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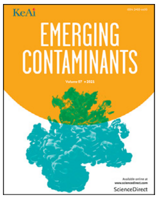

Concentrations of Tetrabromobisphenol-A and hexabromocyclododecane in Thai child daycare centre dust and the exposure risk for young children

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ABSTRACT

Children spend one-third of their day in child daycare centres, where they may be exposed to toxic chemicals, such as tetrabromobisphenol-A (TBBPA) and hexabromocyclododecane (HBCDD), from indoor dust. However, studies on the exposure of TBBPA and HBCDD in schools remain scarce, especially in Thailand, where no such study has yet been reported. Therefore, in this study, we collected dust samples from 20 child daycare centres in Thailand and analysed them for flame retardants, TBBPA and HBCDD. TBBPA was detected in all samples with a median concentration of 35 ng g^{-1} (range, 14–5017.7 ng g^{-1}). The median level of HBCDD was lower than the quantitation limit (LOQ) with <LOQ to 86.6 ng g^{-1} of dust sample. The electronic density score was calculated from the number of electronic appliances divided by each sampled surface floor area that revealed an abundance of TBBPA in the dust in rooms with many electronic items. The use of electronic devices, especially printers, that were found to be related to the high TBBPA concentration in the room, should be minimised in child daycare centres. The children's exposure to these chemicals from dust was estimated for dust ingestion and dermal absorption. The combined exposure was lower than the established reference dose value, and the hazard quotient for children via these two pathways were ranged $0.7 \times 10^{-6} - 2.7 \times 10^{-4}$ for TBBPA and $1.8 \times 10^{-5} - 4.9 \times 10^{-5}$ for HBCDD, which did not exceed 1.0. Although there is uncertainty regarding the potential health effects of long-term exposure to these substances, it is undeniable that child daycare centres represent a source of exposure to these substances for children. Further investigation of other intake routes of several flame retardants in child daycare centres and identification of the sources of these substances is warranted to decrease children's health risks from exposure to these harmful substances in child daycare centres.

1. Introduction

Indoor pollution is an important source of chemical exposure for humans, as most people spend more than 70% of their time indoors [1]. Among indoor pollutants, brominated flame retardants (BFRs) have attracted increasing attention and are a concern in several countries [2]. Tetrabromobisphenol-A (TBBPA) and

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hexabromocyclododecane (HBCDD) are BFRs that are widely used in many materials and consumer products to reduce their flammability [3–5]. HBCDD is primarily added to expanded and extruded polystyrenes, which are generally used as thermal insulation foams for thermal building insulation applications. It also finds application in high-impact polystyrenes, which are used in electronic equipment enclosures and in the back coating of fabrics [3,4,6,7]. TBBPA is primarily applied as a reactive flame retardant to epoxy and polycarbonate resins, however it is also used as an additive flame retardant in acrylonitrile-butadiene-styrene plastics and high-impact polystyrene [8]. It is also widely used in circuit boards, electronic equipment, furniture, and building materials [9]. When applied additively, BFRs do not covalently bind to the original material or polymer. Hence, they can be emitted from products by mechanisms such as abrasion and volatilisation and are widespread in indoor environments [8,10,11]. These pollutants can be associated with dust, pose a risk of human exposure, and adversely affect human health [2], such as through endocrine system disruption, neurotoxicity, and immunological toxicity [9,12]. TBBPA and HBCDD are legacies and emerging contaminants because they are persistent and have the potential for long-range transport in the environment and toxicity, which has caused public health concerns [3,4].

Humans are exposed to indoor chemicals deposited in dust, mainly through dust ingestion and dermal absorption, while they spend time in indoor environments, such as houses and schools [4,13,14]. Children tend to have higher levels of exposure to dust (and therefore the pollutants sorbed to dust) as they are closer to the floor and frequently exhibit hand-to-mouth behaviours and lick objects, which may introduce these pollutants into the body [2,15–17]. Moreover, previous studies suggest that children can be more exposed to these substances in childcare centres than in their homes [17,18]. This is the case because some children spend one-third of each weekday in childcare centres. Therefore, child daycare centres may account for a significant proportion of the exposure of children to toxic chemicals in dust every day [16]. Few studies have focused on TBBPA and HBCDD in schools or in exposure assessments. However, results have shown that childcare centres are contaminated with these chemicals. The median TBBPA and HBCDD concentrations in dust from childcare centres in the UK were 1400 ng g^{-1} and 89,000 ng g^{-1}, respectively [17]. This shows that childcare centres are a significant source of indoor toxic chemicals, and children have a high risk of suffering adverse health effects. In Thailand, our results from previous studies [19,20] indicated that compared to adults, children are more exposed to TBBPA and HBCDD via house dust ingestion. Our findings indicate that children are a sensitive group prone to indoor exposure to these substances, which can adversely affect their health. Therefore, the present issue of children being exposed to toxic substances at childcare centres is of critical worldwide concern. Furthermore, there are no data on the concentrations of TBBPA and HBCDD in child daycare centres in Thailand. This study is the first report of the occurrence of these chemicals in the classroom dust of child daycare centres in this country. The study aimed to quantify the concentrations of TBBPA and HBCDD in dust in the child daycare centres. After investigating the TBBPA and HBCDD levels, the study used the estimated daily intake to assess the exposure of children to these substances through dust ingestion and dermal absorption. The results provide information on children's exposure to TBBPA and HBCDD in child daycare centres in Thailand and recommend guidelines to reduce the exposure of children to such substances.

2. Materials and methods

2.1. Chemicals and reagents

The following standards and internal standards were purchased from Cambridge Isotope Laboratories (Andover, MA, USA): TBBPA; α-, β-, and γ-HBCDD; $^{13}C_{12}$-α-HBCDD; $^{13}C_{12}$-β-HBCDD; $^{13}C_{12}$-γ-HBCDD; and $^{13}C_{12}$-TBBPA (each with purity ≥98%). The d_{18}-γ-HBCDD was supplied by Wellington Laboratories (Guelph, ON, Canada). Concentrated sulfuric acid was acquired from Merck (Darmstadt, Germany). All solvents used in this study, including hexane, dichloromethane, and methanol, were purchased in high-performance liquid chromatography (HPLC) quality from Merck. Silica gel 60 (0.063–0.200 mm) was purchased from Merck. The indoor dust reference material SRM 2585 was supplied by the U.S. National Institute of Standards and Technology (Gaithersburg, MD, USA).

2.2. Sample collection

Detailed sampling, sample extraction, and clean-up procedures were based on previous studies [3,18,22,23]. Briefly, dust samples were obtained from 20 child daycare centres (S1–S20) in Thailand between November 2019 and January 2020. In each child daycare centre, one sample of dust was collected from a classroom where children usually studied, played, and slept. Four square meters of the floor was vacuumed for 4 min. The dust samples were collected using a nylon sock with a 25 μm pore size inserted into the nozzle of a portable vacuum cleaner. The sock was then closed with a twist tie, wrapped with aluminium foil, and sealed in a plastic bag. The dust samples were transported to the laboratory in a cooler with ice. In the laboratory, each sample was passed through a sieve (250 μm) to separate the dust from the coarse particles and refrigerated at −20 °C until further processing. The observation checklist assessed classroom characteristics and the number of electronic devices and furniture in each sampled child daycare centre during the collection time. Electronic devices such as fans, televisions, computers, laptops, printers and air conditioners were counted to determine their number. The electronic density was calculated by dividing the total number of electronic appliances in the sampled classroom by the floor surface area (width × length; m^2) of the classroom. The electronic density score was categorised as high electronic density in the surface room area when its magnitude was greater than 0.2. An electronic density score of 0–0.2 was identified as low electronic density in the surface room area.

2.3. Chemical analysis

The chemical analysis was divided into two processes are sample extraction and clean-up. After weighing a 100 mg portion, the sieved dust sample was spiked with 25 ng each of $^{13}C_{12}$-α-HBCDD, $^{13}C_{12}$-β-HBCDD, $^{13}C_{12}$-γ-HBCDD, and $^{13}C_{12}$-TBBPA, which were used as internal (surrogate) standards. Each sample was extracted with 7 mL of hexane:dichloromethane mixture (1:1, v/v), vortexed for 5 min, sonicated at 20 °C for 30 min, and centrifuged for 5 min at 3500 rpm. After centrifugation, the supernatant was transferred to a clean-up process. The residual sample was extracted in three cycles. The crude extract was then washed with concentrated sulfuric acid to remove interfering organic compounds. After phase separation, the hexane layer was transferred onto a solid-phase extraction cartridge packed with 4 g of pre-cleaned acidified silica (44% concentrated sulfuric acid, w/w), load sample with triplicated 1 mL hexane rinses, and eluted with 25 mL of hexane:dichloromethane mixture (1:1, v/v). The eluate was evaporated to dryness under a gentle nitrogen gas flow. The dried extract was reconstituted in 200 μL of methanol containing 25 pg μL^{-1} d_{18}-γ-HBCDD as a recovery determination (or syringe) standard before liquid chromatography with tandem mass spectrometry analysis to determine the recoveries of the internal standards for quality assurance/quality control purposes.

2.4. Instrumental analysis

Sample analysis was performed using an Agilent 1200SL HPLC system coupled with an Agilent 6400 tandem mass spectrometer. Chromatographic separation of TBBPA and HBCDD isomers (α-, β-, and γ-HBCDD) was performed on an Agilent Pursuit XRS3 C18 reversed-phase analytical column (2 × 150 mm, 3 μm particle size). The mobile phases consisted of (A) 1:1 methanol/water and (B) methanol. The flow rate was set at 0.15 mL/min with an injection volume of 10 μL. The mass spectrometer was operated in electrospray ionisation negative ion mode. The ions were monitored at the transition of m/z 540.8 to 78.8 for native and 552.8 to 78.8 for $^{13}C_{12}$-TBBPA. The ion m/z ranges of 640.4 to 78.8 and 652.5 to 79 were selected for native and $^{13}C_{12}$-HBCDD labelled diastereomers, respectively.

2.5. Quality assurance/quality control

Field blanks ($n = 4$) were collected from every 5th child daycare centre by vacuuming sodium sulfate from pre-cleaned clean floors using the same procedures used to collect dust samples. For each extraction batch of 10 dust samples, one procedural blank was used to check for potential contamination from the process. No analytes of TBBPA or HBCDD were detected in the procedural or field blanks. The certified reference material SRM 2585 ($n = 5$) was used to determine the accuracy and precision of the analytical method. The measured values of SRM-2585 agreed with the certified values (80–123%), with relative standard deviation values below 15%. The limit of quantification (LOQ) value was set as a signal-to-noise ratio of 10, yielding 0.1 ng g^{-1} for TBBPA and 0.7 ng g^{-1} for HBCDD. The recovery values of the internal standards of TBBPA and HBCDD in spiked dust samples were 81–118%, with a mean value of 92 ± 13%, and 93–117%, with a mean value of 105 ± 7%, respectively.

2.6. Calculation of daily intake of TBBPA and HBCDD

Estimated daily intake (EDI, ng kg^{-1} bw day^{-1}) values of TBBPA and HBCDD from child daycare centre dust ingestion and dermal absorption for children (2–4 years) were determined under the median and high-end exposure scenarios. The estimated daily intakes through dust ingestion were calculated using the following equation from previous studies [10,21,22]:

$$EDI_{Ingestion} = (C_{dust} * EF * IR)/BW \tag{1}$$

where C_{dust} is the concentration (ng g^{-1}) of TBBPA or HBCDD in the dust sample, and EF is the estimated fraction of time spent within the child daycare centre each day, which for children was assumed to be 33% of their time (8/24 h) [16]. IR is the daily ingestion rate (g day^{-1}), which assumes 100% absorption of contaminants from dust ingested orally. However, not all fractions are bioavailable, assuming 100%, and the risk assessment may be overestimated. We consider that 100% of our study indicates a worst-case health risk assessment. The ingestion rate for children was assumed to be either 0.05 or 0.2 g day^{-1} [17] to consider both median and high dust ingestion rate scenarios, respectively. BW is body weight (kg), which was taken to be 18.6 kg for children (2–4 years) [23].

The estimated daily intake values of TBBPA and HBCDD from child daycare centre dust via dermal absorption were calculated using the following equation [16]:

$$EDI_{Dermal} = (C_{dust} \times BSA \times DA \times AF \times EF \times 0.001)/BW \tag{2}$$

where BSA is the exposed body surface area (hands, arms, and legs) of the children, which was assumed to be 2564 cm^2/event [18,24], and DA is the amount of dust adhered to the skin, by weight for the exposed body parts (0.04 mg/cm^2) [24,25]. The absorption factor (AF) to the skin was 0.4 for TBBPA and 0.46 for HBCDD [24,25]. EF is the fraction of time spent within the child daycare centre each day and is equal to 33% (8/24 h) [16].

To assess the health risks for children, we calculated the hazard quotient (HQ) according to Equation (3) from previous studies [26]. HQ is the ratio between the total daily intake estimated via dust ingestion and dermal absorption and the oral reference dose (RfD) for each substance.

$$HQ = EDI_{Total} / RfD \tag{3}$$

The HQ values below 1.0 indicate no risk to the children from dust ingestion and dermal absorption. An HQ greater than 1.0 suggests that ingestion and dermal absorption exposure pathways may be an important consideration for adverse health effects in children.

2.7. Statistical analyses

Microsoft Office Excel 2019 was used to analyse the descriptive data. The minimum, maximum, mean, median, and standard deviation of TBBPA and HBCDD in dust were used to calculate the human exposure. All contaminant concentrations below the LOQ were treated as zero for data analysis. Statistical analysis was performed using IBM SPSS statistics software version 21.0. The Shapiro–Wilk test was used to check the normality of the data. After log-transformation, the TBBPA and HBCDD concentrations were normally distributed. A linear regression model was used to determine the relationships between the electronic density score and the target substance concentrations in the child daycare centre dust. A p-value less than 0.05 was considered to indicate statistical significance.

3. Results and discussion

3.1. Concentrations of TBBPA and HBCDD in child daycare centre dust

Table 1 summarises the concentrations of TBBPA and HBCDD in the dust from the 20 child daycare centres in Thailand. TBBPA was detected in all samples analysed (100%, $n = 20$), whereas HBCDD was detected in 35% of the dust samples. The median concentrations of TBBPA and HBCDD in child daycare centre dust were 35 and <0.7 ng g^{-1}, respectively. The concentration of TBBPA in the dust ranged from 13.6 to 5017.7 ng g^{-1} (Fig. 1). Notably, the highest concentration of TBBPA was recorded in child daycare centre S13 (5017.7 ng g^{-1}). In this child daycare centre, the classroom is a large hall divided into three smaller classrooms by boundary partitions. Each small classroom had a television, computer, plastic toys, and foam mattresses. Additionally, two smaller rooms on one side had the doors open all day. One room was used to store several electronic appliances, such as computers, printers, televisions. The

Table 1
Summary of concentrations (ng g^{-1}) of TBBPA and HBCDD in indoor dust from selected countries, reported concentration of this substance as median (min.– max.).

Country	Location	Sampling Year	Sample size (n)	Concentrations (ng g^{-1})					Reference
				TBBPA (range)	α-HBCD	β-HBCD	γ-HBCD	HBCDD (range)	
Thailand	Child daycare centres	2019 and 2020	20	35 (13.6–5017.7)	<0.7	<0.7	<0.7	<0.7 (<0.7–86.6)	This study
Sweden	Preschool	2018	100	0.1 (NR[a]-1.6)	–	–	–	–	[26]
Sweden	Preschool	2015	100	65 (<2.1–3500)	52	15	26	100 (7.6–16,000)	[16]
USA	Childcare centres	NR[a]	14	–	76	14	35	120 (34–3000)	[18]
China	Schools	2014	8	Not detected	–	–	–	94 (84–110)	[13]
Korea	Kindergarten	2016	6	185 (83.5–679.6)	272.1	29.7	110.2	412.1 (184.6–1160)	[4]
UK	Child daycare centres and primary schools	2007 and 2008	43	110 (17–1400)	1400	550	1700	4100 (72–89000)	[17]

[a] Not reported.

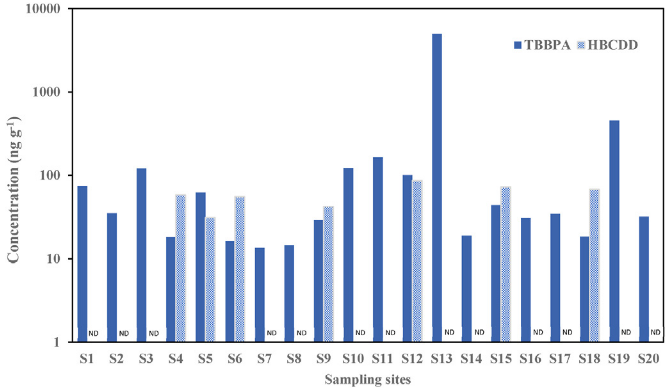

Fig. 1. The concentrations of TBBPA and HBCDD in twenty child daycare centres in Thailand. ND is not detected.

other room was used to store the refrigerators. Furthermore, the air conditioner was turned on throughout the day, and the window was opened for natural ventilation for approximately 20 min when the room was cleaned after school. The air conditioner was turned on while collecting dust samples in the afternoon. TBBPA is used as an additive flame retardant in the high impact polystyrene and acrylonitrile-butadiene-styrene plastic resin in the plastic casings of electrical devices and the components of office electronic equipment [10]. TBBPA can then migrate from the products, leading to widespread pollution in indoor dust [8,9]. In addition, the child daycare centre (S13) had poor ventilation in the classroom. This may be another reason for the accumulation of TBBPA in the classroom dust, resulting in higher TBBPA concentration in this child daycare centre than in other child daycare centres with only a small number of electronic items and natural ventilation flow in the classroom throughout the whole day. This result suggests that higher numbers of electronic items in the classroom and poor ventilation in the child daycare centre lead to higher TBBPA contamination in the child daycare centre dust. We compared our results for TBPPA in child daycare centre dust with those reported in previous studies from various countries worldwide. A recent report [4] found a median concentration of TBBPA in kindergarten room dust in Korea (185 ng g^{-1}) that was higher than that yielded by our results. This might be explained by the fact that although most Thai classroom dust samples were collected in the winter, the room temperature was still high at more than 30 °C. Most classrooms sampled do not have air conditioners, so they use fans and have windows open for natural ventilation most of the day. Unlike in Korea, where classrooms have a closed ventilation system throughout the day due to cold weather Thus, TBBPA may leach from commercial products and become associated with dust, causing TBBPA to be highly contaminated in Korean indoor dust. On the contrary, we found that our median result of TBBPA was 250 times higher than that reported in preschool dust in Sweden (0.1 ng g^{-1}) [26]. Part of the reason may probably be due to the action in the guidance document focusing on reducing children's exposure to toxic substances in Swedish preschools. Reducing the use of electronic devices, toys, and classroom furniture minimises the risk of children from exposure to the hazardous substances contained in these items at preschool [27]. This could explain the lower contamination of TBBPA in the Swedish preschool dust than that reported in our study.

In HBCDD commercial mixture, γ-HBCDD is generally more predominant than α-HBCDD and β-HBCDD. The isomeric distribution of HBCDD in our study presented the highest composition of γ-HBCDD (27–62%), followed by α-HBCDD (19–57%) and β-HBCDD (3–39%). Similar isomeric contribution patterns (γ-, α-, and β-HBCDD) were found in child daycare centre and primary school dust in the UK [17]. However, some previous studies in dust samples from Korea [4], the USA [18], and Sweden [16] showed a predominance of α-HBCDD. It is because γ-HBCDD can shift to α-HBCDD through exposure to natural light, such as in textile products [28]. In addition, thermal treatment (140–160 °C) during the addition of flame retardants to expanded and extruded polystyrene causes transformation from γ-HBCDD to α-HBCDD [29]. Therefore the difference in physical-chemical properties of the isomer, resulting in α-HBCDD in some studies, showed a high contribution pattern than other HBCD isomers. For the sum of HBCDD isomers, the level of HBCDD in seven dust samples from the child daycare centres ranged from <0.7 to 86.6 ng g^{-1} (median <0.7 ng g^{-1}). Dust samples contaminated with HBCDD were found in classrooms with several plastic toys and plastic shelves for books. The highest HBCDD concentration found in the child daycare centres occurred in sample S12 (86.6 ng g^{-1}), followed by samples S15 (72.3 ng g^{-1}) and S18 (67.9 ng g^{-1}). As mentioned, these samples were collected from classrooms that included several plastic toys, some of which were old, and there were also small fabric cushions for the children to sit on. Similarly, a recent study [30] examined contamination in new and second-hand children's toys in the UK and found that some items purchased in the UK before 2017 were contaminated with high HBCDD concentrations ranging from 139,000 to 840,000 ng g^{-1}. Therefore, plastic toys or plastic shelves may be a significant source of HBCDD contamination in the child daycare centre dust collected in this study. Additionally, HBCDD is used as an additive for coating the upholstery fabric for foam furniture [31,32]. This could be another reason for the high concentrations of HBCDD in classrooms of the three child daycare centres. These HBCDD levels may be related to the fabric cushions used for child seating in the classrooms. The median concentrations of HBCDD in our study were much lower than those found in school dust in other countries. For instance, the median concentration of HBCDD in dust from the child daycare centres of this study was more than 4000 times lower than that of UK childcare centres (4100 ng g^{-1}) [17] and >500 times lower than the concentration (412.1 ng g^{-1}) in kindergarten classrooms in Korea [4]. Furthermore, dust samples collected in preschools by previous studies showed median HBCDD concentrations of 120 ng g^{-1} in childcare centres in the USA [18], 100 ng g^{-1} in Sweden [16], and 94 ng g^{-1} in China [13]. This comparison suggests that the HBCDD concentrations in the child daycare centres in our study were substantially lower than those in previous reports, which may be explained by recent information reporting low usage of HBCDD in several applications in Thailand. Moreover, the available data showed that HBCDD compounds have not been imported to Thailand since 2017 [33]. Therefore, the HBCDD contamination in dust from the child daycare centres of our study was very low.

3.2. Association of TBBPA and HBCDD concentrations in child daycare centre dust with electronic density

Details of the types of electronics, number of electronic items, and area of each sampled surface floor are described in Table S1. The electronic density score was determined by dividing by the number of electronic appliances by the area of each sampled surface floor (m^2). The electronic density score was modified from the previous study [34] and categorised into two groups based on half of the total electronic density score: high electronic density with a score greater than 0.2 ($n = 8$) and low electronic density with a score of 0–0.2 ($n = 12$). Rooms with high electronic density scores had a median TBBPA concentration of 124.2 ng g^{-1} (range of 62.2–5017.7 ng g^{-1}) whereas those with low electronic density scores had a median TBBPA level of 24 ng g^{-1} (13.6–43.7 ng g^{-1}). The electronic density scores and concentrations of TBBPA and HBCDD in the dust of each child daycare centre are illustrated in Fig. 2. Linear regression results showed our study has a significant positive relationship between electronic density score and TBBPA concentrations in dust ($p < 0.01$). The higher electronic density score resulted in higher TBBPA concentrations in dust. In contrast, the electronic density score was not related to HBCDD concentration ($p = 0.277$).

Regarding TBBPA related to the electronic density score, the result showed that child daycare centre S13 had the highest TBBPA concentration (5017.7 ng g^{-1}) in dust and the highest electronic density score (0.41). In addition, the classrooms of samples S19, S11, S10, and S3, although the surface floor area was not large, included many electronic items, such as a television, computer, and a printer, which resulted in a high electronic density score that is associated with higher TBBPA concentrations. Interestingly, the classrooms of these samples (S13, S19, S11, S10, and S3) had relatively high TBBPA concentrations in dust. In such classrooms, a printer is used before sampling, and a dust sample was collected immediately after the printer stops operating. During the printing process, fine particles are released into the room. These emitted particles might contaminate with various harmful substances [35]. If TBBPA is used as a flame retardant in the printer, TBBPA was contaminated with the particles released during the printer use. Therefore, printing may be one of the primary sources of TBBPA contamination in classroom dust of child daycare centres. On the contrary, the classrooms of samples (S18 and S20) had a very spacious floor and a small number of electronic devices, resulting in a low electronic density score and low TBBPA concentration. Our results suggest that electronic devices leach TBBPA-contaminated dust into child daycare centres. Thus, electronic devices, especially printers, should not be placed in the classroom. Previous reports [4,10,19] also showed that TBBPA is found at significantly higher concentrations in rooms containing many electronic items than in those with less electronic items. In this study, we only identified a number of electronic items and did not identify other products, such as plastic furniture, plastic toys, and nap mats, in the sample classroom. These results may have provided information relating the concentrations of TBBPA and other flame retardants. Thus, further investigations must consider other sources of flame-retardant substances to identify all sources of toxic substances in the classroom.

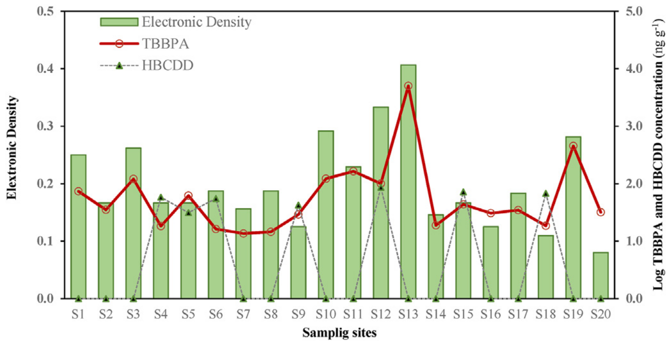

Fig. 2. Electronic density and TBBPA and HBCDD concentration for each child daycare centre in Thailand.

3.3. Daily exposure dose to TBBPA and HBCDD

The concentrations of TBBPA and HBCDD obtained above LOQ (0.7 ng g^{-1}) were used to calculate the daily exposure to these substances. Children's intakes of TBBPA and HBCDD from ingestion and dermal absorption of child daycare centre dust in Thailand were evaluated based on the median and 95th percentile concentrations of TBBPA and HBCDD in dust samples to determine the median and high-end exposure scenarios (Table 2). Briefly, children were exposed to these compounds with slightly different levels of dust ingestion and dermal absorption. For dust ingestion, the estimated daily intake (EDI_{Ingest}) values of TBBPA for children were 0.03 and 2.42 ng kg^{-1} bw day^{-1} for the median and high-end exposure scenarios, respectively. The median exposure scenario of ingestion in this study was half the value reported in a UK daycare centre (0.06 ng kg^{-1} bw day^{-1}) [17]. This difference occurred because the authors of the previous study used the sum TBBPA concentration of daycare centre and office dust to calculate the ingestion exposure, whereas, in this study, we used only the TBBPA in child daycare centre dust to calculate oral exposure. However, the results suggest that UK children do have somewhat higher exposure to TBBPA via dust ingestion. In terms of HBCDD ingestion, the high-end exposure value to HBCDD was 6 times higher than that of the median exposure scenario. A comparison of the EDI_{Ingest} value of HBCDD in dust in our study showed that it was approximately 6 times lower than the value reported in a previous study performed in a childcare centre in the USA (0.29 ng kg^{-1} bw day^{-1}) [18]. This may be because the median concentration of HBCDD in childcare centre dust in the USA was 2 times higher than that in Thai child daycare centre dust. Moreover, they used 10 h for exposure time in a childcare centre in the USA, whereas this study used only 8 h for the exposure time of children in a Thai child daycare centre. Thus, children in the USA were exposed to higher HBCDD levels than those estimated for Thai children. Regarding dermal absorption (EDI_{Dermal}), our results showed that children were exposed to TBBPA and HBCDD via dermal absorption at values of 0.026 and 0.049 ng kg^{-1} bw day^{-1}, respectively. The combined total exposure (EDI_{Total}) values of TBBPA and HBCDD, i.e., the exposure values to TBBPA and HBCDD via both dust ingestion and dermal absorption, were 0.06 and 0.1 ng kg^{-1} bw day^{-1}, respectively, under the median exposure scenario. The total daily intake values were below the oral reference dose (RfD) values for TBBPA

Table 2
The estimated daily intake of TBBPA and HBCDD via dust ingestion and dermal absorption for children in Thai child daycare centres.

Chemicals	EDI_{Ingest} (ng $(kg.bw)^{-1}day^{-1}$)		EDI_{Dermal} (ng $(kg.bw)^{-1}day^{-1}$)		EDI_{Total} (ng $(kg.bw)^{-1}day^{-1}$)		Reference Dose (RfD) (ng $(kg.bw)^{-1}day^{-1}$)	Hazard Quotient (HQ)	
	Median exposure	High-end Exposure	Median exposure	High-end Exposure	Median exposure	High-end Exposure		Median exposure	High-end Exposure
TBBPA	0.031	2.42	0.026	0.50	0.06	2.92	600,000[a] 30,000[b]	9.4×10^{-8} 1.9×10^{-6}	3.1×10^{-7} 9.7×10^{-5}
HBCDD	0.052	0.29	0.049	0.07	0.10	0.36	200,000[c] 3,000[d]	5.0×10^{-7} 3.4×10^{-5}	1.8×10^{-6} 1.2×10^{-4}

[a] Reference dose (RfD) values were estimated by [36].
[b] Reference dose (RfD) values were estimated by [37].
[c] Reference dose (RfD) values were estimated by [38].
[d] Reference dose (RfD) values were estimated by [39].

(600,000 ng kg^{-1} bw day^{-1} [36] and 30,000 ng kg^{-1} bw day^{-1} [37]. For HBCDD, oral reference dose (RfD) values was 200,000 ng kg^{-1} bw day^{-1} [38]. Recently, the UK's Committee on Toxicity of UK suggested that exposure to HBCDD in dust at a high level may lead to adverse health effects and indicated a toxicological reference point of 3000 ng kg^{-1} bw day^{-1} [39]. A previous study reported total exposure values for TBBPA and HBCDD via combined dust ingestion and dermal absorption in Chinese classrooms of <0.01 ng kg^{-1} bw day^{-1} and 0.05 ng kg^{-1} bw day^{-1}, respectively [13]. The total exposure to these compounds in China was 1–2 times lower than that in our study because the concentration of TBBPA in Chinese classrooms was lower than that in Thai classrooms and the HBCDD level was only slightly higher than that in the present study. However, we set the time children spent in their classroom to be 8 h, whereas the study performed in China set the time to 6 h. This difference contributed to the lower exposure values of Chinese children.

The HQ in our study through dust ingestion and dermal absorption was less than 1.0, indicating that children's intake of HBCDD and TBBPA via child daycare centre dust is very low and poses no immediate health risk. However, it is undeniable that child daycare centres are one of the several sources of exposure to these substances. Thus, further investigation must consider other significant exposure pathways, such as inhalation and dermal contact with FR-treated articles for different flame retardants, including those not evaluated in our study. This research should lead to a more thorough understanding of children's total exposure to toxic substances in the classroom. In addition to child daycare centres, children also spend most of their time at home. Therefore, it would be interesting to measure the concentrations of these compounds in both home and child daycare centre and then calculate the children's total exposure in one day. The presence of BFRs in the dust can have long-term health effects on children. The immediate health problems of children exposed to dirty dust are asthma and allergies. Therefore, cleaning the classroom reduces the polluted dust and removes some equipment from the classroom, such as toys and electronic equipment, which are significant sources of dust accumulation in the classroom. This can help reduce acute health effects on children and reduce long-term exposure to BFRs as well.

4. Conclusions

This is the first study to report concentrations of TBBPA and HBCDD in floor dust from child daycare centres (20 centres) in Thailand. Our study showed that the concentration of TBBPA was highest (5017.7 ng g^{-1}) in child daycare centre S13, which included many electronic devices in the classroom. In addition, the electronic density score indicated that high electronic density in the classroom was associated with high TBBPA concentrations. In contrast, HBCDD was detected in comparatively low concentrations because it is rarely used in Thailand and has not been imported since 2017. Although the EDI values of TBBPA and HBCDD via the ingestion route were slightly higher than those for the dermal absorption pathway, the total exposure via the two pathways was well below relevant reference dose (RfD) values. Our information provided an HQ value substantially lower than 1.0, indicating that children do not have any immediate health risks from exposure via dust ingestion and dermal absorption. However, since some children spend much of their daytime hours in a child daycare centre, they can be exposed constantly to toxic chemicals every day in the child daycare centre, which may affect their health over long periods of time. Thus, information about exposure to various hazardous substances in child daycare centres remains an important goal for minimising exposure to potential chemicals and protecting the health of children. Future studies should assess other intake routes of various flame retardants in child daycare centres and develop guidelines to reduce flame retardant intake via several exposure pathways, which are required to lower the health risks to children in the child daycare centre setting.

Conflict of interests declaration

The authors submit this manuscript titled “Concentrations of Tetrabromobisphenol-A and Hexabromocyclododecane in Thai Child Daycare Centre Dust and the Exposure Risk for Young Children” to be considered for publication as a research paper in *Emerging Contaminants*.

I and all co-authors have confirmed that there are no known conflicts of interest.

Ethics approval

The authors declare that this study does not involve animals.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.emcon.2023.100229.

Abbreviations

BFRs	brominated flame retardants
HBCDD	hexabromocyclododecane
TBBPA	tetrabromobisphenol-A
°C	degrees Celsius
EDI	estimated daily intake

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