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Implications of Fire Retardants in Living and Activity Places

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ABSTRACT: *The use of brominated flame retardants has been expanded since the '70s. Nowadays, many of these chemicals are recognized as global pollutants and are associated with impacts to living organisms, including endocrine and thyroid disruptions, immunotoxicity, cancer, and adverse effects on fetal and child development and neurological function. The main commercial forms of brominated flame retardants in plastics and coating materials are mixtures of penta- octa- and deca-brominated diphenylethers. Some retardants have been banned or have gradually voluntarily phased out by manufacturers, because of their environmental persistence and toxicity. The global spreading and propagation of these pollutants is being studied with emphasis in their consistence in indoor environments in residences, offices and other working places as well as in transportation means. The extent of the pollution varies because of the different set of conditions in each microenvironment.*

Keywords: *fire retardants, brominated diphenylether, environmental impact, pollution*

I. INTRODUCTION

Human and ecosystem exposures to persistent organic pollutants occur as a function of compound distribution and time after release across a range of pathways. The early thinking about the exposure to such pollutants was based on industrial and accidental releases leading to exposures such as those of electrical workers to polychlorinated biphenyls from dielectric fluids.

It has been realized that the persistence and bioaccumulation of many pollutants has resulted in their wide spatial distribution in the environment and significant human exposure via the diet. It is now also known that some organic pollutants such as brominated flame retardants, particularly polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane are released from building construction materials, consumer and household goods [1], leading to contamination of domestic and workplace indoor environments [2, 3]. Environmental concentrations of some of these compounds are rising [4], reflecting their increasing use. Given the disproportionate fraction of time spent indoors, such indoor contamination potentially represents a substantial pathway of human exposure.

The production of PBDEs is accomplished through the bromination of diphenyl ether in a solvent (e.g. dibromomethane) in the presence of a Friedel-Craft catalyst, such as $AlCl_3$. Though PBDEs do not naturally occur in the environment, they are widely used brominated flame or fire retardants added to diverse polymers used in a large variety of consumer and industrial products such as electronic and electrical devices, textiles, furniture and plastics to prevent the spread of fires or delay the time of flashover. When heated, they release bromine atoms, which displace the oxygen needed for a fire to burn, generating a very thin protective layer of bromine gas on the surface of the treated product which restricts access of the fire to fuel sources.

The PBDEs group is comprised of a large number of congeners, differentiated by the number and places of bromine atoms on their phenyl rings. The homologue series (mono-, di-, tri-, tetra-, penta-, hexa-, hepta- octa-, nona-diphenylethers) count 3, 12, 24, 42, 46, 42, 24, 12, 3 members, respectively, while the decabromodiphenylether, BDE-209, is the only congener with all free places of the phenyl rings occupied by bromine atoms, thus being the heavier of all congeners [5]. PBDEs present resistance to acids and bases, to reductive or oxidizing compounds, as well as to high temperatures [6, 7].

Polybrominated diphenylethers have been found in many abiotic elements of the environments such as air, soil, sediments, water, sewages and wastes produced by human activities, in modern transportation means and building indoors [8, 9]. The penta-bromodiphenylether congeners are dominating in the atmosphere and aquatic environment [10], while the presence of deca-bromodiphenylether rises in the case of soils, sediments and sludge [8, 10-13].

Ninety percent of penta-BDEs are used in polyurethane foam (furniture), parts of transportation means (upholster, seat cushions), sound isolations, textiles and electronic circuits. Octa-BDEs are mainly used in polymers and rigid plastics (ABS, HIPS).
A list of various resins, polymers, and substrates has been given by WHO [5]. Materials containing polybrominated diphenyl ethers, their major applications and products containing them are presented in Table I.

Table I: Materials containing PBDEs, their applications and products containing them

Resins/polymers/substrates	DeBDE	OBDE	Pe-BDE	Principal applications	Final consumer products
Acrylonitrile-Butadiene-Styrene (ABS)		X		Molded parts	TV-sets/business machines, computer housings, household appliances (hairdryer, curler). automotive parts, electronics, telecommunications
Epoxy-resins (ER) EPOXY	X			Circuit boards, protective coatings	Computers, ship interiors, electronic parts
Phenolic resins (PR)	X		X	Printed circuit boards	Paper laminates/glass pre-impregnated composite fibers for printed circuit boards
Polyacrylonitrile (PAN)	X			Panels, electrical Components	Lighting panels for elevators and rooms, housing of electrical appliances
Polyamide (PA)	X	X		Electrical connectors, automotive interior parts	Computers, connectors, housing in electrical industry, board, electrical connectors, automotive industry, transportation
Polybutylene terephthalate (PBT)	X	X		Electrical connectors and components	Switches, fuse, sw1tch box, computer housings, switchboard electrical connectors, stereos, business machines, military electronics
Polyethylene / Cross-linked polyethylene (PE/XPE)	X			Cross-linked wire and cable, foam tubing, weather protection and moisture barriers	Power cable with cross-linked low density PE, conduit for building with high density PE, portable apparatus building control, instrument, shipboard, automotive, marine appliances, insulation of heating tubes
Polyethylene terephthalate (PET)	X			Electrical Components	Boxes, relays, coils, bobbins
Polypropylene (PP)	X			Conduits, electronic Devices	TV and electronic devices (e.g. yoke, housings, circuit board hangers, conduits); electro-mechanical parts TV, hot waste water pipes, underground junction boxes
Polystyrene / High Impact Polystyrene (PS / HIPS)	X	X		TV cabinets and back covers, electrical appliance housings	TV back panels, computer covers and housings of electrical appliances, office machines, smoke detectors
Polyvinylchloride (PVC)	X		X	Cable sheets	Wire and cables, floor mats, industrial sheets
Polyurethane (PUR)			X	Cushioning materials, packaging, padding	Furniture, sound insulation panels, wood imitations, transportation
Unsaturated (Thermoset) Polyesters (UPE)	X		X	C1rcu1t boards, Coatings	Electrical equipment, coatings for chemical processing plants mouldings, military and marine applications: construction panels
Rubber	X		X	Transportation	Conveyor belts, foamed pipes for insulation
Paints/Lacquers	X		X	Coatings	Marine and industry lacquers for protection of containers
Textiles	X		X	Coatings	Back coatings, impregnation: carpets, automotive seating, furniture in homes and official buildings, aircraft, undergrounds, tents, trains, and military safety clothing

II. THE COMMERCIAL FORM OF PBDEs

The Penta mixture mainly contains BDE 47, 99, 100 and is environmentally persistent. The Octa mixture contains BDE 153, 154, 183 while the Deca comprises mainly of BDE 209 [14]. The Deca- form is a white powder which contains 83% by weight bromine [15]. Is mainly mixed with polymers used in upholster and more condensed plastics for electric electronic appliances, especially TV sets [15, 16].

Penta-BDE is applied in polyurethane foam, mattresses, seat cushions, upholstered furniture, carpet underlay, and bedding [16, 17]. It is also used in epoxy and phenolic resins, polyesters and textile. Octa-BDE is a white powder containing 79% bromine, and comprised mainly of the congener 183 [14]. The octa- form is the less used of the three. It is applied in ABS plastics (PC monitors, housings for televisions, mobile phones, and copy machine parts) [15-17].

In 2009, Penta and Octa mixtures were listed as Persistent Organic Pollutants under the Stockholm Convention. Deca is still in use in North America with the exception of Canada, where its production has been banned. The RoHS directive initially made an exception for deca-brominated diphenylether, but since 2008, it can't be used in electric and electronic applications in EU market. In 2006 polybrominated diphenylethers (tetra- to deca-) were added to the toxic substances list of Canadian Environmental Protection Act (CEPA) [18]. In the same year, China stopped the use of penta- and octa-bromodiphenyl ethers in new electric and electronic products, while Sweden restricted the use of deca-brominated diphenylether in textiles, furniture and some electronics. Deca-brominated diphenylether was banned in Norway in April 1st 2008, with the exception of applications to the transportation sector.

III. PRESENCE OF PBDEs IN DUST - TEST RESULTS

Despite the global regulations, existing PBDE resources are still in use in transportation means, furniture and electric products, where they act as a source of these compounds indoors and then outdoors [19]. Concentrations of PBDEs and their within room and within house variability have been reported [20]. Of special concern is the appearance of PBDEs in the atmosphere and in dust, being responsible for about 80% of human exposure [21].

Table II summarizes concentration values of PBDE congeners measured in the dust of various indoor environments (detached houses, apartments, parliaments, offices, school rooms, public service places, vehicles).

Table II: Polybrominated diphenyl ethers' concentrations in indoor dust from European countries (ng/g)

State, city	**Brominated diphenylether congener**											**□PBDEs**	**Ref**
	28	**47**	**100**	**99**	**85**	**154**	**153**	**183**	**197**	**196**	**209**		
Austria, Vienna – Parliament offices with carpets	3	64	23	72	5.4	9.4	18				510	704.8	[22]
Austria, Vienna – Parliament offices with corridors	2.8	66	15	68		11					340	540	[22]
Belgium, Flanders – 10 offices, average values	2.1	21.1	6.8	45.4		5.5	12.1	23.8	2.3	6.6	443	138	[23]
Belgium, Flanders – 45 residences, average values	0.4	8.1	1.1	8.9		0.9	2.2	1.4	0.9	9.5	313	26.8	[23]
Denmark – 3 residences	3	66	11	<0.1	1.9	1.8	23	11			260	377.7	[24]
Denmark, Copenhagen – Eigtved Pakhus	0.47	21	5	27	1.6	5.2	6.1				470	535	[22]
Denmark, Copenhagen – Parliament	0.91	39	8.3	40		3	8.3				330	430	[22]
Finland – Residence	0.1	9.9	3.5	8.8	1.8	0.8	3.8				100	128.7	[24]
Finland, Helsinki – Parliament south aisle	1.6	180	36	160	7.5	9.4	22				1,100	1,509	[22]
Germany, Berlin – Parliament (Reichstag)	6.9	80	14	50	2.9	6.3	17				1,500	1,677.1	[22]
Germany, S. Bavaria – 34 residences, median values	3.39	23.7	6.47	35.2		3.56	5.02	9.24			354	438	[25]
Germany – 10 residences, median values		<14	<6	10		<6	<6	<6			63	74	[26]
Italia, Rome – Senate	2.8	89	15	59	3.3	5.4	21				6,900	7,095.5	[22]
Italia, Milan – Palazzo Marini	1.5	110	23	170	11.2	9.2	59				4,600		[22]
Sweden, Stockholm – Parliament	0.95	78	19	68	3.1	5	9.8				700	883.85	[22]
Sweden, Stockholm – 10 residences median values	1.3	42		52			6.6	12			320	510	[27]
Sweden, Stockholm – 34 apartments	0.8	37		66			7.8	11			1,100	1,400	[27]
Sweden, Stockholm – 10 offices	1.2	52		92			23	55			780	1,200	[27]
Sweden, Stockholm – 10 care centers	2.8	120		110			12	6.5			580	1,200	[27]
Sweden, Stockholm – 4 vehicles, median values	0.2	7.4		11			3.1	2.2			1,300	1,400	[27]
The Netherlands, Hague – Parliament	<0.16	97	30	130	7.4	11	48				800	1,123.4	[22]
The Netherlands – Internet provider rooms, 3 samples	<0.15	14	3.6	13	0.65	1.24	12.1				360		[22]
UK, West Midlands – 43 schools, care centers, mean	1.4	32	10	54	2.8	5.0	28	5.1	5.6	77	8,500	8,600	[28]
UK – 100 residences mean values	4.14	223	33	287	12,2	16,8	33,8	19,2			9,820	10,449.1	[24]
UK, Scotland – 1 residence	33	1,960	230	2,100	88	110	170	87			5,600		[24]
UK, Birmingham	0.75	20	7	47		5.4	14	64			45,000	45,000	[19]
UK, Birmingham – 2 houses, 112 samples, average	46.4	443.4	139.6	825.7	39.9	88.1	115.9						[29]
UK, Newcastle – 10 residences, median values		22	4	28		3	5	5			10,000	10,000	[26]

In Table III, concentration values of PBDE congeners measured in the dust of various indoor environments in countries outside the European continent. It mast be noted that the sampling methods, the analysis times and the indoor uses are not identical. However, the spread-out of these compounds is shown, even in countries which do not produce but simply use products containing brominated flame retardants.

Table III: Polybrominated diphenyl ethers concentrations in indoor dust from non-European countries (ng/g)

State, city, indoor or outdoor space	**Brominated diphenylether congener**									**□PBDEs**	**Ref**
	28	**47**	**100**	**99**	**85**	**154**	**153**	**183**	**209**		
Australia – 10 residences, mean values		60	100	18		9	13	14	730	1,200	[26]
Australia, Queensland – 5 residences, 3 offices	-	54	15	77	-	7	13	25	619	897/899	[30]
Australia, Brisbane – 10 residences, mean values		91.6	37.5	184.6			23.79	102.38	377.5	904.2	[31]
Canada, Ottawa – 68 residences, mean values	15	1,100	490	1,800	190	380	470	44	1,100	5,700	[32]
Canada,, Toronto – 10 residences, mean values	6.6	300	120	510		69	71	13	670	1,400	[19]
Japan, Hokkaido – Two wooden storey residences	<0.025	2.45	0.54	2.8		0.99	2.01		390	485	[33]
Kuwait – 17 residences	0.6	10.4	1.7	8.9	1.0	1.9	1.8	5.3	202.7	233.6	[16]
New Zealand, Wellington – 20 residences, average values	0.86	36	16	87		8.7	9.8				[19]
Philippines, Manila Observatory, biennial average values	1.19	66.9	7.4	26.3		0.81	0.68	0.31	82.9	134.81	[34]
Singapore – 31 residences, average values	1.2	110	65	340		43	76	18	2,200	2,900	[35]
Taiwan, Kaohsiung & Pingtung County – indoor, mean	7.76	4.97	0.136	0.742		0.415	0.659	1.00	56.8	81.1	[2]
Taiwan, Kaohsiung & Pingtung County – outdoor, mean	0.21	0.595	0.089	0.517		0.509	0.758	1.18	29.9	42.7	[2]
USA – 10 residences, mean values		430	880	150		140	80	70	2,000		[26]
USA – 24 non–smokers residences, mean values		577	220	809							[36]
USA, Amarillo/Austin – 20 residences, mean values	25	810	240	1,400		240	240	28	1,600	4,800	[19]
USA, Boston – 12 samples from 11 residences		0.67	0.17	1.01		0.09	0.11				[37]
USA, Boston – 31 offices in eight buildings	7.5	697	195	915	49.6	115	138	81.2	4,204		[38]
USA, Boston – 108 samples from 20 residences, mean	6.4	337.6	76.9	535.4	19.2	35	47	15.1	1,811	4,269.5	[39]
USA, California – University, campus, median values		3,260	1,041	5,945	117	620	632	75	5,545	23,508	[40]
USA, Massachusetts – 50 residences, mean values	13	543	643	135	33.6	63.2	78.6	20	1,906	4,742	[41]
USA, Michigan – Offices in 10 buildings with various uses	18	1,650	525	3,310	113	182	126	1,270	6,930	15,800	[42]
USA, Dallas – PC, screen and carpet dust	20.3	1,621	429	2,296	96.4	189	199	19.3	8,567	12,136	[43]
USA Washington – 10 residences, mean		1,857	911	2,352	100	156	243	60.4	8,286	13,965.4	[44]
USA Washington – 17 household dryers, average values	20.7	1,220	274	1,700	83.4	156	181	30.7	2,090	5,755.8	[45]

PBDEs were measured in dust in USA, UK, Canada and New Zealand [19]. Concentrations of BDE 209 in two UK samples were the highest ever recorded in a domestic (or office) indoor dust sample. In New Zealand BDE 209 has not been determined. Despite the commercial formulations of PBDEs never having been manufactured in, nor imported into New Zealand, their presence in dusts suggests international trade in PBDE-containing goods is an important pathway effecting their global distribution.

Dust samples from 17 homes were tested for their correlation with total PBDE concentrations [45]. No correlations were observed with year of house construction, type of flooring (i.e., hardwood vs carpet) or the number of television sets or personal computers in the home. However, a significant inverse correlation was observed between the area of the home and the contribution of BDE 209 to the total PBDE concentration in dust. BFR concentrations in floor dust collected from houses, garages, and vehicle interiors varied widely but systematically. In vehicles, concentrations were generally lower than in houses, with the notable exceptions of BDE-208, BDE-207, BDE-206, and especially BDE-209, which were sometimes an order of magnitude higher, indicating extensive use of the deca-formulation in vehicles [46]. Car interior dust reflected the diverse materials, the area sampled (e.g., car seats, carpets), and accumulated dirt. In the garages, BFR compositions resembled that in the houses, although concentrations were about an order of magnitude lower, and lower still for BDE-209. The garage dust samples were highly variable and did not reflect the high abundances of deca-BDEs found in the vehicle interiors, suggesting small contributions from this source.

PBDEs were measured in all samples collected in 17 houses in Kuwait [16], with a mean ΣPBDE concentration ranging between 1 and 393 ng/g. Similar results have been reported in 2011 [47] concerning the PBDE concentrations in the dust of 55 working places in Hong Kong and 23 houses in River Pearl area. There has been no correlation between the concentrations of congeners in domestic dust and the age of the building, between the concentrations of BDE-99 and BDE-47 and the number of furniture containing foam nor between the BDE-209 concentrations and the number of electronic appliances. In Philippines [34], biological and environmental samples collected from fish and from the Manila Metro have been analyzed for PBDEs stressing the need for future studies and directions. Recently published data [40] suggest that the presence of electronics impacts the flame retardant concentrations more in elevated surface dust than in floor dust affecting the true exposure of humans (underestimation).

IV. CONCLUSIONS

All reviewed studies show that the use of chemicals in consumer products leads to the contamination of the indoor environment. The extent of the contamination varies between houses, other built places and transportation means, because of the different set of conditions in each microenvironment. These variations could include differences in the types of products used in each household. In any case, the exposure to dust is an important pathway for human assault by chemicals intended to retard fires.

In many cases the blame of PBDEs is not confirmed, since there is not a good correlation between domestic dust and the residence characteristics, such as the number of electronic appliances, the hours of function etc. However, products containing foam and thermal systems must be more thoroughly examined in view of their role in releasing Brominated diphenyl ethers in indoor environments.

Governments must focus on safer chemicals and materials and be more proactive in pushing green chemistry and sustainable product design. Governments should make the phase out of deca-BDE and all other PBDEs a priority.

Long-term measurements of the factors controlling the atmospheric composition of PBDEs, in conjunction with proper statistical methods, will assure the confrontation of these bio-accumulative pollutants in transportation means, homes and workplaces. The research must be accompanied by correlation studies of phenomena hurting human organs and functions during activities in places with materials emitting brominated diphenyl ethers. Special emphasis must be given to children exposure via dust ingestion.

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