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Concentrations and isomer profiles of hexabromocyclododecanes (HBCDDs) in floor, elevated surface, and outdoor dust samples from Basrah, Iraq†

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Concentrations of the α, β, and γ- diastereomers of hexabromocyclododecane (α-, β-, and γ-HBCDD) were measured in 60 dust samples from 20 homes across Basrah, Iraq. From each home, two indoor dust (ID) samples (specifically one collected from elevated surfaces (ESD) and one from the floor (FD)) were collected from the living room, with one outdoor dust (OD) sample collected from the front yard of the house. Concentrations of HBCDDs decreased in the following sequence ESD > FD > OD. For ID, ΣHBCDD concentrations varied from 5.3 ng g^{-1} in FD to 150 ng g^{-1} in ESD, with median levels of 60 and 40 ng g^{-1} in ESD and FD respectively. Concentrations of γ-HBCDD, and consequently of ΣHBCDDs in ESD, significantly ($p < 0.05$) exceeded those in FD. For adults, this implies that exposure assessments based on FD only may underestimate exposure, as adults are more likely to ingest ESD. Concentrations of ΣHBCDDs in OD ranged between 7.4 and 120 ng g^{-1} with a median of 35 ng g^{-1} and were significantly exceeded ($p < 0.05$) by those in ID samples. Concentrations of ΣHBCDDs in OD from houses with car parking areas exceeded ($p < 0.05$) those in OD from other homes, implying vehicles as potential emission sources of HBCDDs. Simultaneously, there was moderate correlation ($R = 0.510$–0.609, $p < 0.05$) between concentrations in ID and OD, implying that the indoor environment is an important source of OD contamination. The isomer pattern of HBCDDs in dust samples displayed a predominance of α-HBCDD, which represented 56%, 52% and 59% ΣHBCDD in ESD, FD and OD samples respectively. Derived from the concentrations reported in this study, the median and 95th percentile estimated daily intakes (EDI) for Iraqi adults and toddlers through house dust ingestion did not exceed the reference dose (RfD) value for HBCDD.

Environmental significance

Because of its persistent and bioaccumulative properties, hexabromocyclododecane (HBCDD) has been listed in the Stockholm convention since 2013, including specific exemptions for the production and use in EPS and XPS foams. Ingestion of indoor dust constitutes an important pathway of exposure to HBCDDs. This study reveals that concentrations of HBCDD in dust collected from elevated surfaces were significantly higher than those in floor dust from the same rooms. This implies considerable uncertainty in human exposure assessment, particularly for adults, as they are likely more exposed to elevated surface dust. Our data represent a valuable baseline against which responses to actions designed to limit exposure to HBCDD may be evaluated in the future.

1. Introduction

Hexabromocyclododecanes (HBCDDs) are cyclic aliphatic additive brominated flame retardants (BFRs). The main application of HBCDDs is in extruded (XPS) and expanded (EPS) polystyrene foams as thermal insulation in buildings, with another important application being in textiles used in upholstered furniture, curtains, wall coverings, and transportation seating. Other applications of HBCDDs include adhesives and paints, as well as in high-impact polystyrene (HIPS) and styrene–acrylonitrile resins employed in casings for electrical and electronic equipment.[1–3] Due to bans and restrictions on another widely-used class of BFRs – polybrominated diphenyl ethers (PBDEs), HBCDDs were increasingly used as a flame retardant a substitute for penta- and octa -BDEs.[4–7] The commercial mixtures of HBCDDs consist of three main stereoisomers that include γ-HBCDD (75–89%), α-HBCDD (10–13%) and β-HBCDD (1–12%). In addition, δ- and ε-HBCDD are present at minor concentrations in the commercial mixture.[1,8,9] The HBCDD mixture is characterized by low vapour

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pressure (6×10^{-5} Pa at 21 °C), water solubility (3.4 μg L^{-1} at 20 °C), and high affinity to particulate matter and environmental lipids as exemplified by its log K_{OW} of 4.78.[9,10]

Since the late 1960s, HBCDD has been on the world market, produced mainly in China, the United States of America, and the European Union. The global production of HBCDDs increased rapidly from 16 700 tonnes in 2001 to 22 000 tonnes in 2006, and 31 000 tonnes in 2011.[9,11,12] Because of its persistent and bio-accumulative properties, HBCDD has been listed in Annex A of the Stockholm convention since May 2013. However, the listing includes specific exemptions for the production and use of HBCDDs in EPS and XPS foams in building materials[13], where use is still allowed until 2025.[14]

As additive flame retardants and semi-volatile organic compounds (SVOCs), HBCDDs are not chemically bound to the polymeric products and emissions are likely during production, use, disposal and recycling processes.[1,15] It has been reported that HBCDDs can leach from textiles even at room temperature.[16] As a result, they are globally distributed in various environmental samples such as indoor and outdoor air,[17–19] surface water,[5,20,21] wastewater and sewage sludge,[22,23] sediments,[24,26] soil and road dust,[6,27,28] indoor dust,[19,29,30] food,[31–33] and biota.[34,35] In addition, several biomonitoring studies have detected HBCDDs in human tissues such as human milk,[36–38] serum,[39,40] and hair,[41,42] indicating human exposure to HBCDDs. The main routes *via* which such exposure occurs are: inhalation,[43] diet,[31,44] dust ingestion,[45–47] and dermal absorption.[48,49] Based on animal test results, exposure to HBCDDs are reported to cause different toxicological effects and present potential human health concerns. The main targets for HBCDDs toxicity are: the liver, thyroid hormones, the immune and reproductive systems, as well as neurodevelopmental concerns.[14,15,50–52]

Indoor dust intake constitutes an important pathway of exposure to HBCDDs, particularly for young children due to the large time fraction spent indoors, small body mass and extensive hand-to-mouth behaviour.[53–56] For example, in worst case scenarios, dust ingestion represents 95% of toddler intake for HBCDDs.[57] The importance of this pathway is emphasised by the significant correlation between exposure to HBCDDs *via* dust ingestion and its concentration in serum.[58] HBCDDs migrate from products into indoor dust *via* volatilisation, abrasion, and direct contact between dust and the surface of products.[16,59,60] A large number of studies have reported various levels of HBCDD in indoor dust from different microenvironments such as homes,[61–64] offices,[65,66] schools[67,68] and cars.[69,70]

Most studies of exposure *via* indoor dust measure HBCDDs based on floor dust only, however, for other SVOCs, significant differences between concentrations in elevated surface dust (*i.e.* dust from surfaces above floor level, such as shelves, tables, sofas *etc.*) and floor dust from the same microenvironment have been reported.[71–78] In general, concentrations of PBDEs, NBFRs ("novel" brominated flame retardants) and OPEs (organophosphate esters) in ESD are significantly higher than those in FD.[79] This implies considerable uncertainty in human exposure assessment, particularly for adults, as they are likely more exposed to ESD due to greater contact with elevated surfaces compared to floors.[75] Nevertheless, no available data have compared HBCDD concentrations between ESD and FD specifically from homes. To our knowledge, only one study has examined HBCDDs in both ESD and FD collected from various US microenvironments (a bus, scientific laboratory, computer laboratory, gymnasium, and two each of domestic apartments, classrooms, and offices). However, despite the small sample size ($n = 20$ in total comprising paired ESD and FD samples from 10 microenvironments), this study revealed median concentrations of α-HBCDD and γ-HBCDD in ESD exceeded those in FD by factors of 4 and 1.6 respectively.[80] In addition, many studies focusing on human exposure to flame retardants *via* dust ingestion have indicated that exposure to outdoor dust, such as road dust, is not negligible.[81,82] In other words, it can be concluded that both indoor and outdoor dust will be valuable for human risk assessments. Considerable levels of HBCDDs were found in road and street dust from different areas, such as agricultural, residential, industrial[27] and e-waste recycling areas,[28] suggesting that indoor environments and industrial activities are important sources of HBCDDs to outdoor environments. Nonetheless, to the best of our knowledge, so far, no investigation has attempted to measure concentrations of HBCDDs in paired indoor/outdoor dust samples from the same microenvironment to assess to what extent indoor environments may contribute to outdoor contamination. From our previous study of the most widespread OPEs, significant correlations were found in their concentrations between indoor and outdoor locations from the same house.[83] Furthermore, despite a large number of studies that measured human exposure to HBCDDs *via* indoor dust ingestion, only a limited number have been performed in the Middle East.[84,85] This study is the first to report HBCDD concentrations in dust samples from Iraq. Basrah (or Basra) province was selected for dust sampling because it is one of the important cities in the Middle East, a major port, a main economic city, particularly in the oil industry and the second-largest city by population in Iraq.

Given this background, the aims of this work are to: (1) determine the concentrations of three HBCDD diastereomers (α-HBCDD, β-HBCDD, and γ-HBCDD) in dust samples from Basrah, Iraq to provide an indication of the use of these chemicals in Iraq; (2) examine within-room spatial variability of HBCDDs in dust samples taken from elevated surfaces and floor dust from the same microenvironment, to facilitate assessment of the impact of any such variation on human exposure assessment; (3) determine the relationship between concentrations of HBCDDs in house dust and dust from outdoor (house entrance) surfaces adjoining the sampled houses, to assess to what extent that HBCDD outdoor contamination is influenced by indoors; and (4) provide the first evaluation of the exposure of the Iraqi population to HBCDDs *via* ingestion of indoor and outdoor settled dust.

2. Materials and methods

2.1. Sampling

Details of how samples of indoor and outdoor dust were collected during September–November of 2019 are given in our previous study.[83] In brief, from 20 urban houses in Basrah, Iraq, 60 dust samples were collected using a vacuum cleaner with a nylon sock (25 μm pore size) inserted into the nozzle of the vacuum. From each house, three different types of dust were collected: two indoor dust (ID) samples comprising one floor dust (FD) and one elevated surface dust (ESD) sample, along with one outdoor dust (OD) sample. FD samples were collected from the most frequently used floor areas in the main living room, while ESD was collected from available elevated surfaces such as tables, shelves, and chairs in the same living room for 2–4 min.[75,86] OD samples were vacuumed from available areas next to the wall and surfaces above the ground in the main house entrance area which represents the front yard of the house located between the house and the outer boundary of the property. After sampling, socks were closed, sealed in a plastic bag and stored at −20 °C. Characteristics of the participating houses such as floor material, furniture, electronic equipment, ventilation system and the presence of car parking area contiguous with the entrance of the house were recorded in the ESI (Table S1†).

2.2. Sample preparation, extraction, and clean-up

Dust samples were analysed at the University of Birmingham, UK. The sample extraction and clean-up methods were described in detail elsewhere with slight modification.[87] Briefly, dust samples were passed through a stainless steel 125 μm mesh size sieve, weighed accurately (50–75 mg) and spiked with 50 ng of ^{13}C-α-HBCDD, ^{13}C-β-HBCDD and ^{13}C-γ-HBCDD as internal standards. Two mL of extraction solvent (*n*-hexane: acetone, 3 : 1, v/v ratio) were added to each sample, vortexed for 2 min, sonicated for 5 min, centrifuged for 5 min at 3500 rev per min and the supernatant transferred to a separate tube. The extraction process was repeated three times with all extracts combined, evaporated to incipient dryness, and resolubilised in 1 mL of hexane. The concentrated extract was fractionated into two fractions (F1 and F2) using Florisil cartridges. F1 (containing some HBCDDs) was eluted with 8 mL *n*-hexane, with F2 (containing the remainder of the HBCDDs, as well as OPEs (data reported elsewhere[83])) eluted with 10 mL ethyl acetate. F1 was evaporated to 1 mL and transferred onto 2 g acidified silica 44% cartridges, eluted with 10 mL of Hex/DCM (1 : 1, v/v), evaporated until dryness and then reconstituted in *n*-hexane. Following analysis of F2 for OPEs,[83] F1 and F2 were combined and evaporated to incipient dryness before reconstitution in 100 μL methanol containing 50 ng d_{18}-γ-HBCDD as recovery determination standard.

2.3. Instrumental analysis

α-, β- and γ- HBCDDs were separated and analysed using LC-MS/MS. The equipment used was a dual pump Shimadzu LC-20AB Prominence liquid chromatograph (Shimadzu, Kyoto, Japan) equipped with a SIL-20A autosampler, a DGU-20A3 vacuum degasser, and a Varian Pursuit XRS3 C18 reversed phase analytical column (150 mm × 4.6 mm i.d., 3 μm particle size). A mobile phase program based upon (mobile phase A) 1 : 1 methanol/water and (mobile phase B) methanol at a flow rate of 0.18 mL min^{-1}. The elution program was started at 50% (B) then increased linearly to 100% (B) over 4 min, held for 9 min followed by a linear decrease to 88% (B) over 10 min, held for 0.1 min and ending with 50% (A) for 14 min. The three HBCDD stereoisomers were baseline separated with retention times of 10.8, 11.4 and 11.9 min for α-, β- and γ-HBCDD, respectively.

Mass spectrometric analysis was performed using a Sciex API 2000 triple quadrupole mass spectrometer (Applied Biosystems, Foster City, CA) equipped with an ESI ion source operated in negative ionisation mode. MS/MS detection operated in the multiple reaction monitoring (MRM) mode was used for the quantitative determination of the target compounds based on m/z 640.6 → 78.9, m/z 652.4 → 79.0 and m/z 657.6 → 78.9 for the native, ^{13}C-labelled and d_{18}-labelled HBCDD diastereomers, respectively.

2.4. Quality assurance/quality control

All the test tubes were ultrasonicated and Pasteur Pipettes rinsed with *n*-hexane and baked at 450 °C overnight before use. To assess any possible contamination during sample preparation and analysis, one laboratory blank was conducted in with every set of 6 dust samples. In total 12 blanks were analysed. All target HBCDD isomers in the blanks were below the limit of detection (LOD), and hence data are not corrected for blank concentrations. Similar results were obtained for field blanks ($n = 3$). LOD was estimated based on a signal to noise ratio 3 : 1, with limits of quantification (LOQ) estimated based on signal to noise ratio of 10 : 1, (Table S2,† ESI). For on-going evaluation of the accuracy and precision of the analytical method, an aliquot of SRM-2585 organic contaminants in house dust was analysed with each batch of 10 dust samples. The results show good agreement with previously reported indicative concentrations of HBCDDs (Keller *et al.*, 2007)[88] (Table S3†). Recoveries (average ± standard deviation) of the ^{13}C-labelled internal standards added to dust samples were: α-HBCDD = 76 ± 14%, β-HBCDD = 83 ± 12%, and γ-HBCDD = 86 ± 15%.

2.5. Statistical analysis

Statistical analysis of the data was conducted using Excel (Microsoft Office 2019) and IBM SPSS statistics software (V. 20). Due to the skewness of the data, all concentration data were first log-transformed to achieve normal distributions. For the purposes of statistical evaluation, all concentrations below LOQ were assigned a value of 0.5 LOQ. ΣHBCDD indicates summation of α-, β- and γ-HBCDD. Potential correlations between various parameters were investigated using Pearson's correlation. Results were considered significant where P values were less than 0.05.

3. Results and discussion

3.1. Levels and distribution profiles of HBCDDs in dust samples

The three HBCDD stereoisomers were detected in at least 70–80% of samples for α- and β- HBCDDs, while γ-HBCDD was detected in all indoor and outdoor dust samples. The detection frequency, mean, median, minimum, maximum, 5th percentile and 95th percentile concentrations of α-, β-, γ-HBCDD and ΣHBCDDs in ESD, FD, and OD samples are shown in Table 1. The concentrations of the three stereoisomers and ΣHBCDDs in each analysed sample can be found in Tables S4–S6.† Overall, the highest concentrations of ΣHBCDDs were observed in ESD samples ranging from 17 to 150 ng g^{-1} with an average of 65 ng g^{-1} followed by FD and OD with averages of 46 and 38 ng g^{-1}, respectively. Compared with our previous studies of indoor dust in Basrah, Iraq,[75,83] concentrations of HBCDDs were substantially lower than those of PBDEs and OPEs, indicating the limited application of HBCDD in Iraqi homes. Based on the recorded information, only three of the participating houses were insulated using polystyrene foam boards as thermal insulation. Thus, it is likely that the main sources of HBCDDs in Iraqi houses are textiles, as well as electrical and electronic equipment.

The concentrations of HBCDDs in house dust samples detected in this study were comparable with those in indoor dust from Vietnamese houses (median ΣHBCDDs = 29 ng g^{-1}),[89] but higher than those in indoor dust from Egypt (median 6.2 ng g^{-1})[84] and residential homes surrounding e-waste recycling workshops from China (median = 19 ng g^{-1}).[28] By comparison with other regions, median concentrations of ΣHBCDDs in Iraqi houses were lower than those reported from Europe and North America, and substantially lower than in house dust from France,[70] Canada,[53] and Ireland[19] by factors of 34, 16, and 12 respectively (Table 2 and Fig. S1†). Most recently, the highest median concentration of ΣHBCDDs reported in indoor dust (4 homes, 2 offices and 3 schools) was 18 400 ng g^{-1} in Spain,[68] while the highest dust concentration recorded globally was 570 000 ng g^{-1} from a house in Birmingham, UK.[64]

The isomer pattern of HBCDDs in the two indoor (ID) dust (ESD and FD) and OD samples collected from each house, displayed a predominance of α-HBCDD. On average, samples consisted of 56%, 52% and 59% α-HBCDD in ESD, FD and OD samples respectively (Fig. 1). Similar composition profiles were also found in indoor dust collected in the UK,[64] France,[70] Germany,[90] the USA,[62] China[28] and Korea,[63] where α-HBCDD represented 46%, 44%, 55%, 63%, 68%, and 52% of ΣHBCDDs respectively. A higher proportion of α-HBCDD (63–67%) in OD was observed in China.[27,28,91] The proportion of γ-HBCDD in our dust samples was lower than in commercial HBCDD formulations (which are reported to consist of 75–89% γ-HBCDD).[1,5,11] It has been reported that during the production or processing of materials containing HBCDDs, thermal rearrangement at elevated temperatures (>160 °C), will result in a substantially altered profile of HBCDDs (78%, 13%, and 9% for α-, β-, and γ-HBCDD respectively).[1,92] In addition, experimental data on light-exposed dust samples show a significant shift from γ-HBCDD to α-HBCDD.[29] This may explain the slightly higher ratio of α-HBCDDs in outdoor samples of the current study (59%), compared to that in ID (52–56%).

Table 1 Statistical summary of concentrations (ng g^{-1}) of HBCDDs in elevated surface dust (ESD), floor dust (FD) and outdoor dust (OD) from Iraqi homes

	α-HBCDD	β-HBCDD	γ-HBCDD	ΣHBCDDs
Elevated surface dust (ESD)				
Average	36	6.6	22	65
Median	35	7.0	20	60
Min	0.66	0.66	9.2	17
Max	100	17	39	150
SD	26	4.9	10	35
DF%	85	70	100	100
5%	0.66	0.66	10	22
95%	77	13	38	120
Floor dust (FD)				
Average	24	6.3	16	46
Median	23	6.3	12	40
Min	0.66	0.66	4.1	5.3
Max	64	17	41	120
SD	18	4.5	11	32
DF%	80	80	100	100
5%	0.66	0.66	4.5	9.2
95%	55	12	40	100
Outdoor dust (OD)				
Average	22	4.8	11	38
Median	19	4.4	8.4	35
Min	0.66	0.66	4.2	7.4
Max	75	13	32	120
SD	20	3.9	6.9	28
DF%	80	70	100	100
5%	0.66	0.66	4.3	10
95%	60	11	22	86

3.2. Within-room spatial variability of HBCDDs in dust samples

Statistically significant differences were observed between concentrations of HBCDDs in ESD and FD. A paired *t*-test revealed that concentrations of γ-HBCDD, and consequently ΣHBCDDs in ESD exceeded those in FD with *p* values of 0.001, and 0.002 respectively. In addition, median ESD : FD ratios of HBCDD stereoisomers were found to be 1.5, 1.1, 1.6, and 1.5 for α-HBCDD, β-HBCDD, γ-HBCDD, and ΣHBCDDs respectively. Fig. 2A illustrates within-room spatial variability in HBCDDs between ESD and FD. This is consistent with HBCDD concentrations from various microenvironments within a university campus in the USA, where the median ESD : FD ratios exceeded 1.2.[80] However, no significant differences were apparent between ESD and FD levels in that study, this is likely due to the small sample size that was collected from 10 different microenvironments. For other flame retardants such as PBDEs, NBFRs, and OPEs, significant differences in concentrations of such chemicals were apparent between ESD and FD in Japan,[72] Norway,[71] Iraq,[75,83] UK,[76] and China.[93] This implies that HBCDDs are likely to have similar distribution patterns between ESD and FD from the same microenvironment. As SVOCs and additive flame retardants, HBCDDs can be released from treated products into the environment *via* volatilisation and subsequent sorption to dust particles depending on their vapour pressures and octanol–air partition coefficient (K_{OA}).[94] However, for low volatility compounds like HBCDDs, abrasion of fibres or

Table 2 Comparison of median concentrations (ng g^{-1}) of HBCDDs in this study with previous reports

Country	Sampling year	*n*	α-HBCDD	β-HBCDD	γ-HBCDD	ΣHBCDDs
Iraq, this study	2019	20	23	6.3	12	41
Turkey[85]	2012	10	n.a.	n.a.	n.a.	250
Egypt[84]	2013	17	n.a.	n.a.	n.a.	6.0
Kazakhstan[70]	2009	10	78	20	190	280
Japan[46]	2009–2010	10	n.a.	n.a.	n.a.	70
Korea[63]	2011	42	140	12	60	280
Vietnam[89]	2008	33	10	3.0	9.0	29
China[28]	2016–2017	25	13	2.0	4.0	19
China[25]	2014	30	64	21	64	150
Nigeria[70]	2014	10	99	81	130	400
UK[64]	2019	14	130	66	72	280
France[70]	2008	9.0	600	140	420	1400
Sweden[103]	2010	27	56	18	37	330
New Zealand[61]	2009–2010	50	99	12	96	210
Norway[105]	2014	61	94	23	43	190
Germany[90]	NR	20	180	35	110	330
Ireland[19]	2016–2017	32	200	100	200	490
Portugal[104]	2010–2011	28	91	16	35	150
Czech Republic[69]	2008	25	26	7.0	61	94
Romania[106]	2010	47	200	40	30	270
Belgium[58]	2007–2008	16	69	14	31	110
USA[52]	2012	30	210[a]	28[a]	70[a]	340[a]
Canada[53]	2006	8.0	300	72	230	640
Antarctica[107]	2006	11	150	35	43	230

[a] GM, n.a. = not available.

particles from treated products and transfer *via* direct source-dust contact were identified as the dominant mechanisms of emission into the indoor environment.[59,95] This may explain the higher concentrations of HBCDDs in ESD since most such samples were collected directly from sofas, tables, and shelves on which electronic devices were present. Pearson correlation analysis revealed concentrations of both α-HBCDD and γ-HBCDD in our ESD samples were significantly ($p < 0.05$) correlated with those in FD, with respective correlation coefficient values of 0.652, 0.682, and 0.718 for α-, γ-, and ΣHBCDDs, respectively. This implies a similar source of HBCDD to both ESD and FD samples.

3.3. Within-home (between indoor and outdoor) spatial variability of HBCDDs in dust samples

A one-way repeated measures ANOVA test was performed to evaluate to what extent HBCDD concentrations in both ESD and FD exceeded those in OD samples. Our results revealed that γ-HBCDD and subsequently ΣHBCDDs in ESD were significantly higher than those in OD with *p* values of <0.001 and 0.02 respectively. A paired *t*-test also revealed that concentrations of β-HBCDD, γ-HBCDD, and ΣHBCDDs in ID (*i.e.* the average concentration in each house of ESD and FD) were significantly higher than those in OD samples with a *p* value of 0.008, <0.001, and 0.006 respectively. Furthermore, the median ratio of ID : OD concentrations for the three stereoisomers and ΣHBCDDs ranged between 1.1 for β-HBCDD and 1.6 for γ-HBCDD. Concentrations of our target HBCDDs in ID were moderately correlated with those in OD, with correlation coefficient values of 0.609, 0.551, and 0.509 for α-, β-HBCDD and ΣHBCDDs, respectively. This indicates that indoor sources may contribute significantly to concentrations of HBCDDs outdoors *via* windows, doors, mechanical ventilation systems, and shoes. It has been reported that HBCDD concentrations in soil decline

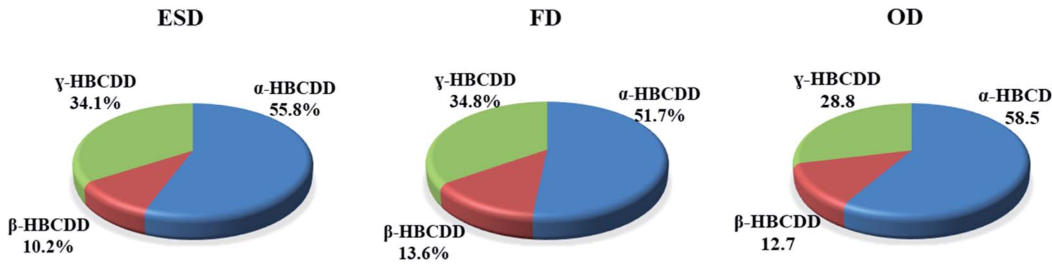

Fig. 1 Average contribution profiles of α-, β- and γ-HBCDD to HBCDDs in elevated surface dust (ESD), floor dust (FD) and outdoor dust (OD) samples from Iraqi homes.

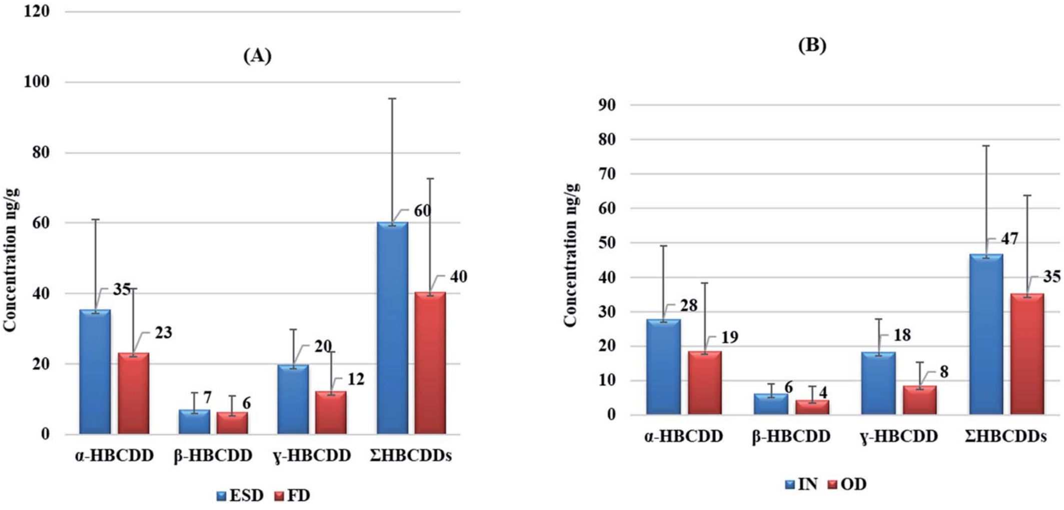

Fig. 2 Comparison of median concentrations (ng g^{-1}) of HBCDDs; (A) between Elevated Surface Dust (ESD), Floor Dust (FD), and (B) between Indoor (IN; the average concentration of ESD and FD) and Outdoor Dust (OD) with standard deviation (*y* error bar).

on moving away from housing which was attributed to the decreasing influence of emissions from HBCDD-treated building insulation.[96] Fig. 2B demonstrates the observed within-home spatial variability between ID and OD and Fig. 3 illustrates correlations between concentrations of HBCDDs in IN and OD dust samples.

It is worth noting that the outdoor locations sampled in this study are not on roads or streets, but the front yards of the investigated homes which are commonly used for parking vehicles. A paired *t*-test revealed that concentrations of ΣHBCDDs in OD from houses with front yards used as parking areas exceeded those in OD from other homes with a *p* value of 0.014 (Fig. S2†). This may indicate vehicles as potential emission sources of HBCDDs in parking areas. Several studies have reported high concentrations of HBCDDs in car dust samples[45,70,97,98] with a higher relative abundance of γ-HBCDD. Further research is needed to investigate the influence of vehicles parked in front yards on the concentrations of HBCDDs and related chemicals in OD and ID environments.

3.4. Estimation of HBCDDs daily intake *via* dust ingestion

Measured concentrations of HBCDD in house dust were used to estimate exposure for the Iraqi population *via* oral ingestion. Two scenarios were considered, based on ingestion of median and 95th percentile concentrations at mean and high rates of 20 and 60 mg day^{-1} for adults, and 50 and 100 mg day^{-1} for toddlers, respectively.[99] Estimated daily intakes *via* house dust

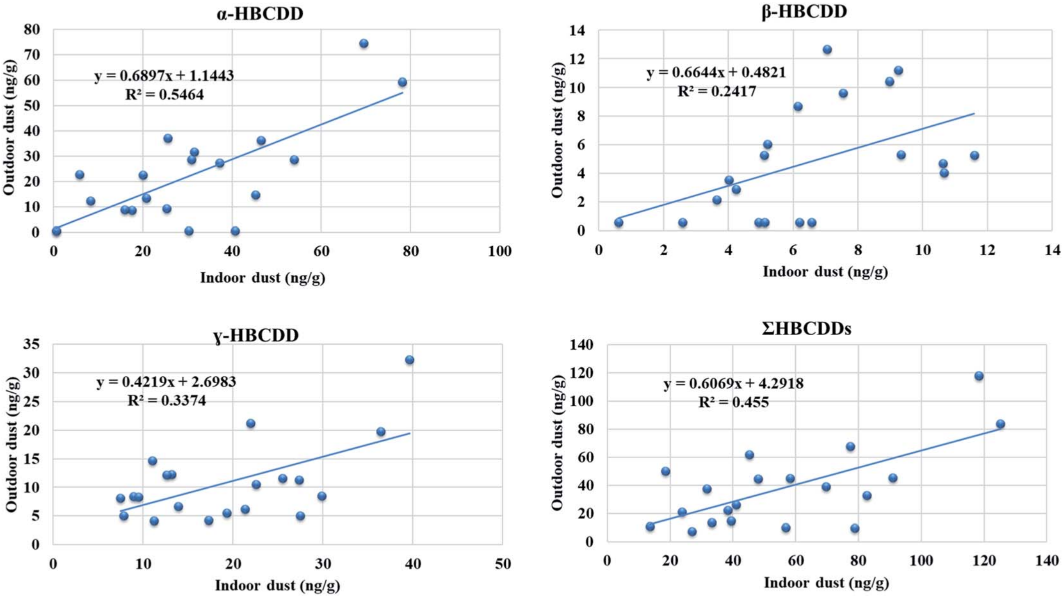

Fig. 3 Correlations between concentrations of HBCDDs in indoor and outdoor dust samples.

Table 3 Estimated exposure (ng kg^{-1} body weight per day) of adults and toddlers of HBCDDs *via* dust ingestion from elevated surface dust (ESD), floor dust (FD) and outdoor dust (OD) in Basrah, Iraq

	ESD				FD				OD			
	Adults		Toddlers		Adults		Toddlers		Adults		Toddlers	
HBCDDs	Median	95th %ile	Median	95th %ile	Median	95th %ile	Median	95th %ile	Median	95th %ile	Median	95th %ile
Mean dust ingestion scenario												
α-HBCDD-ESD	0.0056	0.012	0.11	0.24	0.0037	0.0087	0.072	0.17	0.00022	0.00071	0.058	0.19
β-HBCDD-ESD	0.0011	0.0021	0.022	0.041	0.0010	0.0019	0.020	0.038	0.00005	0.00013	0.014	0.035
γ-HBCDD-ESD	0.0032	0.0061	0.062	0.12	0.0020	0.0064	0.038	0.13	0.00010	0.00026	0.026	0.068
ΣHBCDDs-ESD	0.0096	0.019	0.19	0.38	0.0065	0.016	0.13	0.32	0.00041	0.0010	0.11	0.27
High dust ingestion scenario												
α-HBCDD-ESD	0.017	0.037	0.22	0.48	0.011	0.026	0.14	0.34	0.0089	0.029	0.12	0.38
β-HBCDD-ESD	0.0033	0.0063	0.043	0.082	0.0030	0.0058	0.040	0.075	0.0021	0.0054	0.027	0.070
γ-HBCDD-ESD	0.0095	0.018	0.12	0.24	0.0059	0.019	0.077	0.25	0.0040	0.010	0.053	0.14
ΣHBCDDs-ESD	0.029	0.058	0.38	0.76	0.019	0.049	0.25	0.64	0.017	0.041	0.22	0.53

ingestion ($EDI_{ingestion}$; ng kg^{-1} bw per day) were calculated as shown in the equation below and described elsewhere[83,85,100]

$$EDI = (C_{dust} \times IR_{dust} \times AF_{gastro} \times FTH)/BW$$

where C_{dust} is the concentration of HBCDDs in dust samples (ng g^{-1}), IR_{dust} is the dust ingestion rate (mg day^{-1}), AF_{gastro} is the gastrointestinal absorption fraction (assumed to be 100%), FTH is the fraction of time spent at home (64% for adults and 86% for toddlers indoors, and 4.7% for outdoors for both), and BW is the body weight (80 kg for adults and 13.8 kg for toddlers).

Table 3 summarises EDIs of HBCDDs *via* dust ingestion under various exposure scenarios. For adults, EDIs of ΣHBCDDs assuming median concentrations in dust and mean dust ingestion rates were between 0.00041 (assuming ingestion of OD only) and 0.0096 ng kg^{-1} bw per day (assuming ingestion of ESD only); while for toddlers, they were between 0.11 and 0.13 ng kg^{-1} bw per day assuming ingestion of OD and FD respectively. Assuming ingestion at the high intake rate of dust contaminated at the 95th percentile concentration (*i.e.* high-end exposure scenario); EDIs of ΣHBCDDs for adults were 0.058, 0.059, and 0.041 ng kg^{-1} bw per day, and for toddlers were 0.76, 0.64 and 0.53 ng kg^{-1} bw per day assuming ingestion of ESD, FD and OD respectively. This implies that compared with adults, EDIs of ΣHBCDDs for toddlers are substantially higher (by factors of 13–20) due to their lower body mass and greater assumed dust ingestion rate. However, these EDIs are uncertain as adults are more likely to ingest ESD, while toddlers are more likely to ingest FD. Taking this into account and assuming adults and toddlers ingest ESD and FD respectively, the EDIs of ΣHBCDDs for toddlers will exceed those of adults by approximately 8–13 times. Even under the high-end exposure scenario, our EDIs were orders of magnitude lower than the reference dose (RfD) value for HBCDD of 200 000 ng kg^{-1} bw/day promulgated by the United States National Academy of Sciences (NAS).[101] On this basis, the current study suggest HBCDD exposure of the Iraqi population *via* dust ingestion is not a health concern. As a cautionary note however, the NAS has expressed low confidence in this RfD for HBCDD, and in 2016, the UK's Committee on Toxicity reported that "high levels (of HBCDD) in household dust continues to be a cause for concern" and cited a toxicological reference point of 3000 ng kg^{-1} bw per day.[102]

4. Conclusion

This is the first study to report concentrations of HBCDDs in dust samples from Iraq, in either indoor or outdoor house dust. In general, the concentrations of HBCDDs were much lower than those in Europe and North America. This likely demonstrates lower application in Iraq of EPS and XPS foams as insulation material (the primary use of HBCDDs). Thus, we propose that the low levels of HBCDDs in this study can be attributed to emissions from electronic and electrical equipment, and textiles in the homes studied. Concentrations of HBCDDs were significantly higher in elevated surface dust compared to those in floor dust from the same rooms. This implies that human exposure assessments based on FD only are underestimates, particularly for adults as they interact more with ESD. The presence of non-negligible concentrations of HBCDDs in outdoor dust from front yards indicates the importance of this area when assessing human exposure. Despite the valuable contribution to the body of knowledge on human exposure to HBCDDs *via* indoor and outdoor dust ingestion, the limitation of this study was in sampling design. Monitoring dust from the interior of cars parked in the front yards as well as dust from the same yards would further assist evaluation of the significance of cars as a source of HBCDDs.

Conflicts of interest

There are no conflicts to declare.

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