

COMMERCIAL PENTA-BDE MIXTURES IN DUST SAMPLES FROM INDOOR ENVIRONMENTS IN LUBLIN, POLAND – A CASE STUDY

AMELIA STASZOWSKA ^{a,*}, BERNARD POŁĘDNIK^a, MARZENNA R. DUDZIŃSKA^a,
JACEK CZERWIŃSKI^b

^a Division of Indoor Environment Engineering, Faculty of Environmental Engineering, Lublin University of Technology,
Nadbystrzycka Str. 40B, 20-618 Lublin, Poland

^b Laboratory of Environmental Analyses, Faculty of Environmental Engineering,
Lublin University of Technology, Poland

*Corresponding author: e-mail: A.Staszowska@fenix.pol.lublin.pl, +48 81 5384 404,
fax: +48 81 5381997

Abstract: Levels and distribution of selected polybrominated diphenyl ether congeners in dust samples taken from different indoor environments in Lublin, South-Eastern Poland, are reported. The most abundant congeners, found in the majority of sampling sites, were BDE 47, 100 and 28, respectively. The highest levels of examined congeners were observed in dust samples taken from a computer repair facility and labs. BDE 47, 99 and 100, occurring at highest levels and having the greatest contribution confirm that in Polish indoor environments polymer products contain Penta-BDE technical mixture.

Keywords: Flame retardants, polybrominated diphenyl ethers (PBDEs), Penta-BDE, dust, indoor air quality

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are a group of synthetic organobromine compounds, widely used for over three decades as flame retardants in combustible materials to increase their fire resistance. There are 209 possible PBDE congeners, but commercial PBDE products are marketed as three technical mixtures of several congeners, labelled as Penta-BDE, Octa-BDE and Deca-BDE. The Penta mixture contains mainly BDE 47, 99, 100 with smaller contributions from BDE 28, 153 and 154; the Octa mixture contains BDE 153, 154, 183 while the Deca mixture consists mainly of BDE 209. Presently, the Penta mixture is used as a flame retardant for polyurethane foams in carpet underlays, vehicle interiors, furniture and beddings. However, this mixture was sometimes also used in computer circuit boards until the mid-1990s. The Octa mixture is added primarily to thermoplastics such as high impact polystyrene. The Deca mixture is used in plastic housings for electrical goods such as TVs and computers, as well as in textiles (Alaee et al., 2003; Alcock et al., 2003).

The application of PBDEs in polymer products results globally in annual savings of thousands of lives, serious personal injuries, and reduces property damage. However, because of their similarities to PCBs and PCDD/Fs (ubiquity in the environment, bioaccumulation, resistance to degradation and presence in human tissue) PBDEs have been identified as “new emerging” environmental pollutants. Recent studies have shown that some lower brominated PBDE congeners containing bromine atoms in position 2,2',4,4', e.g. BDE 47 and BDE 99, may cause many health disorders including neurological and carcinogenic dysregulation (McDonald, 2002; Rudel et al., 2003).

In response to these findings, the commercial mixtures Penta- and Octa-BDE were banned in the European Union in 2004. Unfortunately, such restrictions will not eliminate PBDE releases from products currently in-use or new products manufactured with recycled PBDE-containing materials. Additionally, a potentially important secondary source for lower brominated PBDE congeners may be debromination of Deca-BDE. Products originating from this process show enhanced toxicity and an enhanced ability to accumulate in comparison with the parent compounds (LaGuardia et al., 2006).

To date, PBDEs have been measured in air (Shoeib et al., 2004; Wilford et al., 2005), surface water (Alaee et al., 2003), sewage sludge (de Boer et al., 2003), soil (Cai and Jiang, 2006), sediment (Hale et al., 2006) and in several biological matrices and human tissues (Bi et al., 2006; Domingo, 2004; Lind et al., 2003). The highest PBDE levels were detected in indoor samples, namely in dust, at concentrations which were often 50-fold higher than those reported for the outdoors.

It is well known, that indoor dust can be an important pathway for exposure to a range of contaminants found in the indoor environment such as building materials (e.g. PCBs, lead from old paints) and consumer polymer products. PBDEs can accumulate in dust from various pathways, including vaporizing from products, transport of soil particles and aerosols from the outdoor to the indoor environment (Rudel et al., 2003; Kemmelein et al., 2003). PBDEs may enter the human body by inhalation of suspended particles, ingestion of dust particles (especially children and toddlers) and by direct skin contact (Butte and Heinzow, 2002; Jonez-Otazo et al., 2005; Karlsoon et al., 2007; Wu et al., 2007). Analysis of the PBDEs content in the indoor environment and, more particularly, of house dust is thus of prime importance for assessing human exposure to these toxic chemicals.

This study reports levels and distribution of selected PBDE congeners commonly used in technical Penta-BDEs mixture (BDE 28, 47, 99, 100, 138, 153, 183) in dust samples collected from different indoor environments

MATERIAL AND METHODS

Sample collection

Samples of dust were collected from two category sites: occupied and non-occupied. As occupied sampling sites were chosen three different private households (Room 1, Room 2, Room 3) in Lublin, a city of ca. 400 thousand inhabitants, situated in South-Eastern Poland. Non-occupied sites were two computer student laboratories located at the Lublin University of Technology (Lab 1, Lab 2) and a computer repair facility (Computer Facility), located in the neighborhood of the University. As a reference site was chosen a student classroom at the Lublin University of Technology. Dust samples were collected by vacuum cleaner with a water filter. At the occupied sites dust was sampled from the top of bookshelves and furniture, where it had collected for at least the previous three months.

The first occupied site (Room 1) was a small living room with one window to the west. It was located on the second floor of a 60-years-old three-story building. The room was gravitationally ventilated, had an oak wood floor, wood furniture (varnished birch) and a woolen carpet. Inside the room there was a TV set with a liquid crystal display (LCD) screen. The second site (Room 2) was a bedroom located on the second floor of high, 5 years old apartment building. The room had two outer side walls directed to the northeast and southeast and with a window on the southeast wall. It was ventilated gravitationally, with a varnished hornbeam wood floor which was covered with a woolen carpet with an artificial bottom-side. The room was furnished with a typical veneered chipboard bed and varnished pinewood furniture. There were no electronic appliances. At the third site (Room 3) dust was sampled from a children room, with one window to the east. This room was located on the second floor of a 12 years old five-story building. It was gravitationally

ventilated, appointed with hard birch parquet floor, wood furniture (varnished plywood) and tufted polyamide with a textile backing carpet. Inside the room there was a computer with a cathode ray tube (CRT) monitor and ink printer.

Dust in the computer laboratories and in the facility was sampled from inside computer boxes. Lab 1 was a room located on the second floor with two windows to the east and a ceramic tile floor. Inside the room, except for 25 computers with CRT monitors there were a teacher's desk, tables and seats for 25 students, one blackboard for chalk writing and one whiteboard. Lab 2 was a room located on the third floor with two windows to the east and a ceramic tile floor and walls. Inside the room, except for 25 computers with CRT monitors, was a teacher's desk, tables and seats for 24 students and one blackboard for chalk writing. Similarly to Labs 1 and 2, dust samples from the Computer Repair Facility were collected from at least 30 computers under repair, from inside computer boxes.

As a reference site was chosen a classroom, located on the second floor with two stained dark glass windows directed to the north. The floor was covered by ceramic tiles. The room was equipped with tables and hard chairs only. No electronic appliances or textiles were present inside. Samples were taken from the floor, after a holiday period during which the room was closed for one month for any activities.

Chemicals

All chemicals used for PBDE analysis were of high purity. Toluene and dichloromethane (Ultra-Resi Analysed) were purchased from T.J. Baker (Germany). PBDE analytical mixture standards series BDE-CVS-F containing BDE 28, 47, 85, 99, 100, 153, 154, 183 congeners, as well as mass labeled (^{13}C) PBDE recovery solution BDE-MFX containing BDE 77L and 138L were obtained from Wellington Laboratories (Ontario, Canada). Additional standard solutions of native PBDE (BDE-MXF) containing BDE 28, 47, 85, 99, 100, 153, 183 congeners at a concentration 2000 ng/cm³ were obtained from Wellington Laboratories and from Accustandards (mixture M-527 BDE).

Extraction/clean-up

Samples from the vacuum cleaner (in the form of suspended particles in water solution), were passed through glass filters GF/B (Whatman; 1 μm pore size) after addition of ^{13}C labeled standards (Wellington Laboratories – BDE 77L and BDE 138L) to monitor the BDE recovery. The filters were freeze-dried and then extracted with toluene in a Soxhlet apparatus for 16 hours in a dark room. The extract was evaporated under a nitrogen stream and then the solvent was changed to dichloromethane. Further purification was performed using size exclusion chromatography (SEC) (Breeze 1525, Waters) system equipped with a double pump system, 2000 μL loop, combined 19x150 mm and 19x300 mm Envirogel GPC cleanup columns (Waters, USA), tunable detector UV-VIS Waters M2487 working @ 254 nm, FC III fraction collector (Waters, USA), and system control and data acquisition Breeze 3.30SPA software. Dichloromethane was used as a mobile phase with the flow rate 5 cm³/min. Purified extracts were collected from 11 to 17 minutes.

Sample purification is commonly done on the set of classical chromatography columns – a silica gel column followed by an activated carbon column (de Boer et al., 2003). For the PBDE levels measured in this study, size exclusion chromatography was sufficient. Typically, this method is used to clean-up samples for PAHs analysis (Gevao et al., 2007) but it has also been applied for other compounds e.g. pharmaceuticals (Debska et al., 2004), chlorinated and brominated compounds (Janson et al., 1991), and also for PBDEs (Saito et al., 2004). A graphical scheme of the purification procedure is presented in Figure 1.

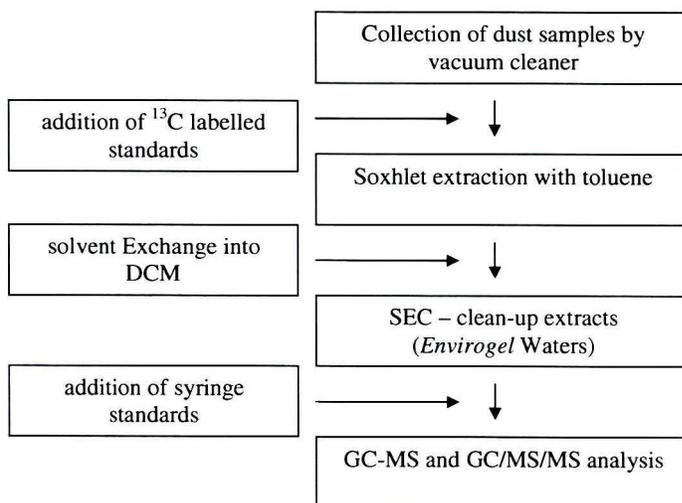


Fig. 1. Scheme of PBDE analysis in dust samples.

Gas chromatography/mass spectrometry (GC/MS)

Chromatographic analysis was done on the Trace Ultra – Polaris Q GC-MS system. The GC-MS technique which is cheaper and enables the measurement of more congeners than LC-MS has recently been used for PBDE analyses by many researchers (de Boer and Cofino, 2002; Hyötylainen and Hartonen, 2002). The parameters of the chromatograph system are presented in Table 1.

Table 1. Conditions of GC-MS system.

The operation conditions of the chromatograph (TRACE ULTRA)	
Injector	PTV (splitless mode) @ 320°C
Capillary column	RTx Dioxin (Restek) 60m x 0.25mm d _i =0.18µm
Oven temperature programming	85°C (0.5 min hold) ramp 15°/min to 210°C then ramp 5°/min to 320°C, 20 min hold
Carrier gas	He (99.9996%) @ 40 cm/s
The MS operating conditions (POLARIS Q)	
The ion source temperature	250°C
The transfer line temperature	275°C
Scanning mode I: Full Scan	50.0 - 850.0 amu
Collision gas	He (99.9996%) @ 2 ml/min
Scanning mode II	SIM: quantitation ion and (qualifiers) Tri-BDE (28) 406 408 (246, 248) Te-BDE (47) 486 (484, 488, 326) Pe-BDE (99, 100) 564 (566, 406, 404) Hx-BDE (138, 153, 154) 643 (645, 484, 482, 186) Hp-BDE (183) 723 (725, 562, 564)

Our research was focused on the Penta mixture, which has been banned in the EU since 2004, therefore 2,4,4'-TriBDE (BDE 28), 2,2',4,4'-TeBDE (BDE 47), 2,2',4,4',6-PeBDE (BDE 99),

2,2',4,4',5-PeBDE (BDE 100), 2,2',4,4',5,5'-HxBDE (BDE 153), 2,2',3,4,4',5'-HxBDE (BDE 138), and 2,2'3,4,4',5',6-HpBDE (BDE 183) were measured. The detection limit for this method was evaluated on 0.5 ng/dm^3 ($S/N \geq 3$) in extracts for PBDE 28 and PBDE 100, and 1.2 ng/dm^3 for PBDE 183. The quantification limit was evaluated on the level $10 \text{ pg / injection}$ ($S/N \geq 5$). Linearity was better than 10-250 for all congeners except BDE-153 (better than 10-200).

Quality assurance/ quality control (QA/QC)

No certified standard reference materials were available for house dust analysis. Therefore for each sample, extraction efficiencies were calculated based on the addition of labeled standards before extraction and at other steps of the analytical procedure. Because of the different character of dust surface and sodium sulfate surface, comparison of recoveries of SDB discs (3M, USA) spiked with native PBDEs was performed instead of application of labeled or native PBDEs onto Na_2SO_4 . 74 to 91% recovery was obtained in triplicate analysis while the relative standard deviation for triplicate analysis of real samples ranged from 12% for BDE 28 to 27% for BDE 181.

Blank runs were performed after each injection of the analyzed sample to prevent "ghost" peaks. The uncertainty, evaluated based on error variance propagation law, was from 17% for BDE 47 and BDE 99 to 31% for BDE 153. Differences in the recoveries of real sample measurements (based on ^{13}C labeled compounds spiking) and from spiked sample were lower than 5%.

RESULTS AND DISCUSSION

Seven tri- through hepta-BDE congeners (BDE 28, 47, 100, 99, 138, 153 and 183) were measured in the dust samples collected from sites including: residential rooms, classroom, computer student labs and computer repair facility in Lublin, Poland.

All analyzed congeners were found in the dust samples, with the exception of BDE 183, which was below the detection limit in all samples. PBDE levels varied with respect to the particular congener and sampling site. A summary of the measured PBDE concentrations observed in this study is presented in Table 2.

Table 2. Concentrations of PBDE congeners measured in dust samples.

Sampling site	PBDE congeners [ng/g]						
	28	47	100	99	138	153	180
Room 1	dl	10	dl	dl	dl	dl	dl
Room 2	dl	dl	dl	dl	dl	dl	dl
Room 3	27	43	10	dl	dl	dl	dl
Classroom	16	41	14	dl	dl	dl	dl
Lab 1	12	48	35	49	21	7	dl
Lab 2	44	91	69	42	22	13	dl
Computer repair facility	78	331	43	157	85	44	dl

dl- sample measurement was below the detection limit

The range of congener concentrations was nd– 331 ng/g dust. The most abundant congeners, found in the majority of sampling sites were BDE 47, 100 and 28, respectively. The highest levels of examined congeners were observed in the dust samples from the Computer Repair Facility, Lab 2, and Lab 1. None of the examined congeners was detected in dust from Room 2, which was somewhat surprising. Levels below the detection limits might be partially explained by the lack of electronic devices in that room.

Only BDE 28, 47 and 99 congeners were found in the samples from occupational sites. PBDE concentrations were in the range nd-43 ng/g for BDE 28, 47 and 99. The highest levels were found in Room 3, where a computer with CRT monitor and printer was located.

In turn, in non-occupied sites (Lab 1 and 2, Computer Repair Facility) PBDE concentrations were higher (nd-331 ng/g) and almost all analyzed congeners (with the exception of BDE 183) were found. In the Lab 2, analyzed congeners were found at approximately double the levels observed for dust samples from Lab 1. The obtained results might be explained by the duration of computer usage in labs during the semester and by the frequency of cleaning intervals of individual computer units (\geq twice per year). Computers from Lab 2 were working longer and were not cleaned as frequently as those in Lab 1.

In the classroom, which was assigned as a reference site, there were no potential indoor PBDE sources. With the intention of avoiding the influence of outdoor PBDE sources, this site was closed for one month prior to removing samples for analysis. Surprisingly for the authors, PBDE levels in this classroom were above detection limits and, in addition, showed the presence of lower brominated BDEs such as 28, 47 and 100. It is difficult to give a convincing explanation of these results.

Our investigation was limited to one city in South Eastern Poland in a less developed, essentially agricultural area. However, our samples were collected at a university, well equipped with audiovisual and electronics and modern buildings (LCD TV, recently furnished, etc.). The PBDE concentrations obtained in this study were lower than those reported in the literature given for comparison in Table 3. There the cited authors collected dust samples in households (Gevao et al., 2006; Ibarra et al., 2006; Stapleton et al. 2005; Suzuki et al., 2006; Wenning et al., 2006) and offices (Ibarra et al., 2006; Suzuki et al., 2006). Moreover, our findings are lower than those presented by Sjödin (2006) for Germany, Australia, UK and US.

Table 3. PBDE concentration range in dust samples in this study compared to other studies.

Authors	PBDE congeners [ng/g]						
	28	47	100	99	138	153	183
This study (Poland)	dl - 27	dl-43	dl-10	dl	dl	dl	dl
Gevao et al., 2006 (Kuwait)	dl-3.47	0.11-65.15	0.01-8.62	0.04-35.74	na	dl-4.36	na
Ibarra et al., 2006 (UK)	dl-2.3	5.2-163	1.1-50	7.4-317	na	1.0-94	na
Stapleton et al., 2005 (US)	4.2-76.5	103-7,610	25.9-2,900	174-13,800	dl-111	11.7-1,510	na
Suzuki et al., 2006 (Japan)	240-67,000	1,100-980,000	160-130,00	180-810,000	na	660-11,000	na
Wenning et al., 2006 (US)	0.9-59	44-2,460	32-1,580	207-7,660	na	21-1,440	na

dl- sample measurement was below the detection limit

na - not analyzed in the study

Differences in PBDE concentrations between our study and other studies may reflect mainly differences on the scale of PBDEs usage. In Poland, PBDEs are not produced and not so widely distributed as inorganic flame retardants. Large amounts of electronic appliances are, however, imported from countries in which PBDEs are added to polymers. In turn, the differences between PBDE levels in household dust could be related to the differences in point sources in the indoor environment (e.g. EE appliances, textiles).

As stated earlier, not all selected PBDE congeners for analysis were found in all dust samples and the distribution of individual congeners varied strongly, as shown in Figure 2.

BDE 28 (substituted with three bromine atoms) was one of the most abundant among the analyzed PBDE congeners. It was found in five of the seven sampling sites. The highest percentage share – 34 %, was obtained for Room 3 and Classroom – 22%, respectively. In turn, BDE 47 (substituted with four bromine atoms) was found in the majority of samples, besides Room 2. In dust collected in Room 1, BDE 47 accounted for 100%. It was also the most abundant congener in Room 3 (54%), Classroom (58%), Lab 1 (30%), Lab 2 (32%) and in the Computer Facility (45%). BDE 100 (substituted with five bromine atoms) was present in highest distribution in the Classroom and in both Labs (20%, 20% and 23%). BDE 99 (also with five bromine atoms in the molecule), 138 and 153 (congeners with six bromine atoms) were detected only in samples from the Labs and Computer Facility. BDE 99 contributed 29% to the all analyzed PBDE congeners in dust from Lab 1, but for Lab 2 it was only 15%.

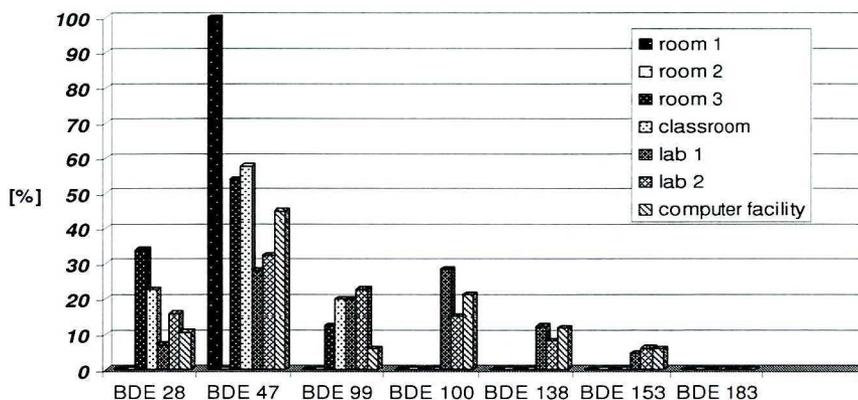


Fig. 2. Percentage distribution of the measured PBDE congeners in dust samples.

In turn, for the Computer Facility, BDE 99 accounted for 21%. BDE 138 and 153 were found at a level of 12% in Lab 1. BDE 153 was the least abundant congener in dust samples, where it was found and accounted for less than 6%.

BDE 47, 99 and 100, occurring at high levels and having the greatest contribution confirm that in Polish indoor environments PBDEs are probably being widely applied. Because the sites studied were equipped with computers, their housings may contain the Octa-BDE or the Deca-BDE mixture (not-analyzed in this work). The latter mixture is used as a flame retardant in computer casings, wires and electrical connectors. The prevalence of BDE 47 and 100 in collected dust samples may result from Deca-BDE transformation caused by specifically indoor factors, e.g. sun- or artificial light. Additionally, it may be significant that the percent distribution of measured PBDE congeners (expressed as a sum percent of BDE 28, 47, 100, 99, 153) is similar to the composition of the technical Penta-BDE mixtures (Sjödin et al., 1998).

CONCLUSIONS

In most of the dust samples examined, congeners typical of the banned Penta-BDE mixtures were found, although their concentrations were extremely low. In only one sample from a residential area, none of the examined congeners was detected. Concentrations in private apartments were lower than in public areas, like laboratories at the university.

In most samples, lower brominated congeners (tri, tetra and penta), which are mainly components of technical Penta-BDE banned in the EU were detected.

The highest concentrations were found for tetra- and penta- congeners in the samples collected in computer laboratories and computer repair facility. They may volatilize from commercial electronic elements produced outside Europe or arise by transformation of higher brominated compounds which are still permitted. Transformation is very probable, as the concentrations of these congeners were significantly lower in samples having experienced exposure to day- and artificial light and collected over longer periods.

Samples were collected at a limited number of sites, but even based on these results, it might be surmised that electronic appliances – probably mainly computers - might be the most important sources of brominated ethers in household dust. Therefore, the potential risk from exposure to PBDE should be measured for employees of computer dismantling facilities, people spending most of their time working with computers, children using computers in their bedrooms (especially when the computer remains in stand-by mode) and children watching TV and playing TV games for several hours a day.

REFERENCES

- [1] Alaei M., Arias P., Sjödin A., Bergman A. 2003. An overview of commercially used brominated flame retardants, their application, their use patterns in different countries/ regions and possible models of release. *Environ. Int.* **29**(6): 683-689.
- [2] Alcock R.E., Sweetman A.J., Prevedouros K., Jones K.C. 2003. Understanding levels and trends of BDE-47 in the UK and North America: an assessment of principal reservoirs and source inputs. *Environ. Int.* **29**(6): 691-698.
- [3] Bi X., Qu W., Sheng G., Zhang W., Mai B., Chen D., Yu L., Fu J. 2006. Polybrominated diphenyl ethers in South China maternal and fetal blood and breast milk. *Environ. Pollut.* **144**(3): 1024-1030.
- [4] Butte W., Heinzow B. 2002. Pollutants in house dust as indicators of indoor contamination. *Rev. Environ. Contam. T.* **175**: 1-46.
- [5] Cai Z., Jiang G. 2006. Determination of polybrominated diphenyl ethers in soil from e-waste recycling site. *Talanta* **70**(1): 88-90.
- [6] de Boer J., Cofino W.P. 2002. First world-wide interlaboratory study on polybrominated diphenylethers (PBDEs). *Chemosphere* **46**(5): 625-633.
- [7] de Boer J., Wester P.G., van der Horst A., Leonards P.E.G. 2003. Polybrominated diphenyl ethers in influents, suspended particulate matter, sediments, sewage treatment plant and effluents and biota from the Netherlands. *Environ. Pollut.* **122**(1):63-74.
- [8] Debska J., Kot-Wasik A., Namiesnik J. 2004. Fate and analysis of pharmaceutical residues in the aquatic environment. *Critical Reviews in Anal. Chem.* **34**(1): 51-67.
- [9] Domingo J. L. 2004. Human exposure to polybrominated diphenyl ethers through the diet. *J. Chromatogr. A* **1054**(1-2): 321-326.
- [10] Gevao B., Al-Bahloul M., Al-Ghadban A.N., Al-Omair A., Ali L., Zafar J., Helaleh M. 2006. House dust as a source of human exposure to polybrominated diphenyl ethers in Kuwait. *Chemosphere* **64**(4): 603-608.
- [11] Gevao B., Al-Bahloul M., Zafar J., Al-Matrouk K., Helaleh M. 2007. Polycyclic aromatic hydrocarbons in indoor air and dust in Kuwait: implications for sources and nondietary human exposure. *Arch. Environ. Contam. Toxicol.* **53**(4): 503-512.
- [12] Ibarra C., Hazrati S., Harrad S., 2006. Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in household dust in Birmingham, UK. *Organohalogen Compd.* **68**: 2151-2154.
- [13] Hale R.C., La Guardia M.J., Harvey E., Gaylor M.O., Mainor T.M. 2006. Brominated flame retardant concentrations and trends in abiotic media. *Chemosphere* **64**(2): 181-186.
- [14] Herzke D., Berger U., Kallenborn R., Nygard T., Vetter W. 2005. Brominated flame retardants and other organobromines in Norwegian predatory bird eggs. *Chemosphere* **61**(3): 441-449.
- [15] Hyötylainen T., Hartonen, K. 2002. Determination of brominated flame retardants in environmental samples. *Trends in Analytical Chemistry* **21**(1): 13-29.
- [16] Jansson B., Andersson R., Asplund L., Bergman A., Litzen K., Nylund K., Reutergårdh L., Seelström U., Uvemo U-B., Wahlberg C., Wideqvist U. 1991. Multiresidue method for the gas-chromatographic analysis of some polychlorinated and polybrominated pollutants in biological samples. *Fresenius' Journal of Analytical Chemistry* **340**: 439-445.
- [17] Jonez-Otazo H., Clarke J.P., Diamond M.L., Archibold I.A., Ferguson G., Harner T., Richardson G.M., Ryan J.J., Wilford B. 2005. Is house dust the missing exposure pathway for PBDEs? An analysis of the urban fate and human exposure to PBDEs. *Environ. Sci. Technol.* **39**(14): 5121-5130.

- [18] Karlsson M., Julander A., van Bavel B., Hardell L. 2007. Levels of brominated flame retardants in blood in relation to levels in household air and dust. *Environ. Int.* **33**(1): 62-69.
- [19] Kemmelein S., Hahn O., Jann O. 2003. Emissions of organophosphate and brominated flame retardants from selected consumer products and building materials. *Atmos. Environ.* **37**(39-40): 5485-5493.
- [20] LaGuardia M.J., Hale R., Harvey E. 2006. Detailed polybrominated diphenyl ether (PBDE) congener composition of the widely used Penta-, Octa-, and Deca-PBDE technical flame retardant mixtures. *Environ. Sci. Technol.* **40**(20): 6247-6254.
- [21] Lind Y., Darnerund P.O., Atuma S., Aune M., Becker W., Bjerselius R., Cnattingius, Glynn A. 2003. Polybrominated diphenyl ethers in breast milk from Uppsala County, Sweden. *Environ. Res.* **93**(2): 186-194.
- [22] McDonald T.A. 2002. A perspective on the potential health risks of PBDEs. *Chemosphere* **46**(5): 745-755.
- [23] Rudel R.A., Camann D.E., Spengler J.D., Korn L.R., Brody J.G., 2003. Phthalates, alkylphenols, pesticides, polybrominated diphenyl ethers and other endocrine disrupting compounds in indoor air and dust. *Environ. Sci. Technol.* **37**(2): 4543-4553.
- [24] Saito K., Sjödin A., Sandau C.D., Davis M.D., Nakazawa H., Matsuki Y., Patterson D.G. Jr. 2004. Development of an accelerated solvent extraction and gel permeation chromatography analytical method for measuring persistent organohalogen compounds in adipose and organ tissue analysis. *Chemosphere* **57**(5): 373-381.
- [25] Shoeib M., Harner T., Ikonoumou M., Kannan K. 2004. Indoor and outdoor air concentrations and phase partitioning of perfluoroalkyl sulfonamides and polybrominated ethers. *Environ. Sci. Technol.* **38**(5): 1313-1320.
- [26] Sjödin A., Jakobsson E., Kierkegaard A., Marsh G., Sellström U. 1998. Gas chromatographic identification and quantification of polybrominated diphenyl ethers in a commercial product, Bromkal 70-5DE. *J. Chromatogr. A* **822**(1): 83-89.
- [27] Sjödin A., Päpke O., Focant J.F., Jones R.S., Pless-Mulloli T., Leontjew-Toms L.M., Herrmann T., Mueller J., Needham L.L., Patterson D.G. 2006. Concentration of polybrominated diphenyl ethers (PBDEs) in household dust from various countries – is dust a major source of human exposure? *Organohalogen Compd.* **68**: 2181-2185.
- [28] Stapleton H.M., Dodder N.G., Offenbergs J.H., Schantz M.M., Wise S.A. 2005. Polybrominated diphenyl ethers in house dust and clothes dryer lint. *Environ. Sci. Technol.* **39**(4): 925-931.
- [29] Suzuki G., Nose K., Takigami H., Takahashi S., Sakai S-I., 2006. PBDEs and PBDD/Fs in house and office dust from Japan. *Organohalogen Compd.* **68**: 1843-1846.
- [30] Wenning R.J., Bock M., Maier M., Luksemburg W.J., 2006. PBDEs, PCDD/Fs, and PCBs in indoor house dust. *Organohalogen Compd.* **68**: 395-398.
- [31] Wilford B.H., Shoeib M., Harner T., Zhu J., Jones K.C. 2005. Polybrominated diphenyl ethers in indoor dust in Ottawa, Canada: Implications for sources and exposure. *Environ. Sci. Technol.* **3**(18): 7027-7035.
- [32] Wu N., Hermann T., Paepke O., Tickner J., Hale R., Harley E., La Guardia M., McClean M., Webster T.F. 2007. Human exposure to PBDEs: Associations of PBDE body burdens with food consumption and house dust concentrations. *Environ. Sci. Technol.* **41**(5): 1584-1589.

Received: September, 2007; accepted: June, 2008.