an additive BFR in plastics (Liu et al., 2016). The highest quantities of TBBPA are found in television and computer casings and the components of office electronic equipment (Covaci et al., 2009). In additive applications, TBBPA does not react chemically with the polymer material and may therefore migrate out of the matrix into the indoor environment due to volatilisation as well as abrasion (Abdallah et al., 2016; Wu et al., 2016; Wang et al., 2018). Notably, living rooms in urban residential houses usually contain several furniture items such as sofas, tables, chairs, and electrical appliances. In addition, the size of living rooms in urban residential houses is small, and most of them use air conditioners and open windows for ventilation. On the contrary, the houses in the e-waste dismantling site and rural areas in our study had few electronic devices and rarely had any furniture in the living room. Moreover, the living rooms in e-waste dismantling site and rural areas were more spacious and had open doors and windows throughout the day for natural ventilation. However, the houses located in the e-waste dismantling site, approximately 5–100 m close to the e-waste dismantling workshop, contained considerable e-waste suggesting that TBBPA is released from e-waste, transported and absorbed to house dust in living rooms. Thus, TBBPA concentrations in house dust in the e-waste dismantling site were higher than in house dust from the urban and rural residential areas.

Most previous reports on e-waste dismantling sites analysed TBBPA concentrations in e-waste workplace dust. House dust samples from a residential house in an e-waste dismantling site have been analysed in only one study from China, which reported a median TBBPA concentration of 10,329 ng g^{-1} (Wu et al., 2016). This was fourteen times higher than that

reported in our study. This is because the houses in China were located in a large e-waste recycling industrial park. Unregulated e-waste recycling activities separate plastic casings from household appliances and electronic devices. The e-waste dismantling site in our study is smaller than the e-waste recycling industrial park in China. Thus, although the e-waste dismantling practises in this study are similar as those in China, the quantity of e-waste is smaller than that in China. Moreover, in this study, the sample collection period coincided with the rice harvesting period. Thus, less e-waste is imported into the area than outside the rice harvesting period. All these factors contributed to the lower TBBPA concentrations detected in house dust in the e-waste dismantling site of our study compared to that in China. The median TBBPA concentrations in urban residential houses in this study (median 68.6 ng g^{-1}) were three times lower than those previously documented in the USA (median 200 and 187 ng g^{-1}) (Dodson et al., 2012; Allgood et al., 2017) and in South Africa (median 120 ng g^{-1}) (Abafe and Martincigh, 2016). This implies that living rooms in the USA and South Africa might have more electronic devices than the living rooms in our study. Therefore, TBBPA in house dust in our study is lower than in other studies. However, other studies, which analysed samples obtained in Nigeria, the UK, and South Korea, reported the following median TBBPA concentrations: 50 (Abdallah et al., 2016), 62 (Abdallah et al., 2008), 69 (Kweon et al., 2018), and 78.9 ng g^{-1} (Barghi et al., 2017), respectively, similar to the median value observed in our study. Furthermore, the median concentration of TBBPA in our study was 2–8 fold higher than those reported in urban residential house dust in the USA (median 7.9 ng g^{-1}) (Stapleton et al., 2014), Kazakhstan (median 13 ng g^{-1}) (Abdallah et al., 2016), and the UK (median 35 ng g^{-1}) (Drage et al., 2020). However, the median TBBPA concentration in rural residential house dust in our study, 16.5 ng g^{-1}, was close to the level of that recorded in Germany (28 ng g^{-1}) (Fromme et al., 2014).

3.1.2. HBCDD

The non-parametric Kruskal-Wallis test showed that concentrations of HBCDD (the sum of α, β, and γ-HBCDD isomers) in house dust were not significantly different between the three areas (i.e., urban, rural, and e-waste dismantling areas; P = 0.398; Fig. 2a). HBCDD concentrations ranged from <0.7 to 528 ng g^{-1} (median 2.10 ng g^{-1}) in the urban residential houses, from <0.7 to 29.8 ng g^{-1} (median 2.05 ng g^{-1}) in the houses in e-waste dismantling site, and from <0.7 to 126 ng g^{-1} (median 1.11 ng g^{-1}) in the rural residential houses. This may be attributable to the small number of locations sampled in this study. A larger sample size would be required to accurately reflect the true range and variation in concentrations of HBCDD between the three different sampling location categories. Remarkably, the highest median and maximum HBCDD concentrations were found in urban house dust sampled at a new condominium less than one year-old. In this home, the living room was covered with carpet and connected to the kitchen without a partition. There were also many pieces of furniture and items such as: fabric sofas, cushions, dolls, double curtains, a television, and a computer. The residents relied on air-conditioning throughout the day as the windows provided poor ventilation. In contrast with other sampled houses, there were fewer electronic devices. Most houses contained more open space with less furniture. Moreover, one known use of HBCDD is as a back coating for fabric covers for foam-filled furniture (Drage et al., 2018). Thus, our study found a high HBCDD concentration in only one house dust sample collected from rooms with several upholstery fabric furniture items.

Previous studies on HBCDD in house dust reported higher median HBCDD concentrations than those observed in our study. Briefly, median HBCDD concentrations in Europe were 1,300 ng g^{-1} in the UK (Abdallah et al., 2008), 270 ng g^{-1} in Turkey (Kurt-Karakus et al., 2017), 280 ng g^{-1} in UK (Drage et al., 2020), and 129 ng g^{-1} in Spain (Corsolini et al., 2021). In the

USA, median HBCDD levels were 160 and 338 ng g^{-1} (Dodson et al., 2012; Stapleton et al., 2014). For comparison, in house dust samples collected from Africa in Egypt and Nigeria contained median HBCDD concentrations of 6.15 (Hassan and Shoeib, 2015) and 405 ng g^{-1} (Abdallah et al., 2016), respectively. Furthermore, house dust samples collected in Asia showed median HBCDD concentrations of 106 ng g^{-1} in Korea (Barghi et al., 2017) and 0.20 ng g^{-1} in China (Peng et al., 2017). These results demonstrate that HBCDD in house dust in Europe, America, and Africa exceeded that in Asia. This may be because Europe was a bigger consumer of HBCDD-containing products than Asia (Barghi et al., 2017), specifically before HBCDD was listed in the Stockholm Convention on POPs in 2013 (UNEP, 2015; Stockholm Convention, 2019). However, HBCDD was listed in Annex A with an exemption to use HBCDD in expanded and extruded polystyrene building insulation foam in many countries. Such foam is clearly labelled and/or identifiable as containing HBCDD throughout its lifetime (UNEP, 2014; Sharkey et al., 2020). Therefore, to date, HBCDD continues to contaminate in house dust globally. The lower HBCDD levels in Asia may also be because HBCDD is rarely used in electronic appliances in Asia (Wang et al., 2018). While one study suggested carpets to be a source of HBCDD in house dust (Barghi et al., 2017), our study found that only a few households used carpets in their homes. Moreover, HBCDD usage in several applications has been reported to be low in Thailand's POP information (MTEC & NSTDA, 2021). In particular, polystyrene building insulation foam (which is a substantial application of HBCDD), is rarely used in Thailand. Therefore, it is not surprising that the concentrations of HBCDD in Thai house dust are lower than those in most other countries.

In general, γ-HBCDD is predominant in commercial mixtures, followed by α-HBCDD and β-HBCDD (Covaci et al., 2006; Abdallah et al., 2016; Wang et al., 2018). However, our analysis showed higher percentages of α-HBCDD (43–55%) compared to those of γ-HBCDD (33–38%)

and β-HBCDD (9–25%) in house dust from houses in an e-waste dismantling site and in the urban residential area (Fig. 3). Similarly, α-HBCDD was predominant in house dust collected in Germany (Fromme et al., 2014), France (Abdallah et al., 2016), Nigeria (Abdallah et al., 2016), Korea (Kweon et al., 2018), the UK (Drage et al., 2020), and Spain (Corsolini et al., 2021). Nevertheless, we observed a slight difference in the proportion of γ-HBCDD in rural residential house dust (γ- HBCDD followed by α-HBCDD and β-HBCDD, respectively) in our study, similar to the results of other studies conducted in Korea (Barghi et al., 2017) and the USA (Dodson et al., 2012). In brief, most of the studies found predominance of α-HBCDD in house dust samples. It is possible that γ-HBCDD transforms into α-HBCDD through various pathways. Such transformation has been shown to occur upon the addition of γ-HBCDD to expanded and extruded polystyrene as a flame retardant at thermal exposure (140–160 °C) (Heeb et al., 2010). Furthermore, conversion of γ-HBCDD into α-HBCDD has been described in dust samples after exposure to natural sunlight (Harrad et al., 2009). In addition, the degradation of γ-HBCDD was faster than that of α-HBCDD in dust samples (Howard and Muir, 2010). Consequently, there may be several explanations for the relatively higher proportion of α-HBCDD observed in our dust samples.

3.2 Association between the levels of HBCDD and TBBPA with housing characteristics

Multiple linear regression analyses were used to assess the housing characteristics associated with HBCDD and TBBPA concentrations in house dust. The data included eight categories of variables (Table S3 and Table S4): building structure, house age, floor type, number of electronic devices, type of furniture, ventilation usage, cleaning frequency, and distance from an e-waste dismantling site. The association analysis between HBCDD concentrations in house dust and residence characteristics is presented in Table 2. Floor type was significantly associated with HBCDD concentration in the multivariate data analysis ($P = 0.004$). However,

our study had a small sample size, and HBCDD was only detected in 15 out of 48 samples, with the non-detected replaced by zero in the other 33 samples. Moreover, 44 houses had PVC tile flooring, while only 4 had carpet. In our study, higher HBCDD concentrations were associated with hard floors. Previous research including timber and PVC flooring did not find that either one had significantly higher concentrations of contaminants, but associations were found between building type and house dust (Kweon et al., 2018). Therefore, further studies with a larger sample size and a significant number of subfactors should be conducted to further explore the potential association between flooring materials and contaminant concentrations.

For TBBPA, the critical factors impacting its concentrations in residential house dust were the number of electronic devices and distance from an e-waste dismantling workshop. TBBPA concentrations in living rooms with many electronic appliances were significantly higher ($P < 0.05$) than in rooms with few electronic items. Moreover, the distance of the houses to an e-waste dismantling site was strongly negatively related to TBBPA concentrations in house dust ($P < 0.05$). A similar relationship between the number of electronic devices and high TBBPA concentration was also reported in Korean houses (Barghi et al., 2017), Chinese houses (Wang et al., 2018), and Vietnamese e-waste workshops (Wannomai et al., 2020).

3.3 Estimated daily exposure dose to HBCDD and TBBPA through indoor dust

Estimated daily intakes (EDIs) of HBCDD and TBBPA through dust ingestion by the Thai general population living in an e-waste dismantling site and in urban and rural residential areas were calculated based on the concentrations of these substances in respective house dust samples. We assumed 100% absorption of HBCDD and TBBPA from dust ingestion. The EDI values for the three population groups were calculated under the median and high-end exposure scenarios, which were evaluated, respectively, from the median and 95th percentile

concentrations of HBCDD and TBBPA in house dust samples. The comparison of EDI values between the two scenarios for toddlers, children, and adults is illustrated in Fig. 4 and 5. It should be noted that HBCDD and TBBPA EDI values for toddlers were higher than those for children and adults. The estimated median exposure to HBCDD was 3–9 times higher in toddlers than in children and adults. Toddlers living in urban residential areas received slightly different exposure levels compared to those living in e-waste dismantling sites or in rural residential areas. In the high exposure scenario, toddlers living in urban and rural residential areas or in e-waste dismantling sites would be exposed to HBCDD concentrations of 6.17, 1.51, and 0.41 ng kg^{-1} bw day^{-1}. Remarkably, our study found that the level of exposure to HBCDD in urban residents was slightly higher than that in residents of e-waste dismantling sites and considerably higher than that in the residents of rural areas. Consequently, the number of furniture items owned by residents may affect HBCDD concentrations in house dust samples (U.S. EPA, 2014; Kurt-Karakus et al., 2017). In particular, we observed during dust sampling that living rooms in urban houses had a greater variety of furniture than rooms in other analysed types of residences. Thus, the population living in urban residential houses may be more greatly exposed to HBCDD than those living in e-waste dismantling sites and in rural areas. Nonetheless, the general exposure of Thai residents, including toddlers, to HBCDD was lower than that reported for several other countries (Table 3). Toddlers in France (Abdallah et al., 2016), the UK (Abdallah et al., 2008), and Germany (Fromme et al., 2014) have HBCDD exposure levels 95, 80, and 19 times higher than Thai toddlers. This is likely because, before 2013, Europe consumed considerable amounts of HBCDD, which maintained toddler exposure to HBCDD at a higher level than that in Asia. After 2013, the amounts of HBCDD decreased worldwide since they were listed in POPs. In a more recent study, Barghi et al. (2017) showed that Korean toddlers are exposed to HBCDD at 4.79 ng kg^{-1} bw day^{-1}, exceeding the exposure of Thai toddlers. Furthermore, our study estimated that HBCDD EDI values in all areas for

toddlers, children, and adults were lower than those in previous studies (Abdallah et al., 2008; Fromme et al., 2014; Abdallah et al., 2016; Barghi et al., 2017) and the reference dose (RfD) (200,000 ng kg^{-1} bw day^{-1}) suggested by the US-National Research Council (U.S. NRC, 2000). However, the RfD for HBCDD was calculated by the US National Academy of Sciences (NAS), using data from an unpublished subchronic study performed in rats in 1970 (Zeller and Kirsch, 1970). The NAS concluded that confidence in this RfD for HBCDD is low, because of a lack of other subchronic and chronic studies.

For TBBPA, the highest median and high-end exposure scenario EDIs were also for toddlers. Briefly, toddlers living in all three areas surveyed showed a similar trend in TBBPA exposure compared to other age groups. The estimated level of exposure to TBBPA in toddlers was between 3 and 18 times higher than the levels of exposure in children and adults under the median exposure scenario. A comparison of TBBPA exposure levels of toddlers in the three areas under the median exposure scenario is illustrated in Fig. 5. According to this scenario, toddlers living in houses in e-waste dismantling sites experienced significantly higher exposure (2.58 ng kg^{-1} bw day^{-1}) than toddlers in the urban (0.24 ng kg^{-1} bw day^{-1}) and rural areas (0.06 ng kg^{-1} bw day^{-1}). Under the high exposure scenario, the highest TBBPA exposure was predicted in toddlers living in e-waste dismantling sites (median level of 25 ng kg^{-1} bw day^{-1}). However, this level of exposure was substantially lower than the oral RfD (600,000 ng kg^{-1} bw day^{-1}) that has been observed to cause uterine hyperplasia in rats (NTP, 2013; Wikoff et al., 2015). Currently, some research has revealed that dysfunction of the liver is a health concern, following exposure to TBBPA, even at very low, environmentally relevant concentrations (30,000 ng kg^{-1} body weight) (Yao et al., 2021). Therefore, chronic effects induced by continuous exposure to TBBPA and its derivatives should be further investigated.

Our results indicated that toddlers are exposed to higher levels of TBBPA than older children and adults. In particular, toddlers living in e-waste dismantling sites may have higher potential health risks associated with TBBPA exposure through dust ingestion. A comparison of our findings with the findings of previous studies revealed a trend of higher EDIs of TBBPA for urban residents. Previous results were similar to ours: toddlers in South Africa (Abafe and Martincigh, 2016) and Korea (Barghi et al., 2017) were estimated to be exposed to TBBPA at higher levels than children and adults. Abdallah et al. (2016) reported average exposure of French toddlers to TBBPA of 5.6 ng day^{-1} from various indoor dust types, including house, office, and car dust. It is worth noting that the equation used by Abdallah et al. (2016) was different to the one used in the present study in that they did not report exposure per kg body weight. Assuming an average toddler body weight of 12 kg was used in the present study, the average exposure of French toddlers to TBBPA via indoor dust would be 0.47 ng kg^{-1} bw d^{-1}. Thus, French toddlers were exposed to TBBPA levels that were approximately two times higher than those experienced by Thai toddlers. With regards to the residents of e-waste dismantling sites, Wu et al. (2016) collected Chinese house dust samples in an area surrounding a large e-waste recycling processing park. The median concentration of TBBPA in Chinese house dust was fourteen times higher than that in Thai house dust. Thus, Chinese residents were exposed to TBBPA levels sixty times higher than those estimated for Thai residents. This may be because Wu et al. (2016) used the sum of TBBPA concentrations in indoor and outdoor dust to calculate the EDI of TBBPA. The general population can be exposed to TBBPA from indoor and outdoor dust because of their daily activities. Moreover, the surveyed Chinese residents lived in an e-waste recycling processing park that was a source of TBBPA. They were more likely to be exposed to TBBPA from indoor and outdoor dust than the general population living outside e-waste recycling processing parks. Although we only collected samples of indoor house dust, our data also suggests that e-waste dismantling site could be a significant

source of TBBPA. Thai residents living in e-waste dismantling sites are more likely to be exposed to TBBPA than the rest of the population. Further studies should examine the levels of TBBPA in both indoor and outdoor dust to more fully reflect daily human experience. In addition, in daily life, people usually do not stay at home all the time, but engage in various activities in many places, so it is essential to study the exposure to TBBPA from office, school, car, and other dust types. Moreover, people are likely to be exposed to TBBPA not only through dust ingestion, but also through other pathways. Therefore, additional studies are needed to examine the exposure to TBBPA from such other routes, which would enable a more conclusive assessment of total levels of human exposure to TBBPA.

4. Conclusions

In this study, 48 house dust samples were collected from houses in an e-waste dismantling site and urban and rural residential areas for the determination of HBCDD and TBBPA concentrations and an assessment of human exposure via dust ingestion and possible associated health risks. To the best of our knowledge, this is the first study of concentrations of these BFRs in Thai house dust. Our results show HBCDD is present at lower concentrations than TBBPA. The median TBBPA concentration was highest (720 ng g^{-1}) in residential houses located in e-waste dismantling sites, implying that e-waste dismantling activities significantly increase TBBPA concentrations in house dust. Correlation analysis between HBCDD and TBBPA concentrations and housing characteristics indicated that flooring type, number of electronic devices, and distance from e-waste dismantling shops might be associated with higher concentrations of these compounds in house dust. The small sample size in each residential area limited our study, and the strong correlations reported in this study must be verified with a larger sample size. The EDI of HBCDD and TBBPA via house dust was much higher in toddlers than in children and adults under both median and high-end scenarios.

Therefore, it is important to be aware of human exposure to HBCDD and TBBPA in Thailand and its potential adverse health effects, especially in toddlers. Further studies are required to determine the occurrence of other emerging flame retardants in house dust and potential health risks from co-exposure to other contaminants through various pathways. Such studies will improve our understanding of the health impacts of these chemicals.

Acknowledgements

This study was supported by research funding from the Royal Golden Jubilee (RGJ) PhD. Programme scholarship of the Thailand Research Fund (PHD/0129/2559) and Fundamental Fund (BRF2-NDFR29/2564) from Mahidol university, and the On-site Laboratory Initiative by Kyoto University, Japan.

Author Contribution Statement

Sonthinee Waiyarat: Conceptualization, Funding acquisition, Sample collection, Methodology, Formal analysis, Data curation, Writing-original draft preparation, Writing review and editing. Suwanna Kitpati Boontanon: Conceptualization, Investigation, Funding acquisition, Project administration, Supervision, Writing review and editing. Narin. Boontanon: Methodology and Validation, Writing review and editing. Shigeo Fujii: Conceptualization, Writing review and editing. Stuart Harrad: Conceptualization, Methodology, Writing review and editing. Daniel Simon Drage: Methodology, Writing review and editing. Mohamed Abou-Elwafa Abdallah: Methodology, Writing review and editing.

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Exposure, Risk and Predictors of Hexabromocyclododecane and Tetrabromobisphenol-A in House Dust from Urban, Rural and E-Waste Dismantling Sites in Thailand

Sonthinee Waiyarat [a], Suwanna Kitpati Boontanon [a, b*], Narin Boontanon [c], Shigeo Fujii [b], Stuart Harrad [d], Daniel Simon Drage [d, e], Mohamed Abou-Elwafa Abdallah [d]

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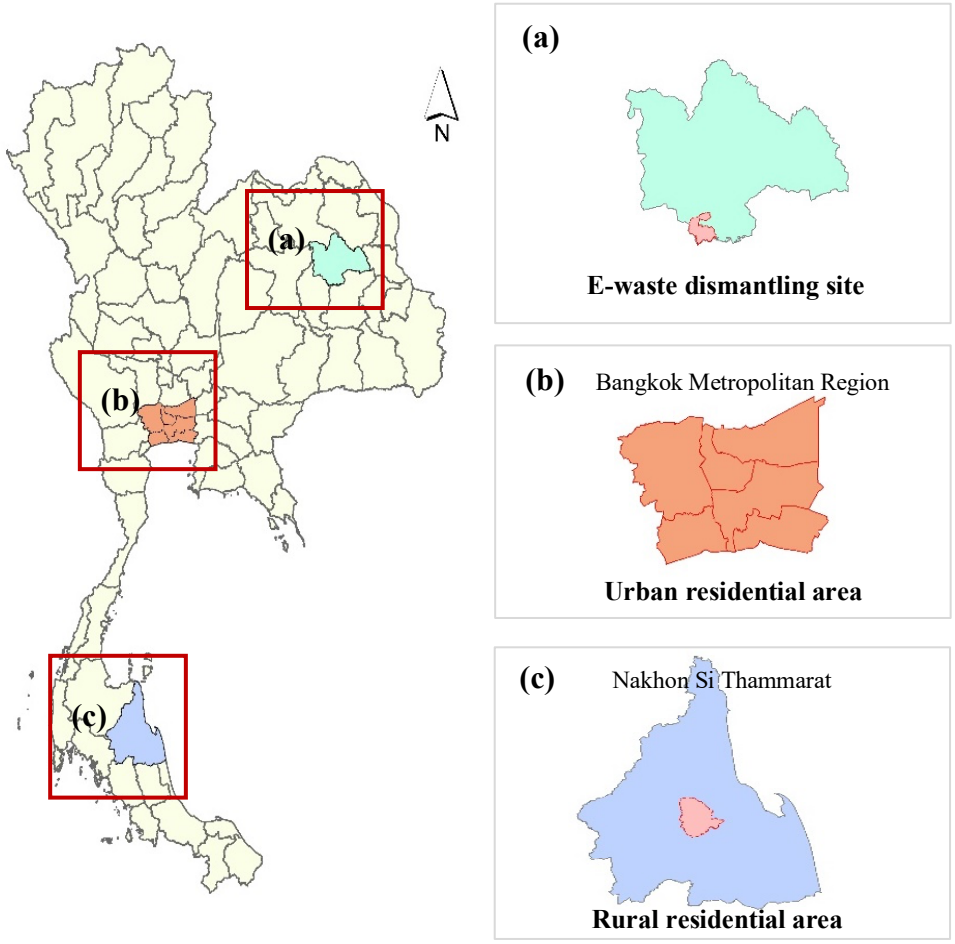

Fig. 1. Map of Thailand showing locations of sampling areas.

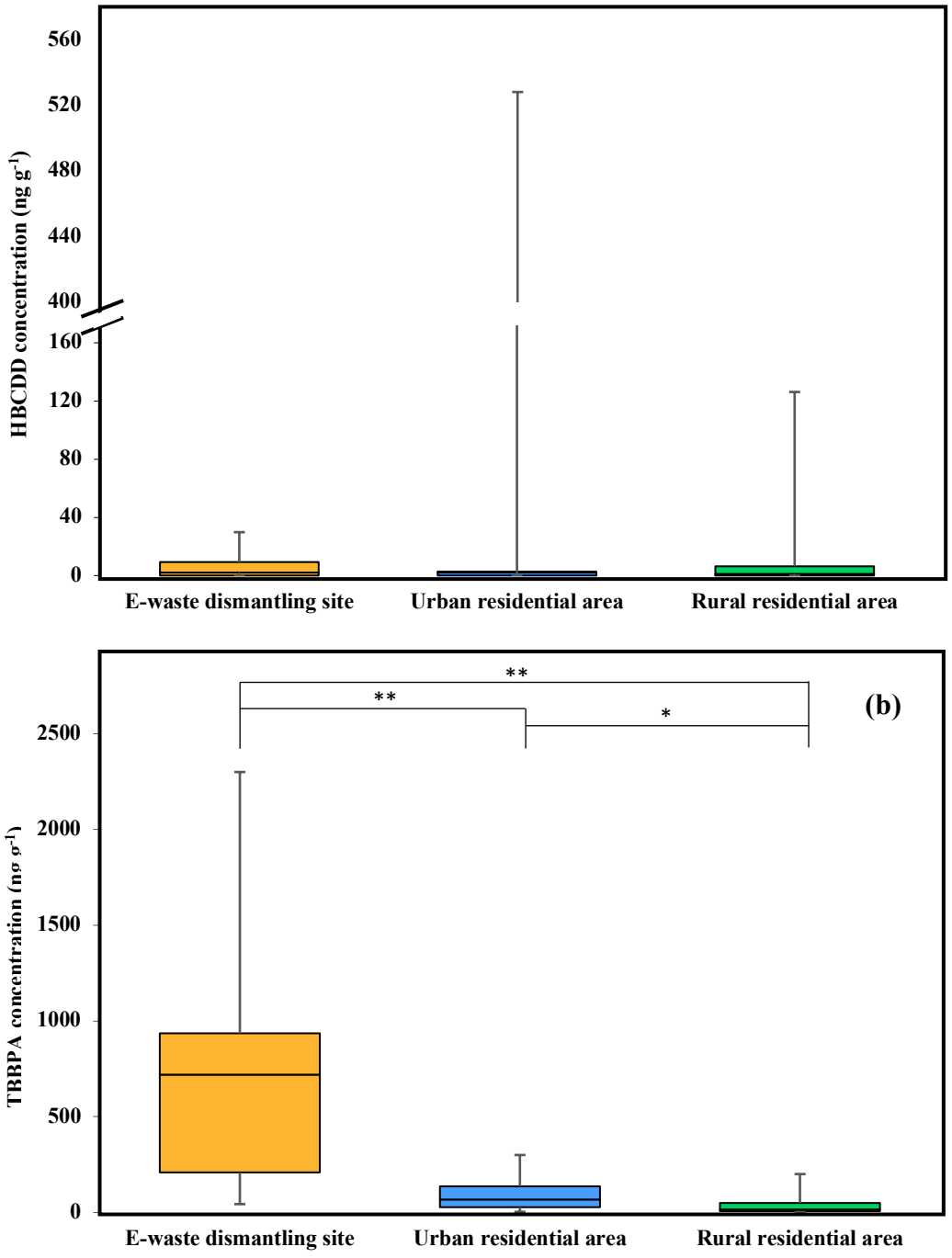

Fig. 2. Median concentrations of (a) HBCDD and (b) TBBPA in house dust from the three areas in Thailand. The horizontal line in each box is the median; the lower and upper boundaries of each box are the 25th and 75th percentiles, respectively. Error bars indicate the minimum and maximum values. Statistical significance of differences is indicated as follows: *$P<0.05$; **$P < 0.001$.

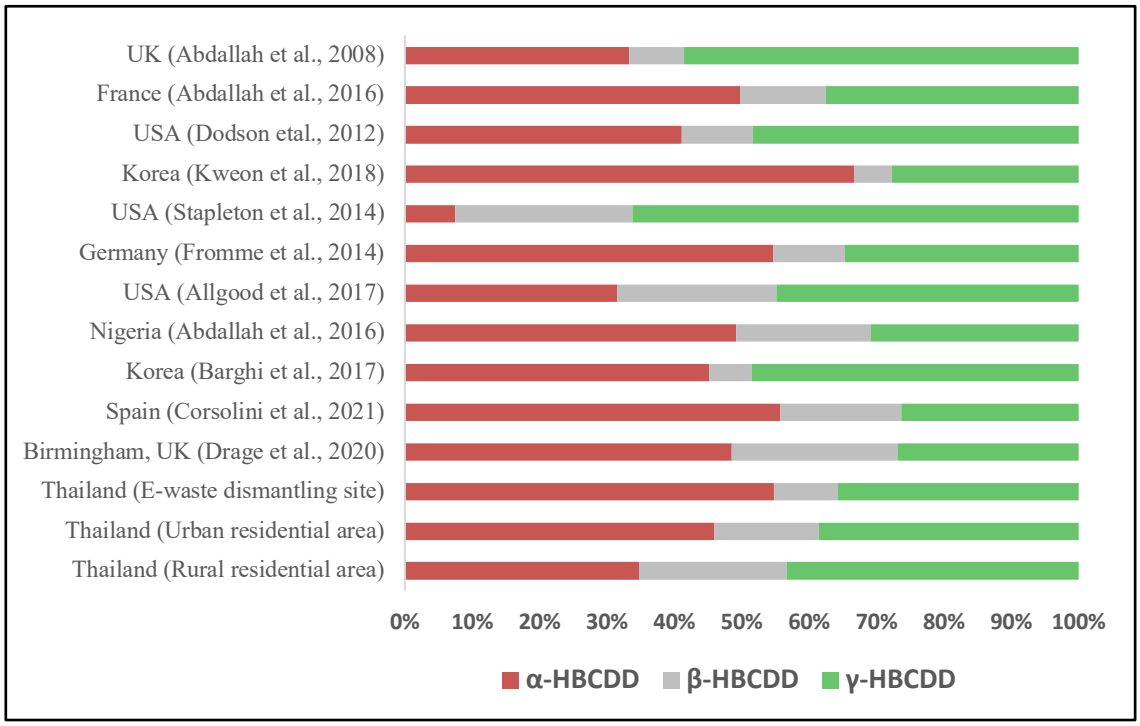

Fig. 3. Relative contributions of different HBCDD isomers to the total HBCDD content in house dust in different studies.

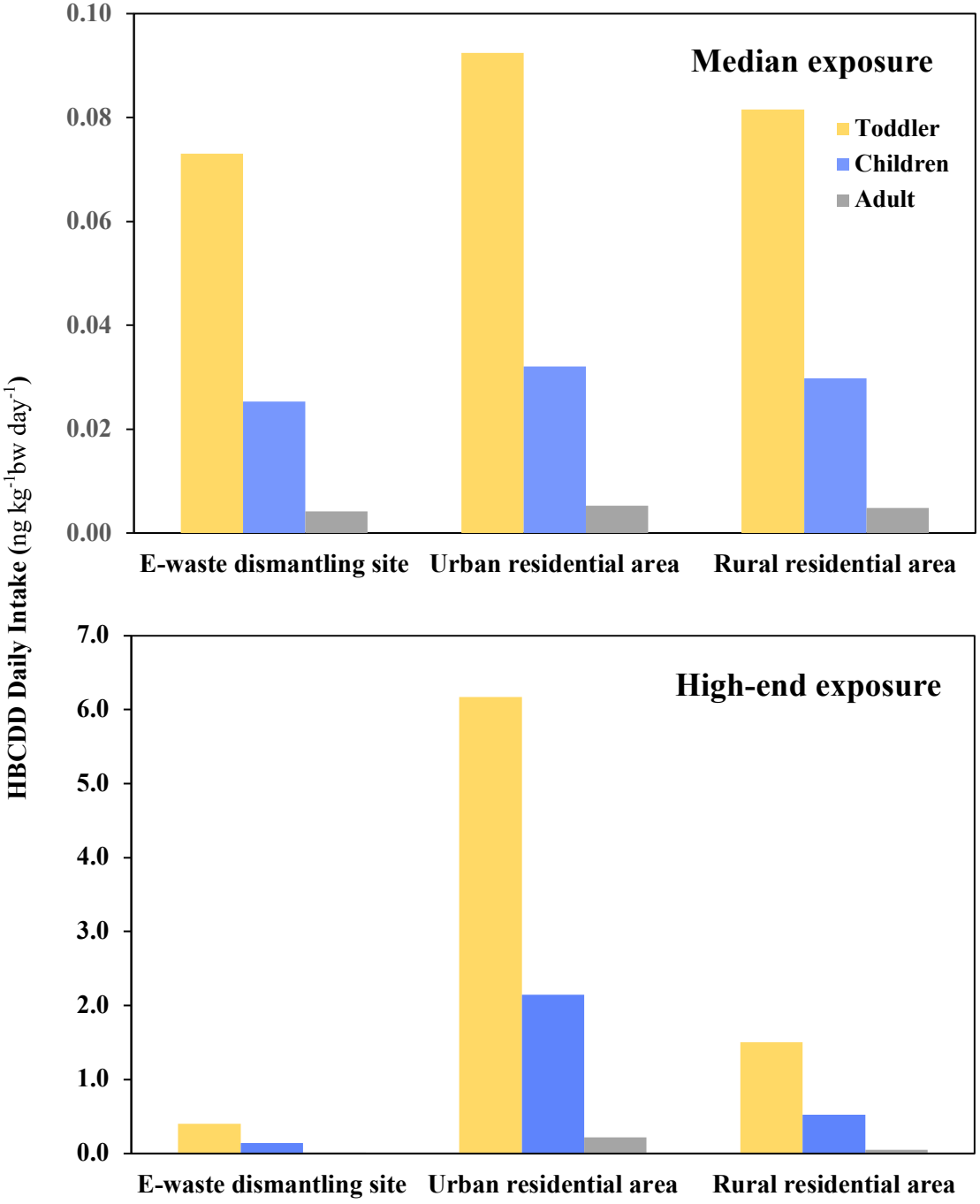

Fig. 4. Estimated daily intake of HBCDD through indoor dust for the Thai population living in an e-waste dismantling site, urban and rural residential areas under the median and high-end exposure scenarios.

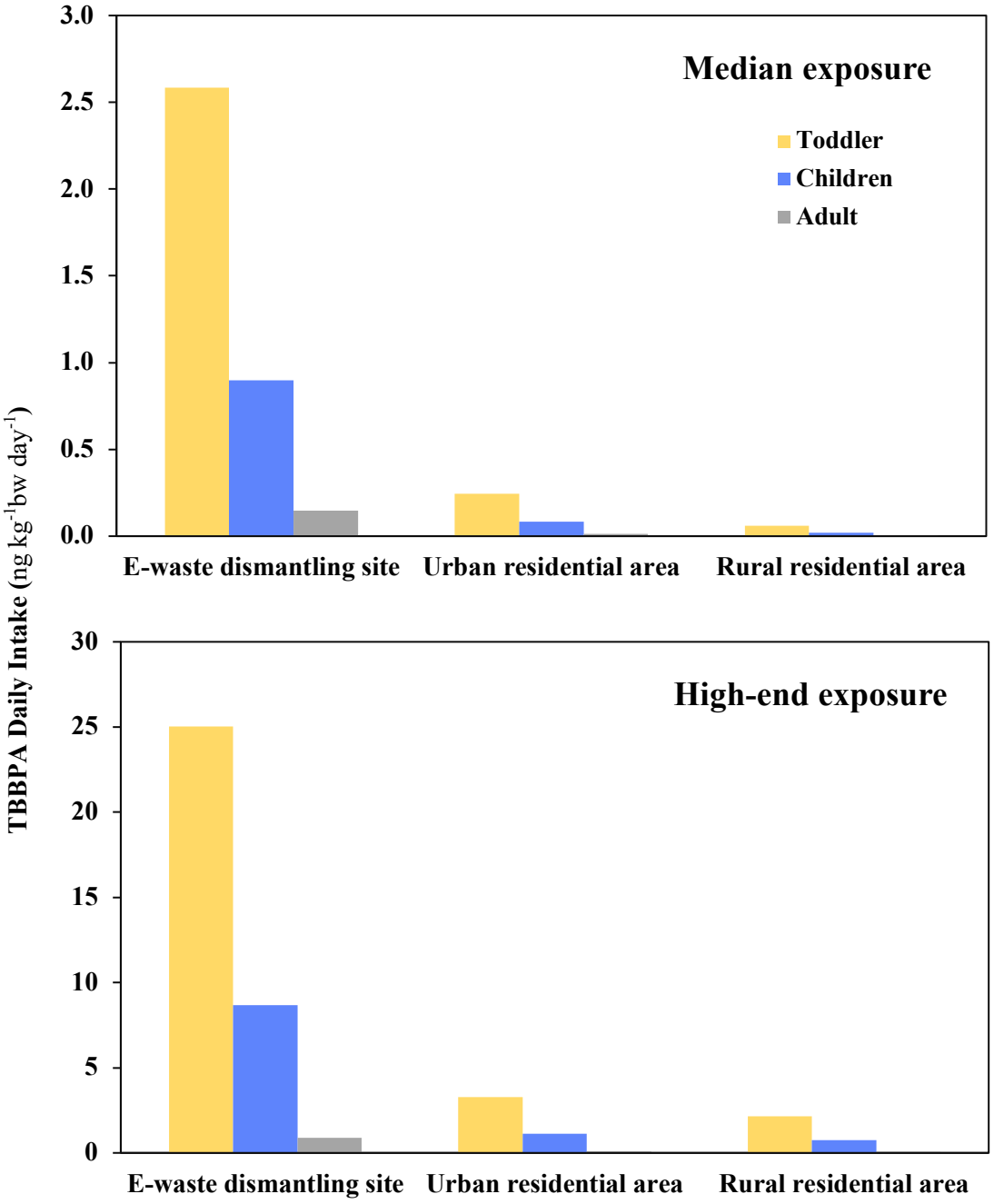

Fig. 5. Estimated daily intake of TBBPA through indoor dust for the Thai population living in an e-waste dismantling site, urban and rural residential areas under the median and high-end exposure scenarios.

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Exposure, Risk and Predictors of Hexabromocyclododecane and Tetrabromobisphenol-A in House Dust from Urban, Rural and E-Waste Dismantling Sites in Thailand

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Table 1 Median concentrations (ng g^{-1}) of HBCDD and TBBPA in house dust in this study and selected countries.

Sampling Year	Country	Sampling site	Sample size (n)	Concentrations (ng g^{-1})					Reference
				α-HBCD	β-HBCD	γ-HBCD	HBCDD (range)	TBBPA (range)	
2006 and 2007	UK	Urban residential area	45	380	93	670	1,300 (140–140,000)	62 (<MQL–382)	Abdallah et al., 2008
2011	USA	Urban residential area	16	62	16	73	160 (39–1,800)	200 (22–2,000)	Dodson et al., 2012
2011	Korea	Urban residential area	42	144	12	60	278 (<LOD–3,132)	69 (<LOD–2,092)	Kweon et al., 2018
2008	France	Urban residential area	9	559	144.00	422	1125 (363–1865)	44 (7–165)	Abdallah et al., 2016
2009	Kazakhstan	Urban residential area	10	78	20.00	189	287 (112–450)	13 (<0.06–83)	Abdallah et al., 2016
2012	USA	Urban residential area	30	7.90	27.80	70	338 (77.6–2,658)	7.9 (<0.20–245)	Stapleton et al., 2014
2012	China	E-waste recycling park	7	-	-	-	-	10,329 (5460–46,191)	Wu et al., 2016
2012	South Africa	Urban residential area	7	-	-	-	-	120 (<LOD –3,767)	Abafe and Martincigh, 2016
2012	Turkey	Urban residential area	3	-	-	-	270 (50-8,800)	-	Kurt-Karakus et al., 2017
2013	Germany	Rural residential area	20	180	35	114	345 (53–4,041)	28 (2.9–233)	Fromme et al., 2014
2013	Egypt	Urban residential area	17	-	-	-	6.15 (1.39–153)	-	Hassan and Shoeib, 2015
2013	USA	Urban residential area	10	52	39	74	326 (104–636)	187 (0–7,260)	Allgood et al., 2017
2014	Nigeria	Urban residential area	10	199	81.00	125	405 (41–1,863)	50 (19–127)	Abdallah et al., 2016
2016	Korea	Urban residential area	46	42.87	5.96	46.18	106.30 (18.92–2,645.49)	78.9 (13.51–1,212.38)	Barghi et al., 2017
2016 and 2017	Spain	Urban residential area	10	74.60	19.60	34.90	129 (12.0–1321)	-	Corsolini et al., 2021
NR[a]	China	Urban residential area	15	-	-	-	0.20 (0.08–1.4)	-	Peng et al., 2017
2019	UK	Urban residential area	14	130	66	72	280 (76–570,000)	35 (<0.5–71)	Drage et al., 2020
2019	Thailand	E-waste dismantling site	13	<LOD	<LOD	<LOD	2.05 (<LOQ–29.8)	720.06 (44.01–2,300.32)	This study
2020 and 2021	Thailand	Urban residential area	23	<LOD	<LOD	<LOD	2.10 (<LOQ–528.4)	68.6 (3.51–300.50)	This study
2019	Thailand	Rural residential area	12	<LOD	<LOD	<LOD	1.11 (<LOQ–126.05)	16.5 (1.96–201.01)	This study

[a] Not reported.

Table 2 Residence characteristics associated with chemical concentrations of TBBPA and HBCDD in house dust samples. Two significant categorical variables at $P < 0.05$ are shown in the table.

	Variable (n)		β (95%CI)	*P*-value
TBBPA ($R^2 = 0.577$)	No. of electronic devices			
	(32)	Small number (<5 items)	Reference	
	(16)	Large number (>5 items)	285.648 ((81.582; 489.714)	0.007
	Distance from e-waste dismantling shop			
	(13)	Close to e-waste dismantling shop	−550.118 (-834.762; -265.474)	0.000
	(35)	Long-distance from an e-waste dismantling shop	Reference	
HBCDD ($R^2 = 0.357$)	Floor Type			
	(44)	Hard floor	121.162 (42.294; 200.030)	0.004
	(4)	Carpet	Reference	

Country	Site	HBCDD (ng (kg-bw)$^{-1}$ day^{-1})						TBBPA (ng (kg-bw)$^{-1}$ day^{-1})						References
		Median exposure scenario			High-end exposure scenario			Median exposure scenario			High-end exposure scenario			
		Toddler	Children	Adult	Toddler	Children	Adult	Toddler	Children	Adult	Toddler	Children	Adult	
UK	Urban residential area	7.24	-	0.52	20.9	-	1.29	0.28	-	0.02	1.13	-	0.05	Abdallah et al., 2008
Germany	Rural residential area	1.73	-	0.15	8.91	-	0.76	0.14	-	0.01	0.53	-	0.05	Fromme et al., 2014
China	E-waste recycling park	-	-	-	-	-	-	-	58.5	7.5	-	-	-	Wu et al., 2016
South Africa	Urban residential area	-	-	-	-	-	-	0.6	0.08	0.08	2.41	0.19	0.2	Abafe and Martincigh, 2016
France	Urban residential area	8.51	-	0.65	61.6	-	2.93	0.47	-	0.03	5.61	-	0.27	Abdallah et al., 2016
Kazakhstan	Urban residential area	0.83	-	0.06	4.88	-	0.23	0.07	-	<0.01	0.83	-	0.04	Abdallah et al., 2016
Nigeria	Urban residential area	2.16	-	0.17	22.4	-	1.07	0.19	-	0.01	1.57	-	0.07	Abdallah et al., 2016
Turkey	Urban residential area	0.95	-	0.02	3.45	-	0.38	-	-	-	-	-	-	Kurt-Karakus et al., 2017
Korea	Urban residential area	4.79	4.02	0.90	7.91	4.58	1.01	0.61	0.48	0.10	1.32	0.61	0.13	Barghi et al., 2017
Spain	Urban residential area	-	-	0.24	-	-	0.61	-	-	-	-	-	-	Corsolini et al., 2021
Thailand	E-waste dismantling site	0.07	0.02	<0.01	0.41	0.14	0.02	2.58	0.90	0.15	25.03	8.69	0.88	This study

Thailand	Urban residential area	0.09	0.03	<0.01	6.17	2.14	0.22	0.24	0.08	0.01	3.26	1.13	0.11	This study
Thailand	Rural residential area	0.08	0.03	<0.01	1.51	0.52	0.05	0.06	0.02	<0.01	2.16	0.75	0.08	This study

Table 3 Estimated daily exposure dose to HBCDD and TBBPA via indoor dust.