



Distribution of brominated flame retardants and phthalate esters in house dust in Korea

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ABSTRACT

We examined the levels of brominated flame-retardants (BFRs) including polybrominateddiphenyl ethers (PBDEs), tetrabromobisphenol-A (TBBPA), hexabromocyclododecane (HBCD), and phthalates in indoor dusts in residential houses in Korea, and their distribution patterns depending on building characteristics. Mean concentrations of phthalate esters ($1,825 \mu\text{g g}^{-1}$) were significantly higher than that of BFRs (PBDE: $1,332 \text{ ng g}^{-1}$, HBCDs: 459 ng g^{-1} , and TBBPA: 213 ng g^{-1}), indicating more frequent use of phthalate-containing products such as PVC flooring in the Korean houses. PVC flooring house was associated with higher concentrations of DEHP ($p = 0.001$) and BBP ($p = 0.012$), indicating that exposure to phthalate was higher in the PVC flooring house. Building age was significantly related with levels of PBDEs especially BDE-47 ($p = 0.062$), BDE-203 ($p = 0.007$), DEHP ($p = 0.004$), and BBP ($p = 0.070$), respectively, indicating that older buildings can produce higher amounts of PBDEs and phthalates. Our study can provide important information on the sources of BFRs and phthalates in residential houses in Korea.

Keywords: Construction age, DEHP, House dust, PBDE, Phthalates, PVC flooring

1. Introduction

There has recently been increasing concern about chemicals that may disrupt the endocrine systems of humans and animals. One such group of chemicals is brominated flame-retardants (BFRs). BFRs, especially polybrominateddiphenyl ethers (PBDEs), tetrabromobisphenol-A (TBBPA), and hexabromocyclododecane (HBCD), have been widely used as fire retardants and plasticizers in numerous plastic products [1-3]. PBDEs can act as endocrine disruptors that mimic or interfere with the function of hormones, such as those in the thyroid gland [4]. Due to the risk posed by PBDEs, international movements to regulate their use in home appliances and plastic equipment have arisen, including the Restriction of Hazardous Substances Directive (RoHS) and the Waste Electrical and Electronic Equipment Directive (WEEE) [2].

The movement to regulate the use of HBCD and TBBPA, which are alternatives to PBDEs, has also emerged [5, 6]. HBCD is considered persistent and bio-accumulative, and might cause reproductive, developmental, and neurological disorders. TBBPA is known to have adverse effects on thyroid hormones.

Under the EU's REACH (Regulation, Evaluation, Authorization, and restriction of Chemicals) program, HBCDs were phased out in European trade by mid-2015 [5]. TBBPA was also registered under REACH in October 2010 [7, 8].

Phthalates have been widely used as plasticizers, added to plastics to increase their flexibility, transparency, durability, and longevity. They are used in a variety of electrical and electronic goods (connectors, capacitors, and cables), household applications (shower curtains, vinyl upholstery, food containers, and wrappers), and construction materials (adhesives, floor tiles, and wall coverings) [9]. Phthalates may mimic estrogenic hormones and have inhibitory developmental effects on humans [10, 11]. Among the phthalates, bis (2-ethylhexyl) phthalate (DEHP) in particular causes damage to the liver and testes [12]. As a result, an EU risk assessment classified DEHP, dibutyl phthalate (DBP), and benzyl butyl-phthalate (BBP) as hazardous substances in 2005, and has issued directives to ban these materials from products, particularly toys and cosmetics [13].

The sources of human exposure to BFRs and phthalates have been reported to be indoor environments and diet [14, 15]. Even



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diet is a main source of PBDE and phthalate exposure, sources such as indoor house dust are also another significant human exposure routes. Recent studies reported that BFRs and phthalates in residential houses [16-18]. However, there are no studies on the measurement of both BFRs and phthalates together in indoor environment.

In this study, we determined the concentrations of the main BFRs (PBDEs, TBBPA, and α -, β -, and γ -HBCD) and phthalate esters by analyzing settled house dust from residential houses in Korea. The results were compared to the levels reported in related researches in other countries. In addition, the correlations between the levels and building characteristics such as flooring type (PVC or wood) and building age were performed.

2. Methods and Materials

2.1. Chemicals and Materials

N-hexane (PRA grade) was purchased from SK chemical (Korea), ethyl acetate (HPLC grade) was purchased from Burdick & Jackson (USA), and methanol (HPLC grade) was purchased from J.T. Baker (USA). Phthalate esters, PBDE congeners, α -, β -, γ -HBCD, and TBBPA were all purchased from Accustandard (USA).

The mixture standard solution of phthalate esters contained 15 kinds of phthalates; dimethyl phthalate (DMP), diethyl phthalate (DEP), diisobutyl phthalate (DIBP), DBP, bis(methoxyethyl) phthalate (DMEP), bis(4-Methyl-2-pentyl)phthalate (DMPP), diethoxyethyl phthalate (DEEP), diamyl phthalate (DAP), BBP, dihexyl phthalate (DHP), di(butoxyethyl)phthalate (DBEP), dicyclohexylphthalate (DCHP), bis(2-ethylhexyl) phthalate (DEHP), di-n-octyl phthalate (DNOP), and dinonyl phthalate (DNP). Likewise, the mixture standard solution of PBDE congeners was prepared by the product named 'Custom PBDE congeners' which was composed of 13 kinds commonly used in industry circle; 2-bromodiphenyl ether (BDE-1), 2,4-bibromodiphenyl ether (BDE-7), 2,4,4'-tribromodiphenyl ether (BDE-28), 2,2',4,4'-tetrabromodiphenyl ether (BDE-47), 2,2',4,4',5-pentabromodiphenyl ether (BDE-99), 2,2',4,4',6-pentabromodiphenyl ether (BDE-100), 2,2',4,4',5,6'-hexabromodiphenyl ether (BDE-154), 2,2',3,4,4',5,6'-heptabromodiphenyl ether (BDE-183), 2,2',3,4,4',5,5',6-octabromodiphenyl ether (BDE-203), 2,2',3,3',4,4',5,5',6-nonabromodiphenyl ether (BDE-206), 2,2',3,3',4,4',5,6,6'-nonabromodiphenyl ether (BDE-207), 2,2',3,3',4,5,5',6,6'-nonabromodiphenyl ether (BDE-208), and decabromodiphenyl ether (BDE-209). The mixture solution of HBCDs and TBBPA was prepared by mixing each single standard solution (Accustandard, USA) using methanol.

The deuterium ring-labelled reference materials of dibutyl-phthalate (di-n-butyl phthalate-3,4,5,6-d₄, DBP-d₄) as a surrogate material for phthalates was purchased from Fluka (USA) [9]. 2,3,5,6-tetrachlorobiphenyl (PCB-65) as a surrogate for low mass PBDEs, HBCDs, and TBBPA was purchased from Accustandard [16]. Tetrachloro-m-xylene (TCMX) was purchased from Accustandard [16] and served as an internal standard, which calibrates final volumes and measuring

sensitivity. PCB-166 was used as a surrogate for high mass PBDE congeners.

The individual stock solutions were prepared appropriately, and kept in a refrigerator at 4°C.

2.2. Sampling

Dust samples were collected from residential houses (n = 42) from different indoor environments in Seoul and Kyung-Gi Province, Korea. During sampling, the factors such as residential type (22 apartments, 13 multiplex, and 7 single studio rooms), building construction age (before 1990, 1991 to 2000, and after 2001), and flooring materials (PVC linoleum and wood type) were also examined using a checklist form [19]. The detailed information of sampling houses is provided in the supplementary materials (Table 1).

Table 1. Summary of House Dust Sampling Information

Items	House
Number of samples	42
Sampling period	May 2011 to September 2011
Locations	Seoul/Kyung-gi Province
Sampling device	Vacuum cleaner
Remarks	Floor: PVC, Wood, laminate Ceiling and wall: laminated paper
Residential type	Apartment (22), Multiplex (13), Single studio room (7)
Floor material	PVC (34), Wood (8)
Construction age	Before 1990 (10), 1991-2000 (21), After 2001 (11)

Duplicate samples were obtained from the residential homes using a vacuum cleaner. After the sampling, large debris, hair and non-dust particles in the samples were removed using a coarse metal sieve. To avoid contamination, the collected samples were immediately transferred to amber glass bottles and stored at 20°C until analysis.

2.3. Sample Extraction

To reduce the risk of lab- and cross-contamination, the extraction method developed by Abb et al. [13] was used. A detailed description of the extraction and analysis procedures of BFRs and phthalates were provided in Fig. S1. Approximately 250 mg of dust was put in a 30 mL extraction vial and added 20 mL of ethyl acetate and n-hexane solution (1:1). After spiking 50 μ L of the surrogate which was the mixture solution made up of PCB-65 (40 μ g mL⁻¹), PCB-166 (5 μ g mL⁻¹), and DBP-d₄ (100 μ g mL⁻¹) [9], the ultrasonic extraction was performed for 60 min at 40°C, and then followed by letting them cool at room temperature. The amount of 2 spatulas of anhydrous sodium sulfate was added to each the extract for water elimination. The supernatant was filtered with a glass syringe (Jamsil Medical) and a hydrophobic filter (AD.25JP050AN, ADVANTEC) for a simple clean up thereafter.

Due to the lower concentrations but higher method detection

limits of PBDEs, exactly 10 mL volume of each the extract was transferred with a volume pipette to a concentration vial separately, and then concentrated into 0.5 mL using nitrogen gas in a Turbovap@II (Zymark). The rest extracts was directly used to analyze phthalates with GC/MS and to analyze HBCDs/TBBPA with LC/MS/MS. Especially, since DEHP showed higher deviation far from the calibration range, each the extract was diluted to 1:10 or 1:100 with n-hexane in additional vials. The final extract for phthalates and HBCDs/TBBPA were carried to the 2 mL vials and set up to 1.5 mL as final volume, but that for PBDEs were set up to 0.5 mL. Prior to analyze them, an internal standard, TCMX ($2 \mu\text{g mL}^{-1}$) were spiked into them skillfully. Each sample was analyzed in duplicate for each determination [20].

2.4. GC/MS Analysis

After the extraction, PBDEs and phthalates were analyzed by GC/MS (Perkinelmer Clarus-500). The separation of PBDEs was operated by a DB-5MS capillary column ($15 \text{ m} \times 0.25 \text{ m} \times 0.1 \mu\text{m}$). The samples were injected by an amount of $1 \mu\text{L}$ in splitless mode. Helium was used as carrier gas at constant flow (1 mL min^{-1}) and nitrogen was used as make-up gas at constant flow (4 mL min^{-1}). The temperature condition was programmed from 80°C , kept for 1 min, then increased with $10^\circ\text{C min}^{-1}$ to 170°C , kept for 2 min, then increased with $10^\circ\text{C min}^{-1}$ to 260°C , kept for 0 min, and then increased with $20^\circ\text{C min}^{-1}$ to 300°C , kept for 8 min, so took totally 31 min. The MS was operated with a selected ion monitoring (SIM) mode in electron ionization inner source. The ion source, quadruple, and interface temperature were 300, 280, and 300°C , respectively.

The determination and separation of phthalates using GC/MS were as follows; the column is different from PBDEs, which was DB-5MS capillary column ($30 \text{ m} \times 0.25 \text{ m} \times 0.25 \mu\text{m}$). The injection volume was $1 \mu\text{L}$ in split mode (20:1) and injector temperature was 260°C . The temperature condition was set from 100°C , kept for 4 min, and then increased with $10^\circ\text{C min}^{-1}$ to 260°C , kept for 10 min, so took 30 min in total. The ion source quadruple and interface temperature were all 280°C . The other conditions were identical with PBDEs.

The contamination of extraction solvents (n-hexane and ethyl acetate) with BFR and phthalate ester residues was also checked by LC/MS-MS.

2.5. LC/MS/MS Analysis

The determination of ΣHBCDs and TBBPA was achieved using a MS/MS system of AB-Sciex (Q-Trap 3200) and a HPLC system of Perkinelmer (FX HPLC system). α -, β -, and γ -HBCD and TBBPA were to be separated and quantified in one method with Thermo CCS Hypercarb C18 column ($150 \times 4.6 \text{ mm i.d.}$, $5 \mu\text{m}$ particle size). A mobile phase of de-ionized water and methanol at a flow rate of total $400 \mu\text{L min}^{-1}$ was applied for elution of HBCD isomers. The mobile phase flowed at the gradient condition whose step was starting 90% methanol was held for 1 min then decreased linearly to 10% over 2 min. This rate was held for 3 min followed by a linear increase to 90% methanol over 4 min and held for 2 min. Totally it took 12 min. Using this method, HBCDs were separated with retention times of 7.93, 8.66, and 9.20 min for α -, β -, and γ -isomers, respectively. The peak of TBBPA also was shown at 5.40 min. Mass spectrometric analysis was performed in the electro-spray negative ionization mode and in Multiple Reaction Monitoring (MRM) mode for quantitative determination. HBCD isomers were analyzed based on m/z 604.3 to 80.8 and TBBPA was based on m/z 542.6 to 78.7.

The detailed QA/QC during the analysis of PBDEs and phthalates is provided in the supplementary materials.

2.6. Data Analysis

For statistical analysis, the mean value was obtained by the application of recommended environmental exposure point concentrations ver2.2 (ALCOA Inc.), which was a simple statistical tool using a personal computer. This representative value was called 'EPC (exposure point concentration)' and was calculated by distinction of the distribution characteristics. Results were considered statistically significant, if the p-value was less than 0.05.

3. Results and Discussion

3.1. Levels of BFRs and Phthalates in House Dust

The concentration ranges of phthalate esters in all dust samples were summarized in Table 2. Mean concentrations of phthalate esters ($1,825 \mu\text{g g}^{-1}$) were significantly higher than that of BFRs (PBDE: $1,332 \text{ ng g}^{-1}$, HBCDs: 459 ng g^{-1} , and TBBPA: 213 ng g^{-1} , respectively). This result indicates that phthalates have been

Table 2. Concentrations of PBDEs and Phthalates in the House Dust Samples

Site	Compound	Phthalates ($\mu\text{g g}^{-1}$) ¹⁾	PBDEs (ng g^{-1}) ²⁾	HBCDs (ng g^{-1}) ³⁾	TBBPA (ng g^{-1})
house (n = 42)	Min	175	396	< LOD ⁴⁾	< LOD
	Max	4,491	3,346	3,132	2,092
	Mean	1,825	1,332	459	213
	median	1,550	1,257	278	69

1) esters: DMP, DEP, DIBP, DBP, DMPEP, DMPP, DMPP, DEEP, DAP, DHP, BBP, DBEP, DCHP, DEHP, DNOP, DNP (total 15 compounds)

2) congeners: BDE-1, BDE-7, BDE-28, BDE-47, BDE-100, BDE-99, BDE-153, BDE-183, BDE-203, BDE-207, BDE-208, BDE-206, BDE-209 (total 13 compounds)

3) isomers: α -HBCD, β -HBCD, γ -HBCD

4) below LODs (limit of detection)

widely used in building materials and electronic equipment, compared to BFRs. Among the BFRs, PBDE concentrations were 3-6 times higher than HBCDs and TBBPA concentrations ($p < 0.05$). This result might be due to that HBCDs and TBBPA have been used as alternatives to PBDEs only recently compared to PBDEs. Thus, PBDEs levels were still higher than HBCD and TBBPA, indicating that PBDEs have been accumulated to dust since they were used vastly until banned [21].

3.2. Σ PBDE levels in House Dust

The concentration of each PBDE in the house dust presents in Table 3. The median concentration of Σ PBDEs in the house dust was $1,257 \text{ ng g}^{-1}$ (ranged $396\text{-}3,346 \text{ ng g}^{-1}$). Most PBDE congeners were detected at a high frequency (frequency of 70-100%), except BDE-28 (23% frequency) (Table 3). Among PBDE congeners, BDE-206, -208, and -209 were detected in all dust samples (100%). BDE-47, -99, and -207 were detected with 97% frequency. BDE-7 (79%), BDE-28 (23%), BDE-100 (85%), and BDE-203 (56%) were detected at a lower frequency. In contrast, BDE-1, BDE-153, and BDE-183 were not detected in house dust samples. This was consistent with those observed in house dust samples collected in other Korean city [17, 22].

The median concentration of BDE-209 in house dust was highest at 315 ng g^{-1} (contribution of 25%). Similar to our result, Lee et al. [22] showed that the predominant congener of PBDEs observed in house dust samples was BDE-209, accounting for $83 \pm 12\%$ of Σ PBDEs. Other Korean study [23] reported that the three congeners of BDE-206, 207, and 209 in all dust samples were detected, and in particular, BDE-209 was found as a dominant congener most of them. In addition, Kim [23] and La Guardia et al. [24] found the relatively high proportions of deca- and nona-BDEs in the profile congeners of the technical mixture products of deca-BDE, such as Saytex 102E or Bromkal 82-0DE. Thus, this result might be due to the intensive use and no regulation of deca-BDE product in Korea [25].

Unlike previous Korean studies [17, 22, 23], BDE-47 (a median

$= 304 \text{ ng g}^{-1}$) was the secondarily abundant congener, accounting for 24% of Σ PBDEs (Table 3). According to Kim [23] and La Guardia et al. [24], tri- to hexa-BDEs (particularly, BDE-47, 99, and 100) are dominant congeners in penta-BDE product such as DE-71 or Bromkal 70-5DE. From our study, penta-BDE product, containing lighter congeners as the dominant ingredient, might have been used consistently in relative proportion with deca-BDE product in Korea, or breakdown from the heavier congeners to lighter ones in house dust.

As shown in Table 4, the levels of BFRs in our study were compared with those reported in other countries. Although the levels of BFRs in this study are significantly higher than that reported a median concentration of 227 ng g^{-1} ($35\text{-}576 \text{ ng g}^{-1}$) in Korean other study [23], the levels of Σ PBDE concentration (median; $1,257 \text{ ng g}^{-1}$ ($396\text{-}3,346 \text{ ng g}^{-1}$) in this study was lower than that in the United States ($4,200 \text{ ng g}^{-1}$) and U.K. ($8,500 \text{ ng g}^{-1}$). DIBP concentration in this study was comparable to that reported from the China and Australia (ranging from 571 to $1,876 \text{ ng g}^{-1}$).

3.3. HBCD and TBBPA Levels in House Dust

As the alternatives to PBDEs, the median concentrations of HBCDs and TBBPA in house dust were 278 ng g^{-1} ($< \text{LOD}\text{-}3,132 \text{ ng g}^{-1}$) and 69 ng g^{-1} ($< \text{LOD}\text{-}2,092 \text{ ng g}^{-1}$), respectively (Table 5). As shown in Table 5, the Σ HBCD concentrations of home dust in the U.K. ($2,401$ and 730 ng g^{-1}), Canada (640 ng g^{-1}), and the United States (390 ng g^{-1}) showed relatively higher concentrations compared to our results. On the other hand, the studies conducted in New Zealand and Japan reported levels similar with our result [35, 36]. The lower concentrations of Σ HBCD in home dust in Korea could be because Koreans generally do not use carpet in their homes. One previous study reported that carpet probably was one source of HBCD in home dust [39].

As shown in Table 5, the median TBBPA levels in house dust from the United States (200 ng g^{-1} ; ranged $22\text{-}2,000 \text{ ng g}^{-1}$) [27], Japan (505 ng g^{-1} ; ranged $490\text{-}520 \text{ ng g}^{-1}$) [37], and the U.K. (87 ng g^{-1} ; ranged $< \text{LOD}\text{-}382 \text{ ng g}^{-1}$) [5] showed relatively

Table 3. Summary of Concentrations of PBDE Congeners in the House Dust Samples

Compound ¹⁾	Mean ²⁾ (ng g^{-1})	Median (ng g^{-1})	Range (ng g^{-1})	s.d. (ng g^{-1})	Proportion ³⁾ (%)	Frequency (%)
BDE-7	91	87	< LOD-229	69	7	79
BDE-28	11	< LOD	< LOD-70	21	-	23
BDE-47	285	304	< LOD-585	147	24	97
BDE-99	162	76	< LOD-1,701	324	6	97
BDE-100	60	45	< LOD-454	75	4	85
BDE-203	37	46	< LOD-122	36	4	56
BDE-206	84	77	33-166	30	6	100
BDE-207	75	68	40-165	33	5	97
BDE-208	82	78	36-144	28	6	100
BDE-209	444	315	61-1,652	321	25	100
Σ PBDEs	1,332	1,257	396-3,346	585	-	-

1) BDE-1, BDE-153, BDE-183 were detected as below LOD

2) Application of recommended environmental exposure point concentrations ver2.2 (ALCOA Inc.)

3) Proportion data was based on the median value

Table 4. Comparisons of the Concentration of Σ PBDEs with Other Related Studies

	Country	Median (ng g ⁻¹)	Range (ng g ⁻¹)	Reference
House dust	South Korea (n = 42)	1,257	396-3,346	This study
	South Korea (n = 4)	227	35-576	Kim [23]
	United States (n = 10)	4,200	520-29,000	Sjödín et al. [26]
	United States (n = 20)	13,723	3,020-192,100	Allen et al. [6]
	United States (n = 16)	3,931	534-85,550	Dodson et al. [27]
	United Kingdom (n = 30)	8,500	12-220,000	Stuart et al. [28]
	Australia (n = 10)	1,200	500-13,000	Sjödín et al. [29]
	Australia (n = 30)	571	61-82,400	Stasinska et al. [29]
	Germany (n = 10)	74	17-550	Sjödín et al. [26]
	China (n = 76)	1,876	186-9,654	Huang et al. [30]
	China (n = 44)	721	132-3,887	Yu et al. [31]
Sources	TV dust	72,134		Huang et al. [30]
	Computer dust	6,272		Huang et al. [30]

Table 5. Comparisons of HBCDs and TBBPA as the Median Values with Other Related Studies

ng g ⁻¹	α -HBCD	β -HBCD	γ -HBCD	Σ HBCDs	TBBPA
This study (n = 42)	144 (< LOD-1,752)	12 (< LOD-322)	60 (< LOD-2,897)	278 (< LOD-3,132)	69 (< LOD-2,092)
Korea (n = 46) (Barghi et al. [32])	43 (10-1,327)	6 (1-217)	46 (ND-1,101)	106 (19-2,645)	79 (14-1,212)
U.K. (n = 42) (Abdallah and Harrad [33])	876 (112-40,653)	300 (25-25,513)	886 (80-74,607)	2,401 (228-140,774)	-
U.K. (n = 31) (Abdallah et al. [34])	170 (22-66,000)	66 (9-7,800)	440 (70-37,000)	730 (140-110,000)	-
Canada (n = 8) (Abdallah et al. [34])	300 (25-670)	72 (6-130)	230 (34-470)	640 (64-1,300)	-
U.S. (n = 42) (Abdallah and Harrad [33])	80 (17-1,800)	28 (6-300)	300 (79-2,000)	390 (110-4,000)	-
New Zealand (n = 50) (Ali et al. [35])	99 (3-1,790)	12 (3-270)	96 (8-3,020)	190 (20-4,100)	-
Japan (n = 33) (Tue et al. [36])	48 (3-340)	4.7 (1-34)	8 (2-110)	120 (5-400)	-
Belgium (n = 18) (Geens et al. [8])	-	-	-	-	10 (1-1,480)
U.K. (n = 35) (Abdallah et al. [5])	-	-	-	-	87 (< LOD-382)
U.K. (n = 35) (Geens et al. [8])	-	-	-	-	60 (190-340)
U.S. (n = 16) (Dodson et al. [27])	-	-	-	-	200 (22-2,000)
Japan (n = 2) (Takigami et al. [37])	-	-	-	-	505 (490-520)
Korea (n = 16) (Wang et al. [38])	-	-	-	-	84 (43-370)

higher concentrations compared to our study. However, the median TBBPA levels in house dust from Belgium (10 ng g⁻¹) and the U.K. (60 ng g⁻¹) showed relatively lower concentrations compared to our study (Table 5). Recently, Barghi et al. [32] reported the concentrations of TBBPA in indoor dust samples from indoor environments in Korea. The highest TBBPA median concen-

tration was found in offices (463.81 ng g⁻¹), followed by schools (204.16 ng g⁻¹) > Kindergartens (185.03 ng g⁻¹) ≥ public indoor environments (183.39 ng g⁻¹) > cars (81.37 ng g⁻¹) > homes (78.87 ng g⁻¹).

The levels of HBCDs and TBBPA in the U.K., the United States, and Canada are generally higher than the levels measured

in this study. This might be because these substances began to be used earlier in these countries as alternatives to PBDEs.

Interestingly, α - and γ -HBCDs were generally detected at higher levels than β -HBCD at each site (Table 5). In particular, α -HBCD levels were consistently higher than that of γ -HBCD and β -HBCD. Especially, β -HBCD was barely detected (α :- 52%, γ :- 24%, and β :- 4%). Barghi et al. [32] reported that the isomeric distribution of HBCD showed a higher contribution of α -HBCD compared to β - and γ -HBCD in the collected indoor dust samples except the office samples from Korea. Similar patterns (α - > γ - > β -HBCD) were found in house dust samples from Canada (α :- 47%, γ :- 37%, and β :- 16%) [34] and Japan (α :- 79%, γ :- 13%, and β :- 7%) [36]. However, Abdallah et al. [5] reported that γ -HBCD was present at the highest concentration, followed by α -HBCD and β -HBCD in the U.K. (γ :- 60%, α :- 23%, and β :- 9%) and the U.S. (γ :- 77%, α :- 21%, and β :- 7%).

3.4. Phthalates in House Dust

The median concentration of total phthalates in house dusts

in this study was 1,550 $\mu\text{g g}^{-1}$ (175-4,491 $\mu\text{g g}^{-1}$) (Table 6). Among the phthalates, DEHP was highly dominant (median 1,488 $\mu\text{g g}^{-1}$), detected at the levels ranging from 114 to 4,321 $\mu\text{g g}^{-1}$ with 100% frequency, then followed by DBP (median 18.3 $\mu\text{g g}^{-1}$), BBP (median 12.8 $\mu\text{g g}^{-1}$), and DIBP (median 2.9 $\mu\text{g g}^{-1}$), respectively. DBP, BBP, and DIBP were also detected in the majority of house dust samples. The trend that DEHP was detected at the highest levels, followed by DBP and BBP in dust samples was similar with other studies. Li et al. [18] also reported that DEHP, DBP, and DIBP were the dominant phthalates in dormitory dust. In contrast, DIBP, DNOP₁ and DHP were detected less frequently. The other phthalates were detected as below LOD.

The concentrations of phthalate esters reported in this study were generally higher than in the levels reported from Europe and the United States, and similar with those measured in Asia (Table 7). The median range for phthalates reported was 500-1,000 $\mu\text{g g}^{-1}$ in Germany, 412-662 $\mu\text{g g}^{-1}$ in the U.S., 869 $\mu\text{g g}^{-1}$ in Denmark, and 1,100 $\mu\text{g g}^{-1}$ in Sweden, respectively. On the other hand, higher median values of phthalates were

Table 6. Concentrations ($\mu\text{g g}^{-1}$) of Phthalate Esters in the House Settled Dust

$\mu\text{g g}^{-1}$	Mean ¹⁾	Median	Range	Frequency (%)
DIBP	4.9	2.9	< LOD-21.1	81
DBP	34.6	18.3	< LOD-190.7	98
DAP	0.4	< LOD	< LOD-4.5	17
DHP	0.1	< LOD	< LOD-1.1	14
BBP	52.1	12.8	< LOD-444.4	74
DEHP	1,762	1,488	114-4,321	100
DNOP	6.2	3.9	< LOD-15.4	69
Σ Phthalate ²⁾	1,825	1,550	175-4,491	

1) Application of recommended environmental exposure point concentrations ver2.2 (ALCOA Inc.)

2) DMP, DEP, DMEP, DMPP, DEEP, DBEP, DCHP and DNP were detected as below LOD

Table 7. Comparisons of the Concentrations of Σ Phthalate Esters in the House Dusts with Other Studies

Median ($\mu\text{g g}^{-1}$)	Σ Phthalates	DEHP	DBP	BBP	Reference
South Korea (n = 42)	1,550	1,488	18	13	This study
Germany (n = 29)	1,049	970	51	28	Butte et al. [40]
Germany (n = 30)	780	703	47	30	Fromme et al. [9]
Germany (n = 65)	769	600	47	19	Kersten and Reich [41]
Germany (n = 272)	564	450	87	24	Pöhner et al. [42]
USA (n = 278)	662	480	29	13	Nagorka et al. [43]
USA (n = 120)	412	340	20	45	Rudel et al. [44]
Denmark (n = 30)	869	604	87	15	Langer et al. [45]
Sweden (n = 346)	1,100	770	150	135	Bornehag et al. [19]
China (n = 23)	1,272	1,190	77	5	Ait Bamai et al. [46]
Japan (n = 128)	1,126	1,107	17	2	Ait Bamai et al. [46]
Kuwait (n = 21)	2,400	2,256	45	8.6	Gevao et al. [47]
(Sources) ¹⁾					
PVC linoleum (n = 8)	8,183	7,834	9	271	This study
Deposition adhesive (n = 2)	22	22	nd	nd	This study
PVC adhesive (n = 2)	2,656	3	2,653	nd	This study

1) Source concentrations are shown as geometric mean values

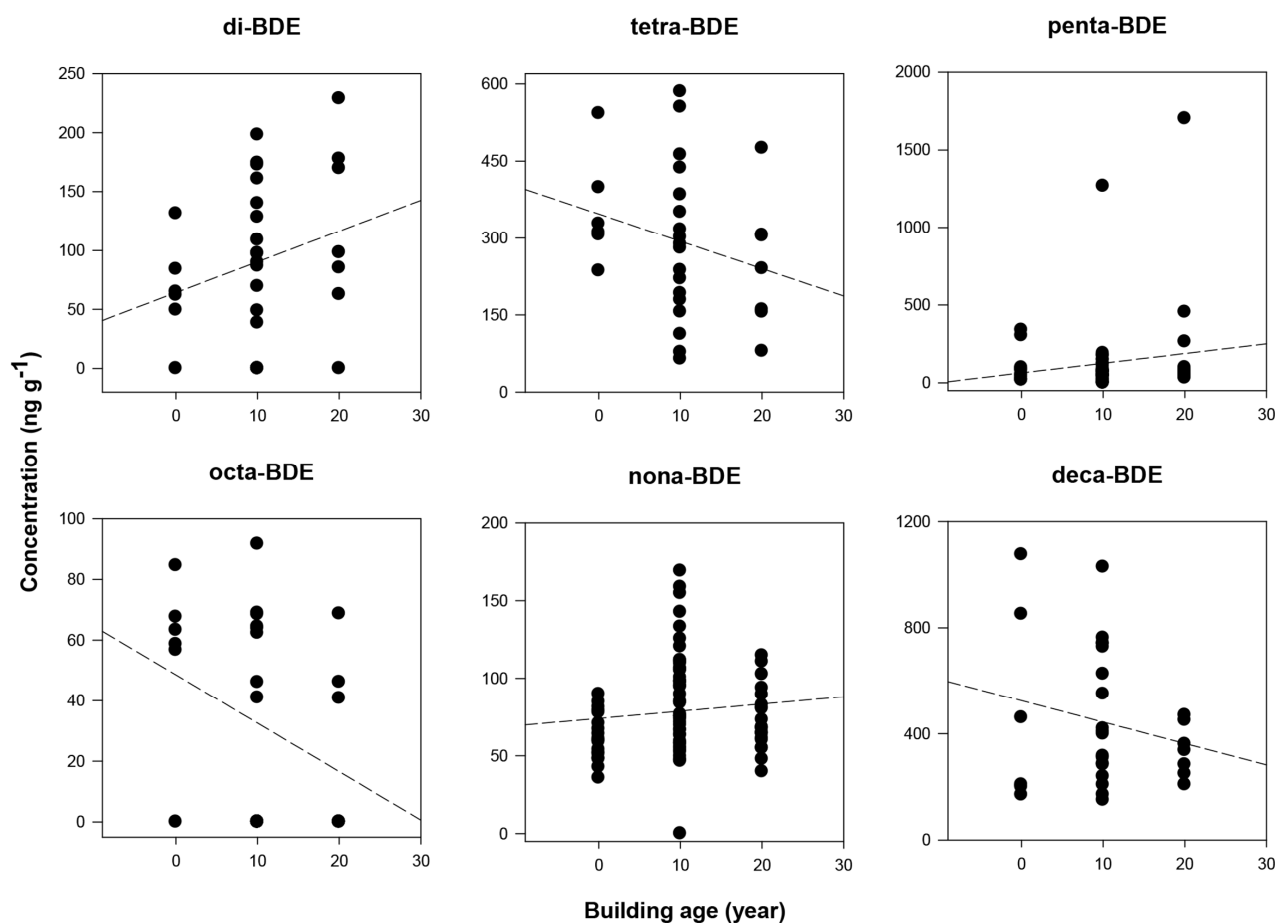


Fig. 1. The relationship between some BFRs concentrations and house age (di-BDE = BDE-7; tetra-BDE = BDE-47; penta-BDE = BDE-99 + BDE-100; octa-BDE = BDE-203; nona-BDE = BDE-206 + BDE-207 + BDE-208; deca-BDE = BDE-209).

observed in Asia (1,550 $\mu\text{g g}^{-1}$ in this study, 1,126 $\mu\text{g g}^{-1}$ in Japan, 1,272 $\mu\text{g g}^{-1}$ in China, and 2,400 $\mu\text{g g}^{-1}$ in Kuwait). The higher the levels of phthalates in Korean houses than other countries can be explained by the fact that PVC linoleums, including synthetic PVC adhesives, are more extensively used in the floors in Korean houses. In fact, we measured the phthalates levels in PVC linoleum ($n = 8$) and PVC adhesive ($n = 2$) used in the sampled houses. The mean concentration of total phthalates in PVC linoleum was 8,183 $\mu\text{g g}^{-1}$, mostly DEHP (7,834 $\mu\text{g g}^{-1}$), and in PVC adhesive, mostly as DBP (2,653 $\mu\text{g g}^{-1}$), indicating that the main source for high concentrations of phthalates are PVC flooring in Korean houses (Table 7).

3.5. Correlations between the Levels of BFRs and Phthalates with Building Characteristics

The association analysis between flooring type (PVC or wood) and building age and the concentrations of 13 congeners of PBDEs, HBCDs, and TBBPA was performed. Table S2 presents that the levels of PBDEs, HBCDs and TBBPA showed no association with PVC flooring type, but there was a weak association with building age. Among the PBDE congeners, BDE-47 ($p = 0.062$),

and BDE-203 ($p = 0.007$) were significantly correlated with building age; total HBCDs were also correlated with building age.

As shown in Fig. S2, deca-BDE can be debrominated over time in indoor environment, and the various PBDE congeners can be detected in dust. Hua *et al.* [48] also observed the reductive debromination of BDE-209 leading to BDE-7 upon solar irradiation. Similar to our result (refer Table 3), Kim [23] reported that the ratio of isomers other than BDE-209 detected in dust was not high, and it could be the result of degradation of environment rather than direct product (penta- or octa-BDE) [23]. Based on these results, although BDE-47 is a dominant congener in penta-BDE product such as DE-71 or Bromkal 70-5DE, BDE-47 might be byproduct of the debromination of BDE-209. Because the concentrations of light congeners (di-BDE to penta-BDE) increased with increasing age of the building and the concentrations of heavy congeners (octa-BDE to deca-BDE) decreased with increasing age of building (Fig. 1).

Next, the association between flooring type and building age, with the concentrations of seven phthalate esters detected was examined. The distribution of surface materials on floors is presented in Table 8. The study cases were PVC-contained linoleums and the wood flooring materials were used as a control. Although

Table 8. The Associations of the Levels of Σ Phthalate Esters with the Types of Floors in Settled Dust

$\mu\text{g g}^{-1}$	Type of flooring		p-Value ¹⁾	Construction Age			p-Value ²⁾
	PVC (n = 34)	Wood (n = 8)		< 10 y (n = 6)	10-20 y (n = 19)	> 20 y (n = 9)	
DIBP	2.9 (0.0-21.1)	2.6 (0.0-6.6)	0.718	4.5 (2.6-21.1)	2.4 (0.0-16.9)	3.7 (0.0-6.1)	0.499
DBP	22.9 (1.1-190.7)	9.4 (0.0-38.1)	0.012	13.9 (3.3-47.4)	21.9 (2.7-190.7)	34.3 (1.1-85.0)	0.070
DAP	0.0 (0.0-4.5)	0.0 (0.0-2.7)	0.648	0.0 (0.0-4.5)	0.0 (0.0-1.8)	0.0 (0.0-3.5)	0.308
BBP	12.8 (0.0-444.4)	8.6 (0.0-47.1)	0.718	9.8 (0.0-152.5)	8.1 (0.0-403.0)	14.1 (3.1-444.4)	0.397
DHP	0.0 (0.0-0.8)	0.0 (0.0-1.1)	0.498	0.0 (0.0-0.7)	0.0 (0.0-0.7)	0.0 (0.0-0.8)	0.565
DEHP	1,720 (415-4,321)	699 (114-1,534)	0.001	1,179 (415-1,602)	1,690 (519-4,321)	2,194 (1,262-4,279)	0.004
DNOP	4.5 (0.0-15.4)	0.0 (0.0-8.4)	0.087	3.7 (0.0-10.8)	4.9 (0.0-15.4)	5.4 (0.0-8.6)	0.992
Σ Phthalate	1,823 (451-4,491)	764 (175-1,576)	0.001	1,253 (451-1,675)	1,784 (634-4,491)	2,302 (1,295-4,370)	0.004

1) Mann-Whitney U-test regarding differences in phthalate concentration between PVC flooring and wood as flooring material

2) Spearman correlation test of between construction age and phthalate concentration (correlative in case p-value is below 0.01)

a greater number of PVC floors and fewer wood floors were found among the cases, this disparity was due in part to selection bias, and this imbalance did not influence the results of the study. As shown in Table 8, total phthalates were detected at a median concentration of 1,823 ng g⁻¹ (451-4,491 ng g⁻¹) for PVC flooring, which is considerably higher than the 764 ng g⁻¹ (175-1,576 ng g⁻¹) for wood flooring ($p = 0.001$). The median DEHP and DBP concentrations in dust samples were also significantly higher for PVC flooring compared to wood flooring. In case of DEHP, the median concentration for PVC flooring was 1,720 ng g⁻¹, and that for wood flooring was 699 ng g⁻¹, indicating that PVC flooring was highly associated with a higher DEHP level ($p = 0.001$). Regarding DBP, the median concentration in PVC was 22.9 ng g⁻¹, higher than the 9.4 ng g⁻¹ found for wood ($p = 0.012$). The correlation between PVC flooring and the phthalate concentrations in dust was higher for DEHP than for BBP. However, in case of other phthalate esters, there were no significant differences between PVC and woods. This result is in agreement with other results. It is reported that PVC flooring is one of major sources of phthalates in indoor dust [49]. In Korea, PVC flooring is common in most residential or commercial buildings [50].

Interestingly, there was a strong correlation between levels of DEHP and total phthalates and older building construction age. The median DEHP levels were 1,179 ng g⁻¹, 1,690 ng g⁻¹, and 2,194 ng g⁻¹ in DEHP ($p = 0.004$) and 1,253 ng g⁻¹, 1,784 ng g⁻¹, and 2,302 ng g⁻¹ in Σ phthalates ($p = 0.004$) for building construction age less than 10 y, 10-20 y, more than 20 y, respectively. This result implies that phthalate levels increase in older houses. Table 7 also presents that DBP showed a partial correlation, and were 13.9 ng g⁻¹, 21.9 ng g⁻¹, and 34.3 ng g⁻¹ ($p = 0.070$) as building construction age increases. Similar result was obtained by Bornehag et al. [44]. They found associations between their dust concentrations of DEHP and the amount of VC used as flooring and wall material in the home [51]. Furthermore, high concentrations of DEHP were associated with buildings constructed before 1960.

4. Conclusions

We examined the levels of BFRs and phthalate esters in indoor

house dust in Korea. Among PBDE congeners, BDE-209 (deca-BDE) was dominant, followed by BDE-47, BDE-7, and other. We found the ratio of isomers other than BDE-209 detected in dust was not high. In addition, we observed that not only heavy congeners but also light BDE-7 concentrations increased with increasing age of the building. Thus, this might reflect the debromination of BDE-209. The concentrations of Σ HBCDs and TBBPA were lower than reported in other countries. Among HBCD levels, α -HBCD was present at the highest concentrations, followed by γ -HBCD and β -HBCD. The levels of phthalates were higher than those in Europe and the United States. This result is caused by the more frequent use of phthalate sources such as PVC flooring in Korea. Among the phthalates, DEHP was detected at the highest concentrations, followed by DBP and BBP, indicating that DEHP is widely exposed in indoor environment. We also examined the correlation between flooring type (PVC) and building age based on a survey of building characteristics. For PBDE congeners, BDE-47 ($p = 0.062$) and BDE-203 ($p = 0.007$) were significantly correlated with building age. PVC flooring was highly associated with levels of DEHP ($p = 0.001$) and BBP ($p = 0.012$), indicating that PVC flooring is a source of DEHP and BBP, and building construction age was associated with DEHP ($p = 0.004$) and BBP ($p = 0.070$) levels. This result implies that the older buildings can discharge the greater the quantity of phthalate from sources, resulting in higher the exposure to indoor residents.

Our results can increase understanding of the distribution of BFRs and phthalates in indoor dust in houses, and help to provide the basic data for the establishment of guidelines for indoor air quality in Korea.

Supporting Information (Supplementary Materials)

The results of MDLs for phthalates in dust sample are provided in Table S1. Associations of BFR concentrations of the settled dusts with building characteristics in 42 houses is provide in Table S2. A detailed description of the extraction and analysis procedures of BFRs and phthalates is provided in Fig. S1. The chart of debromination of deca-BDE is provided in Fig. S2.

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