

# Detection of Toxic Flame Retardants in Aquatic and Terrestrial Environment: An Emerging Global Concern

Abioye Fayiga1\*, Mabel Ipinmoroti2

1Soil and Water Science Department, University of Florida, Gainesville, Florida, USA. 2Osun State University, Osogbo, Osun State, Nigeria.

\*Corresponding author. Tel: +2348171143705; E-mail: abioyeg@aol.com

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### **Research Article**

# Abstract

**Background:** Flame retardants are used to delay the onset of fire in products such as furniture, textiles and electrical appliances. But they have been found at elevated concentrations in the global environment and even in remote locations such as Antarctica. They are toxic, persistent and bioaccumulate in the environment. They cause reproductive health problems, nervous disorders, learning problems in children, affect the unborn and infants.

Method and findings: A detailed review of literature was done on Scopus and Science Direct, Pub-Med and Google. Results show that halogenated flame retardants such as PCBs, PBBs, PBDEs, and HBCDs have been banned or restricted in many parts of the world. But despite the ban, they are still being found in high concentrations indicating persistence, absence of regulations or non compliance to regulations. Levels are highest in North America which is consistent with the usage in the continent. Numerous studies have shown that PCBs and PBDEs bioaccumulate and biomagnify in marine mammals, fishes, bivalves, polar bear, birds and human body. Concentrations of PBDE in human milk are highest in North America and lowest in Africa. Even though there is no production of flame retardants in Africa, e-waste recycling is a major source. Levels of PBDEs in indoor dust of Asian countries like China and Korea are the second highest in the world with elevated concentrations in e-waste recycling areas. Replacements for the banned flame retardants such as NBFRs and PFRs have also been detected at high concentrations in the environment. Some of them are as toxic as the replaced ones raising questions about their suitability as alternatives.

**Conclusion:** Toxicological studies are needed before marketing and usage to determine suitable alternative flame retardants.

**Keywords:** Halogenated flame retardants; Phosphate flame retardants; Novel brominated flame retardants; Indoor pollution; Water pollution

# 1. Introduction

Flame retardants are chemicals added to commercial

products such as television, computers, textiles and carpeting, building insulation and furniture to reduce their ignition and flammability thereby providing fire protection [1]. There are four main groups of chemicals used as flame retardants; inorganic salts, phosphorus compounds, nitrogen based compounds, and halogenated organic compounds [2]. Organo-halogenated flame retardants such as Polychlorinated Biphenyls (PCB) produced in 1929 were banned in the late 1970's due to their persistence, bioaccumulation and toxicity in the environment [3-5]. The Stockholm convention signed a global treaty in 2001 that banned and restricted the production, trade and use of Persistent Organic Pollutants (POPs) including PCBs. These polychlorinated compounds were replaced by polybrominated compounds such as Polybrominated Biphenyls (PBBs) which was also later found to be toxic. PBBs were later replaced by other Brominated Flame Retardants (BFRs).

There are three main groups of BFRs which are considered typical or conventional flame retardants: Polybrominateddiphenyl Ether (PBDE), Hexabromocyclododecane (HBCD) and tetrabromobisphenyl A (TBBPA) [6]. PBDE is used in electronics, building materials and textile products; HBCD is the principal flame retardant in polystyrene foam and is also used as thermal isolation in the building industry while TBBPA, the primary flame retardant used in electronic circuits is preferred because it is covalently bonded to the polymer during production thereby reducing migration unlike the others which are additives [7]. This may explain why TBBPA has been found in lower concentrations than PBDE and HBCD in environmental samples [8].

PBDEs are marketed as three commercial mixtures; pentabromodiphenyl ether (penta-(octa-BDE), BDE), octabromodiphenyl ether decabromodiphenyl ether (deca-BDE). These 3 PBDE's have been banned or voluntarily phased out while HBCDs joined the list of restricted chemicals (POPs) at the Stockholm convention in May, 2013 due to their toxicity and persistence in the environment [9-12]. These were replaced by new or Novel BFRs (NBFRs) such as decabromodiphenyl ethane (DBDPE) and 1, 2 bis-2,4,6-tribromophenoxy ethane (BTBPE) [13]. However, some studies have reported



the detection, persistence and toxicity of some NBFRs in the environment [14-16]. Experimental studies in rodents have shown that BFRs have the potential to disrupt endocrine functions by interfering with thyroid systems [17] and may also have adverse effects on the central nervous and reproductive systems in humans [18,19].

Even though flame retardants are not new, there is a growing concern about their toxicity and persistence in the environment. Especially since their occurrence and toxicity has been reported in virtually every continent indicating that flame retardants are a global problem. It is important to know what is happening in all continents since long range transport can distribute these toxic flame retardants everywhere.

Previous reviews on flame retardants have either discussed only one class of flame retardants or levels on a national scale. This paper has compared levels of different classes of flame retardants on a global scale. This paper provides an update on levels of banned and alternative flame retardants in the global aquatic and terrestrial environment, their bioaccumulation and regulation in different countries and continents.

### 2. Flame Retardants in the Americas

#### 2.1 Flame retardants in dust

Flame retardants (Table 1) have been detected, quantified and regulated in North America while there has been little work done in marine environments of South America. They have been detected in various environments including indoor and outdoor air [20-22]; household and office dust [23-26]; sediments [27]; marine environment [28]; and biota indicating their ubiquitous presence in the environment [29-31].

Indoor levels of PBDE vary widely in the United States with the highest concentration detected in California

due to the state's high flammability standards for furniture [32,33]. Concentration of NBFRs were several times lower than concentrations of PBDEs in dust from California homes and fire fighters living quarters correlating with the production, usage and persistence of PBDEs [34]. Bradman et al. [35] reported low airborne but higher levels of PBDE and non PBDE in the dust of a child education facility in California. Their results indicate that furnishings in the facilities were associated with increased penta BDE contamination in the dust.

Dodson et al. [36] reported the detection of 55 flame retardant compounds (in at least one sample) and 41 in at least 50%, including carcinogenic flame retardants such as chlorinated PFRs and "Tris" in household dust samples collected. Concentrations of Firemaster 550 components increased from 2006 to 2011 consistent with its use as a penta-BDE replacement.

Results of a recent study in homes and offices in Canada showed that the concentrations of 10 PBDE congeners and 12 halogenated novel flame retardants in home and office dust were positively correlated with concentrations measured in surface wipes of electronic products suggesting that products with the highest halogenated flame retardants concentrations contribute most to concentrations in dust [26].

It has been shown in Washington State, USA, that flame retardant can be transferred from dust on clothing to the laundry wastewater and subsequently to the aquatic environment [37]. Out of the 21 flame retardant compounds (including PBDEs, HBCDs and TBBPA) detected in house dust (3.6 to 82,700 ng  $g^{-1}$ ), 18 were detected in laundry water (47.1 to 561,000 ng  $L^{-1}$ ).

# 2.2 Bioaccumulation in aquatic and terrestrial environments of the Americas

A study in Canada using European starlings (Sturnus

Class	Name	Abbreviation
Chlorinated flame retardants		
	Polychlorinated biphenyls	PCB
	Dechlorane plus	DP
Brominated flame retardants		BFR
	Polybrominated diphenyl ether	PBDE
	Hexabromocyclododecane	HBCD
	Tetrabromobisphenyl A	TBBPA
Novel brominated flame retardants		NBFR
	Decabromodiphenyl ethane	DBDPE
	1,2, bis-2,4,6,tribromophenoxyethane	BTBPE
Organophosphate flame retardants		PFR
	Tris-(2-chloroisopropyl) phosphate	TCPP
	Tris(chloroethyl)phosphate	TCEP
	Tris(1,3-dichloro-2-propyl)phosphate	TDCPP
	Tris (2-butoxyethyl)phosphate	TBEP
	Triphenyl phosphate	TPP
	Tricresyl phosphate	TCP

**Table 1.** Some common flame retardants in the environment.

*vulgaris*) as bioindicators showed that landfills are an important source of bioaccumulative flame retardants to Canadian terrestrial ecosystems [16]. Manufacturing industries are also a primary source of surface water contamination in North America. Previous studies have reported contamination of sediment from surface water bodies in USA and Canada due to their proximity to BFR or DP manufacturing facilities [38-39].

PCBs were accumulated in large amounts in both marine mammals like stellar sea lions and terrestrial animals such as polar bear (Table 2) despite restrictions on its use. PBDEs were also present in high concentrations in dolphins, stellar sea lions and white croaker in North and South America. The concentrations of flame retardants in aquatic and terrestrial organisms in the Americas (Table 1) show a trend of PCB>>PBDE>>HBCD>TBBPA.

Bioaccumulative behavior of PCBs and PBDEs in a marine food web containing fish, mammals and bird species was compared. Recalcitrant PCB congeners exhibited a high degree of biomagnifications while PBDEs exhibited negligible biomagnifications [40]. However, results of a study on Steller sea lions in Canada later indicated that both PBDEs and PCBs biomagnify in marine food webs and bioaccumulate in Steller sea lions [41].

A study screened PBDEs and non-PBDEs in gull eggs from 25 colonies from the Pacific to Atlantic Canada and found that flame retardants were in the order PBDE>>HBCD>Dechlorane plus (DP) while other screened flame retardants were generally non-detectable and non quantifiable. The study also reported higher concentrations of flame retardants than other parts of the world [42].

PBDEs were determined in adipose tissue of adult and subadult female polar bears sampled between 1999 and 2002 from sub-populations in Arctic Canada, Eastern Greenland, and Svalbard. Results showed similar source regions for PBDE and HBCDs for long range transport to the Arctic and bioaccumulation pathways in the arctic marine food web [43]. Analysis of liver samples of cetaceans in highly industrialized region of Southeast Brazil showed PBDE concentrations that were found to be similar to the Northern hemisphere dolphins. The methoxylated PBDE concentrations in dolphins from Brazil were among the highest detected in cetaceans [44]. Another study in Brazil determined the PBDE concentrations in the blubber of small cetaceans in a coastal area. The pattern of contamination found indicated that penta-PBDEs are a major source of PBDEs to top predators in the Southwest Atlantic coast of Brazil [45].

Whole body homogenates of predatory fish collected from select water bodies across Canada were selected for screening of PFRs to determine the current status of environmental contamination. 6 PFRs out of 15 were detected above quantification limits in at least one individual fish. The low to non detectable PFR concentrations was attributed to degradation [46].

Human exposure to flame retardants may also lead to bioaccumulation in human tissues. Sandanger et al. [47] measured plasma concentrations of selected organobromine compounds and polychlorinated biphenyls in postmenopausal women of Québec, Canada. PBDE-47 was the major PBDE congener, with a mean (geometric) concentration of 8.1 ngg<sup>-1</sup> lipid weight (Iw) and extreme values reaching 1,780 ngg<sup>-1</sup> Iw while the mean concentration of the major PCB congener (PCB-153) ranged from 41.7 ng/g Iw to 177 ngg<sup>-1</sup> Iw. Their result suggest that exposure to PBDE-47 likely occurs through direct contact with the penta-PBDE formulation, whereas exposure to PBDE-153 may originate partly from the food chain.

HBCD was found in placental tissues from Canadian women and results indicated that HBCDs was present in developing fetuses from as early as 6.5 weeks [48]. Human milk samples collected from Canada and Texas were analyzed for BFRs. Milk samples from Canada contained lower amounts of PBDEs in two time periods than those from Texas, USA. HBCD levels in most milk samples were usually less than 1  $\mu$ gL<sup>-1</sup> lw [49]. PBDE levels in blood samples of Inuit

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Country	Biota	Flame Retardant	Concentration	References	
Brazil	Dolphins	PBDE	3-5960 ng/g lw	Dorneles et al., 2010	
Canada	Steller sea lions	PCB	272-14280 µg/kg	Alava et al., 2012	
Canada	Steller sea lions	PBDE	50-3780 µg/kg	Alava et al., 2012	
Canada	Dungeness crab	PBDE	24-1200 ng/g lw	Ikonomou et al., 2006	
East Greenland	Polar bear	PCB	10537 ng/g lw°	McKinney et al., 2011	
East Greenland	Polar bear	PBDE	43.2 ng/g lw °	McKinney et al., 2011	
USA	Shark	TBBPA	0.87 ng/g lw <sup>a</sup>	Johnson-Restrepo et al., 2008	
USA	Shark	HBCD	54.5 ng/g lw <sup>a</sup>	Johnson-Restrepo et al., 2008	
USA	White croaker	PBDE	1670 ng/g lw⁵	Klosterhaus et al., 2012	
USA	White croaker	HBCD	<6 ng/g lw⁵	Klosterhaus et al., 2012	
USA	Fish	PBDE	772 ng/g lw <sup>b</sup>	Xia et al., 2008	
USA	Osprey eggs	PBDE	98-897 ng/g ww <sup>c</sup>	Henny et al., 2009	

 Table 2. Bioaccumulation of flame retardants in aquatic and terrestrial animals in the Americas.

amean; bmedian concentration; cgeometric mean; lw-lipid weight; ww-wet weight



children (mean age 25.2 months) attending childcare centers in Nunavik, Canada were higher than those observed in many children and adolescents around the world but lower than those reported in some U.S. cities [50].

# 2.3 Regulation

Asbestos was used as a flame retardant in the USA in the late 19th century until it was discovered to cause a respiratory disease called asbestosis leading to cancer [4]. The use of asbestos as a flame retardant was discontinued and PCBs were used as from 1929 for five decades in the USA and was banned in the 1970's due to its toxicity and persistence. PCB's were replaced by PBBs but the production of PBB was voluntarily stopped in 1976 after a contamination incident that affected livestock and human health.

Though banned, Shen et al. [51] reported that PCBs were still detected in the North American atmosphere either due to their persistence in the environment or due to non compliance to regulations. PCB concentrations up to 14280  $\mu$ gkg<sup>-1</sup> in stellar sea lions from Canada and 10537 ngg<sup>-1</sup> in polar bear from East Greenland (Table 2) was recently reported decades after it was banned [41,52].

A report by UNEP said that though the open use of PCBs was banned in North America in 1970's, they were still in use in closed electrical systems [53]. However, the ban and restriction of PCBs led to an increased demand for PBDEs as flame retardants. Ivory Gull eggs collected in 1976, 1987 and 2004 from Seynour Island in the Canadian Arctic were analyzed for organochlorines and brominated flame retardants. Concentrations of organochlorines either decreased or showed little changes while concentrations of the PBDEs steadily increased between 1976 and 2004 reflecting the change in flame retardant source [54].

In 2004, the United States phased out the manufacture and import of penta and octa-BDE because of their toxicity and persistence in the environment [55]. Three main manufacturers of deca-BDE in the USA began to voluntarily phase out the export and sale of deca-BDE for certain applications since 2010 while the production, importation and sales of deca-BDE were discontinued in the United States in 2013 [55,56].

Recently, Massachusetts State, USA passed a bill that will ban about 11 flame retardants including penta-, octa-, deca-BDEs; HBCD, TCPP [2-chloroisopropyl) phosphate] and TCEP while Washington DC, USA banned the use of flame retardants such as HBCD, deca-BDE, and TCEP at concentrations above 1000 mgkg<sup>-1</sup> in children's products and home furniture [57].

Several studies have reported decreasing levels of PBDE and increasing levels of alternatives after phase out of PBDEs [52,58-61]. However, Quiros-Alcala et al. [62] have shown that the ban or restriction of toxic flame retardants will not reduce exposure and health effects if the sources (e.g. furniture) are not removed.

This was demonstrated in a study with low income families who have older, deteriorated or poorly manufactured furniture treated with PBDE. Analysis of dust samples from these low income California households showed maximum concentrations of BDE which are the highest reported to date. Likewise, BFR's were measured in offices in Michigan, USA in a study by Batterman et al. [24]. Elevated levels of BFR's (24,300-33,700 ngg<sup>-1</sup>) were found in offices with likely BFR sources like computer servers. BFR's were found in new buildings despite being built after PBDE phase out.

### 3. Flame Retardants in Europe

#### 3.1 Flame retardants in dust

Exposure to dust contaminated with toxic flame retardants constitute a health risk for all living organisms [63]. A contamination pathway from source to dust has been reported for flame retardants in the UK [64]. Even though, persistent flame retardants like PCBs are still being detected in dust in the European environment, previous study has shown that PCB concentrations in house dust was higher in US samples than UK samples [63].

of Concentrations dust in the European follows environment the trend; PBDE>>PFR>HBCD>NBFR>TBBPA (Table 3). Low concentrations of TBBPA may be due to it being covalently bonded and not an additive like the others. Concentrations of flame retardants were higher in cars than offices and homes except for TBBPA [65-67]. Though ingestion of car dust is a significant exposure pathway for BDE 209 (deca BDE), it's a minor contributor to overall exposure for BDE 99, HBCDs and TBBPA [65-68].

The ban and restriction of persistent flame retardants such as PCBs and PBDEs has resulted into increased detection frequencies of alternative flame

**Table 3.** Concentrations of flame retardants in dust in Europe.

	∑PBDE	∑HBCD*	TBBPA*	∑PFR*	DBDPE
Car	340,000 ng/g	13,000 ng/g	2 ng/g	200, 000 ng/g	400 ng/g
Homes	260,000 ng/g	1300 ng/g	62 ng/g	42, 000 ng/g	270 ng/g
Offices	31,000 ng/g	760 ng/g	36 ng/g	NA	170 ng/g
References	Harrad et al., 2008	Abdallah et al., 2008	Abdallah et al., 2008	Brandsma et al., 2014	Harrad et al., 2008

\*median concentrations



Country	Sample	Flame Retardant	Concentration	References
UK	Harbour porpoises	PBDE	54.6-913 µg/kg ww	Law et al., 2013
UK	Harbour porpoises	PFR	6.7-246 µg/kg ww	Papachlimitzou et al., 2015
UK	Otter livers	PBDE	12-17,000 ng/g lw	Pountney et al., 2015
UK	Otter livers	PCB	1.8-140,000ng/g lw	Pountney et al., 2015
Spain	Bottlenose dolphin	PBDE	17.3-1947 ng/g lw	Baron et al., 2015
UK	Fish	TBBPA	0.29-1.7 ng/g lw	Harrad et al., 2009b
UK	Fish	HBCD	14-290 ng/g lw	Harrad et al., 2009b
Denmark	Human placenta	PBDE	0.51-17.1 ng/g lw	Frederiksen et al., 2009
Belgium	Human Liver	PCB	380 ng/g lw	Covaci et al., 2008
Belgium	Human Liver	PBDE	3.6 ng/g lw	Covaci et al., 2008
Norway	Human serum	PBDE	3.6 ng/g lw	Cequier et al., 2015
France	Human serum	TBBPA	20-104 ng/g lw	Cariou et al., 2008
France	Human adipose tissue	TBBPA	ND	Cariou et al., 2008
Belgium	Human adipose tissue	PCB	490 ng/g lw	Covaci et al., 2008
Belgium	Human adipose tissue	PBDE	5.3 ng/g lw	Covaci et al., 2008
Belgium	Chicken eggs	HBCD	8.52 ng/g lw	Covaci et al., 2009
Belgium	Chicken eggs	PBDE	7.77 ng/g lw	Covaci et al., 2009

#### Table 4. Bioaccumulation of flame retardants in Europe.

lw-lipid weight; ww-wet weight; ND-not detected;

retardants such as PFRs and NBFRs [69]. Ali et al. [15] reported detection frequencies between 64-100% for NBFRs in indoor dusts from Belgium and UK. Tetrabromobisphenol A-2, 3, dibromopropyl ether (TBBPA-DBPE) was the major NBFR detected in UK classroom dust while decabromodiphenyl ethane (DBDPE) was the major NBFR in Belgian homes and office dust.

However, Dirtu et al. [70] reported that concentrations of NBFRs were lower than PFRs in Romania dust. The most abundant NBFR was DBDPE while BDE 209 was the most important contributor to the sum of PBDE because it's the most used PBDE formulation (deca-BDE). This is consistent with reports that BDE 209 is the most dominant in UK dust, sediment and water samples [65].

Dirtu et al. [70] showed that PFRs were the dominant flame retardant in Romania dust at concentrations which were higher than PBDEs. The most important contributors to PFRs in Romania dust were TBEP [tri(2-butoxyethyl) phosphate] and TCPP [2-chloroisopropyl) phosphate]. Similarly, Brommer et al. [69] reported that concentration of PFRs in classroom dust in the UK was higher than previously determined concentrations of PBDEs (penta- and octa-BDE). This suggests that PFRs are the main alternatives to the banned PBDEs in Romania and UK.

# 3.2 Bioaccumulation in aquatic and terrestrial environments of Europe

Atmospheric deposition of contaminated dust may also contribute to elevated concentrations of flame retardants in aquatic environments. Several studies have also reported pollution of the European marine and freshwater environments via discharges of wastewater [71,72]. Gorga et al. [7] reported high concentrations of PBDEs (up to 2326 ngg<sup>-1</sup> dry weight-dw), TBBPA (up to 472 ngg<sup>-1</sup> dw), HBCD (up to 97.5 ngg<sup>-1</sup> dw) and alternative BFR like DBDPE with concentration up to 257 ngg<sup>-1</sup> dw in wastewater in Spain. Discharges of untreated industrial and domestic waste into surface waters eventually cause pollution of water, sediment and biota [12,73].

A recent study has reported that an alternative flame retardant, PFR (TCPP) was detected at a much higher concentration (26,050 ngL<sup>-1</sup>) than BDE-209 (295 ngL<sup>-1</sup>) in River Aire, UK near highly populated areas and waste water discharges [74]. Total PBDE concentrations ranging from 1 to 2,645  $\mu$ gkg<sup>-1</sup> and mean concentration of 287  $\mu$ gkg<sup>-1</sup> has been reported in the sediments of Clyde Estuary in UK [75]. In comparison, TBBPA concentrations in the sediments of English lakes were very low with a range of 330 to 3800 pgg<sup>-1</sup> dry weight (dw) while total HBCD concentrations vary from 880 and 4800 pgg<sup>-1</sup> dw [76].

Presence of flame retardants in the marine environment contaminates aquatic organisms (Table 4). PBDEs were still accumulated more than alternatives (PFRs) in marine animals even after the ban (Table 4). The concentrations of PFRs reported were lower than total PBDEs in harbor porpoises and dolphins in the UK [1]. Total PBDE concentrations (Table 4) in dolphins from southern European waters were lower than in harbor porpoises from UK [77]. However, another study reported a decline in total concentration of BDEs in marine mammals from 2003 just before they were banned in Europe [77,78].

A previous study reported high concentrations (Table 4) of PBDE and PCB in Eurasian Otter livers from England and Wales collected between 1995 and 2006 which has been associated with the decline in population of Eurasian Otters indicating their toxicity [79]. Deca-BDE and HBCD have been found to be bioavailable to terrestrial and aquatic birds from

Northern Europe though with little bioaccumulation [80]. The two flame retardants were detected at higher frequencies in sparrow hawks and peregrine falcon eggs.

High bioaccumulation of flame retardants may pollute the food chain for both humans and wildlife especially since it has been reported that dietary exposure is a dominant intake pathway for PBDE in an adult population in Germany [17,81]. For example, PBDE and HBCD have been detected in chicken eggs while TBBPA and HBCD has been reported in fish [11,76].

Exposure to toxic flame retardants through diet and dust inhalation or ingestion may lead to bioaccumulation in humans. A past study has reported PCB concentrations that were much higher than PBDEs concentrations in blood serum of people living in the UK despite restrictions [82]. Similarly, PCB concentrations were about 100 times higher than PBDE concentrations (Table 4) in human adipose tissue and liver in Belgium [83]. Total PBDEs were very low in human serum from France suggesting low usage of PBDEs in France [84]. Concentrations of TBBPA in human milk ranged from 0.06-37.3 ngg<sup>-1</sup> lw while it was not detected in human adipose tissue from France. PBDEs were detected in human placenta in Denmark indicating prenatal exposure to PBDEs [85].

#### 3.3 Regulation of flame retardants in Europe

The Directive on the Restriction of Hazardous Substances (RoHS Directive) (2002/95/EC) in Electrical and Electronic Equipment (EEE) banned producers of EEE containing more than the permitted maximum concentration of hazardous substances such as PBB and PBDE in the European Union (EU) [86]. A new RoHS Directive 2011 (Directive 2011/65/ EU) replaced the previous directives on 21 July 2011 restricting the same hazardous substances but with broader scope and additional obligations [87,88].

Penta- and octa-BDE were banned from use in the EU in 2004 while deca-BDE was banned from use in electrical gadgets in 2008 [59, 88]. The ban created a market for alternatives such as NBFRs and PFRs which have also been detected in the environment [13,89]. PBDE concentrations decreased in human serum of cohabiting UK couples after the ban [88]. PBDE concentrations also decreased in bank voles (Myodesglareolus) in Northern Finland [90].

Early this year, the EU published regulations prohibiting the production, marketing and usage of HBCD [91]. Denmark Environmental Protection Agency conducted a study on PFRs such as TCPP, TCEP and TDCP in kid's products and plans to include its report in assessments by EU for restrictions of these chemicals [92].

# 4. Flame Retardants in Asia

#### 4.1 Flame retardants in dust

China is the largest producer and consumer of flame

retardants in Asia with a total annual production volume of 200,000t which is mostly used in electrical and electronic equipments [16]. This has led to massive environmental contamination in China due to waste disposal problems for these electronic products. E-waste recycling is a major source of contamination in developing countries because they lack the technology, infrastructure and expertise to properly harness their resources [93,94].

High concentrations (227-160,000 ngg<sup>-1</sup>) of PBDEs have been reported in indoor dust in e-waste recycling areas of China while urban areas had lower concentrations (530-44,000 ngg<sup>-1</sup>). Substantial amounts of the alternative DBDPE was dominant amongst NBFRs detected at concentrations ranging from 100-47,000 ngg<sup>-1</sup> in indoor dust from China [95]. Similarly in India, elevated levels of PCBs, PBDEs and DBDPE have been reported in dust from e-waste recycling areas [96]. TBBPA has also been detected in outdoor and indoor dust in the e-waste recycling area of China at concentrations between 1998 (outdoor) and 3435 ngg<sup>-1</sup> dw (indoor) [97].

A recent study in China found high concentrations  $(25 \ \mu gg^{-1})$  of PFRs in indoor dust from rural e-waste recycling area while indoor dust in urban homes and college dormitory rooms had lower concentrations  $(7.48-11 \ \mu gg^{-1})$ . Tricresyl Phosphate (TCP) was the dominant PFR in indoor dust from the e-waste area while TCEP was the dominant PFR in urban homes and college dormitory rooms [98]. The concentrations were lower than what has been reported for Europe and Canada because BFRs are still the major flame retardants in China [98].

This does not agree with the reports that PFRs were found at higher concentrations than PBDEs and NBFRs in office dust in China [99]. BFRs did not change with season while PFRs varied with season; winter > autumn > summer probably due to higher volatilities of PFRs which is sensitive to temperature changes [99]. PFRs were found in higher concentrations in schools than in homes in a study in Japan due to some products (floor polisher/ wax products) being used at the schools. PFR concentrations were a thousand times higher than BFRs [100].

The total concentration of NBFRs in indoor dust of China varied from 6.3-20,000 ngg<sup>-1</sup> with a median concentration of 720 ngg<sup>-1</sup>. The dominant NBFR in indoor dust was DBDPE, followed by HBCD suggesting a shift to greater usage of NBFRs as alternatives to PBDEs [101]. PBDE concentrations in house dust (80-16,000 ngg<sup>-1</sup> dw) in Korea are lower than China suggesting greater usage of flame retardants in China [102].

# 4.2 Bioaccumulation in aquatic and terrestrial environments of Asia

Pollution of marine environments is usually due to discharges of municipal and industrial effluents



from waste water treatment plants. Even though waste water treatment plants removes some of the contaminants, 100% removal is not possible leading to contamination of surface waters and sediment due to discharges of treated and untreated effluents from industries and urban settlements. A recent study has found that removal of PBDEs observed from wastewater only redistributed contaminants from sewage to dewatered sludge. Results showed that a substantial amount (0.38-38g) of PBDEs was discharged into Victoria Harbor from waste water treatment plants in China while 0.17-17g PBDE was disposed to landfill sites in the form of dewatered sludge [103].

Higher concentrations of the alternative DBDPE than BDE 209 reported in sediments of Pearl River Delta in Southern China [16]. It is probably due to national and international restrictions on the use of PBDEs as flame retardants. However total PBDE (17.1-588 ngg<sup>-1</sup> dw) was higher than NBFRs (11.3-454 ngg<sup>-1</sup> dw). PFR concentrations between 8.3-470 ngg<sup>-1</sup> dw have been reported for sediments of the Pearl River Delta in China with high levels in urban and e-waste recycling areas [104]. High concentrations (551-618 ngL<sup>-1</sup>) of an alternative PFR (TCEP) was reported in Yellow Sea and East China Sea in China [105]. Wang et al. [106] reported higher concentrations (9.6-1549 ngL<sup>-1</sup>) of total PFRs in Bohai Sea, China. TCPP and TCEP were the most abundant PFR in the sea water [106].

In Korea, flame retardants such as PBDE (0.55-300 ngg<sup>-1</sup> dw), TBBPA (0.05-150 ng/g dw) and HBCD (0.11-19 ngg<sup>-1</sup> dw) were detected in the sediments of Nakdong River basin [107]. In Japan, HBCDs concentrations up to 7800 ngg<sup>-1</sup> dw were detected in sediments of Kuzuryu River which receives effluents from textile industries which use HBCDs in flame retardant finishes [108]. Pollution of surface water and sediments eventually lead to contamination of aquatic organisms. HBCD concentrations in oyster and mussels in Japan were the highest levels reported from Asia and Europe [109].

Biomagnification and bioaccumulation of PCBs, DechloranePlus (DP), PBDEs and DBDPE has been reported in the mangrove ecosystems of Pearl River Estuary in China. PCB concentrations (32.1-466 ngg<sup>-1</sup> lw) in biota were much higher than PBDE (3.88-59.8 ngg<sup>-1</sup>) which was also higher than DBDPE (not detected-30.6 ngg<sup>-1</sup>) consistent with the strong bioaccumulation potential of PCB [110]. On the contrary, PCB concentrations (23-720 ngg<sup>-1</sup> dw) in Eurasian tree sparrows (Table 5) in China were lower than PBDEs (100-2600 ngg<sup>-1</sup> dw) with the lowest being DBDPE [111]. This was attributed to lower production and consumption of PCB in China. PBDEs and other NBFRs like DBDPE were detected in marine mammals (humpback dolphin and finless porpoise) from the South China Sea. Though PBDEs were dominant, results indicate a shift from PBDEs to the alternatives like DBDPE [112].

Bioaccumulation of PCBs, PBDEs and HBCDs (Table 5) was reported in blue mussels from the coastal waters of Korea [113]. DP has been detected in fish in South Korea at concentrations between 0.61 and 126 ngg<sup>-1</sup> lw [114]. DP, PBDEs and HBCDs have also been detected in marketed fish in Japan though at relatively low concentrations. DP concentrations were the lowest while HBCDs were higher than PBDEs [115]. Similarly, PBDEs have been detected in seafood in Korea at concentrations between 0.06-6.25 ngg<sup>-1</sup> ww [102]. Dominant congeners were BDE 47, 99 and 100.

PFRs have been detected in drinking water in China at concentrations between 85.1-325 ngL-1. The dominant PFRs in drinking water are TCPP, TBEP and TPP (triphenyl phosphate) [116]. Exposure to contaminated seafood, drinking water and dust may result in accumulation of flame retardants in different parts of the human body.

Human serum concentration in China was comparable with European samples but lower than values from North America [117]. Similar to Canada, BDE-47 and BDE-153 were the dominant BDE congener in the serum samples contributing about 60% of total PBDE. PBDEs have been detected in maternal and umbilical cord sera in Korea. Dominant congeners were BDE 47, 99 and 153 [118].

Another study in Korea detected PBDEs in human breast milk (0.9-8.07 ngg<sup>-1</sup> lipid weight) and umbilical cord blood (2.79-85.9 ngg<sup>-1</sup> lw) at concentrations that were comparable to EU and Asian countries but lower than USA [119]. Total PBDE concentrations in human breast milk from Japan (0.01-23 ngg<sup>-1</sup> lw) were higher than Korea [120]. Lower average concentrations (2.1-4.3 ngg<sup>-1</sup> lw) of total PBDEs in breast milk was reported

Country	Sample	∑ PBDE	∑ HBCD	∑РСВ	DBDPE	References	
China	Mud crab	7.66-20.4	NA	32-118	5.38-17.7	Sun et al 2015	
China	Tree sparrow	100-2600	NA	23-270	ND-330	Yu et al 2014	
China	Shrimp	1.13-24.6	ND-14.2	NA	NA	Su et al 2014	
China	Yellow catfish	28.8-97.6	35.2-92.1	NA	NA	Su et al 2014	
Korea	Blue mussels	6.6-440	6-500	17-1000	NA	Ramu et al 2007	
Korea	Crucian carp muscle	NA	4.8-6.6	NA	NA	Jeong et al 2014	
Japan	Fish	2.2-878 pg/g ww	0.02-21.9 ng/g ww	NA	NA	Kakimoto et al 2012	

**Table 5.** Bioaccumulation of flame retardants (ngg<sup>-1</sup> lipid weight) in Asia.

NA-not available; ND- not detected; ww-wet weight



in Japan in 2012. Results indicated that sources of contamination were both anthropogenic and natural [121]. A recent study analyzed PFRs in breast milk from Japan, Vietnam and Philippines and results showed that concentrations were lowest in Vietnam followed by Japan and highest in Philippines [122].

#### 4.3 Regulation

Use of PCBs in dielectric fluid was banned in 1979 while importation of PCB was prohibited in 1984 in Korea [113]. Even though there is a high demand for PBDEs in Korea due to rapid growth of the electronics industry, there is no regulation of PBDEs in Korea [102]. This was not the case for China which developed regulations similar to Europe. China established its Restriction of Hazardous Substances directive (RoHS) by restricting the use of PBDEs to below 1000 mgkg<sup>-1</sup> limit in electrical gadgets [16]. This has led to a decrease in PBDE concentrations in sediments in China, though e-waste dismantling resulted in elevated concentrations of discontinued PBDEs in rural areas [16]. Though deca BDE, dechlorane plus (DP) and DBDPE are still being used in China [110]. China banned the usage of penta and octa BDEs in 2006 as a signatory to the Stockholm Convention [123]. This resulted into decreasing atmospheric PBDE concentrations from 2005-2010 while concentrations of alternative halogenated flame retardants increased from 2008-2013 in China [123]. Li et al. [124] also reported a change in commercial pattern of BFRs after the phase out of PBDEs in China. There was a shift from lower molecular weight to higher molecular weight congeners in China. Concentrations of PBDEs and HBCDs were monitored in China's aquatic environments for four years (2009-2012) after the ban. Results indicate that levels of PBDEs in biota decreased in the first three years but increased during the last year due to biomagnifications through food webs. However, HBCD concentrations were not influenced by the phase out of PBDEs [125].

# 5. Flame Retardants in Oceania

#### 5.1 Flame retardants in dust

Dust has been shown in a study in New Zealand to

be an important human exposure source of PBDEs in the environment. Results of the study showed that concentrations of PBDEs in breast milk were correlated to PBDEs (BDEs 47, 153, 154, 209) in mattress dust and PBDEs (BDEs 47, 183, 206, 209) in floor dust [126]. A similar study reported a correlation between BDE-153 in dust and BDE-183 in breast milk in Australia [127].

PCBs measured in indoor dust from New Zealand were found to be lower than North America but slightly higher than Europe [63]. This suggests historical lower usage of PCB in New Zealand relative to North America. Another study in New Zealand reported that NBFRs such as DBDPE were detected in floor and mattress dust at concentrations similar to Sweden and UK house dust but lower than concentrations in China house dust, UK schools and US homes [128]. They also found that concentrations of HBCDs in New Zealand dust which ranged from 20-4100 ng/g, was lower than concentrations in UK but similar to USA. This suggests that HBCDs were not used as alternatives to PBDEs in the region.

Low concentrations of PBDEs were reported in dust from offices (583-3070 ngg<sup>-1</sup>) and homes (87-733 ngg<sup>-1</sup>) in Australia [129]. The results of a more recent study however showed high PBDE concentrations ranging from 60.4-82400 ngg<sup>-1</sup> with median 571 ngg<sup>-1</sup> in residential dust of Australia [130]. The median concentration is higher than what is reported for Europe but about ten times lower than North America and Asia.

# 5.2 Bioaccumulation in aquatic and terrestrial environments

PBDEs have been detected in marine animals in Australia at concentrations between 6.3-130 ng/g in marine megafauna and 7-64 ngg<sup>-1</sup> in seafood [131]. Lal et al. [132] also detected PBDEs and PCBs in eel and shellfish in Fiji at concentrations (Table 6) below the EU recommended maximum residue limits in food. BDE 47 and BDE 209 were dominant in shellfish probably from combustion sources.

A previous study investigated the biomagnification potential of PBDEs in invertebrates and fishes from

Country	Sample	Flame retardant	Concentration	References		
Australia	Green turtle	PBDE	6.3	Hermanussen et al. 2008		
	Degong	PBDE	1.4			
	Mud crab	PBDE	64			
	Prawn	PBDE	43			
Fiji	Eel	PCB	36.9	Lal et al. 2014		
	Mussels	PCB	0.1-5.89			
	Clam	PCB	0.1-0.28			
Australia	Fanbelly	PBDE	115	Losada et al. 2009		
	Tailor	PBDE	107			
	Mudcrab	PCB	35 pg/g	Matthews et al. 2008		
	Tailor	PCB	53 pg/g			

 Table 6. Bioaccumulation of flame retardants (ng/g lw) in Oceania.

Sydney harbor, Australia. They detected PBDEs in aquatic animals at concentrations ranging from 6.4-115 ngg<sup>-1</sup> lw suggesting bioaccumulation in the aquatic food chain [133]. They also found that BDE 47 was the dominant congener. Matthews et al. [134] reported elevated levels of PCBs in seafood species from Moreton Bay, Australia even though the toxic equivalent concentrations were below EU maximum limits. Food chain contamination exposes the human population to toxic effects of these chemicals.

Concentrations of total PBDEs in human blood serum was lower (7.17 ngg<sup>-1</sup>) in New Zealand than North America but similar to Europe [135]. A similar study reported PBDE concentrations ranging from 2.44-258 ngg<sup>-1</sup> with median 9.97 ngg<sup>-1</sup> in plasma of pregnant women in Western Australia [136] PBDE concentrations in breast milk in Australia was lower than from North America but higher than Europe and Asia [137].

# 5.3 Regulation

New Zealand ratified the Stockholm Convention in 2004 and 2009 which regulated persistent organic pollutants (POPs) including PCBs. This appears to be effective because concentrations of PCBs in breast milk are 54% lower than 10 years ago [138]. This was not the case in Australia where total PBDE concentrations (especially BDE 209) of sediments of Sydney estuary have not decreased since their ban [139]. Concentrations of HBCDs and PBDEs (especially BDE-209) increased from 1980-2014 in Sydney estuary, Australia. Australia has not ratified the Stockholm Convention since 2004 but the regulatory body for industrial chemicals for the Australian Government (National Industrial Chemicals Notification and Assessment Scheme, NICNAS) banned penta and octa BDEs in 2007. However, deca-BDE is still being imported and used in Australia [130].

# 6. Flame Retardants in Africa

# 6.1 Flame retardants in dust

There are few studies on flame retardants in Africa which indicates lack of attention to environmental health. It could also be due to poor economic status of most African countries. However among the few studies, PBDEs and non PBDE flame retardants have been detected in dust from homes, offices and cars in several countries in Africa including Egypt, Nigeria and South Africa [140-142]. In Egypt, average concentrations of PBDEs in dust from offices (14, 993 ngg<sup>-1</sup>) were the highest, followed by cars (6943 ngg<sup>-1</sup>) and the homes (248 ngg<sup>-1</sup>) were the lowest [140].

Nevertheless, these values are substantially lower than from developed countries in North America and Europe. There was a positive correlation between PBDE concentrations in dust and number of electronic items in homes and offices in Nigeria suggesting that electrical gadgets are important sources [141]. The levels of flame retardants in Africa homes are low probably because electronic items are not affordable for the poor population. E-waste recycling is another source of contamination since Africa is basically a dumping ground for used electrical equipments.

Large amounts of used electronic equipment with BFR concentrations above the RoHS limit have been exported to developing countries including Nigeria, a major importer in Africa [143]. Sindiku et al. [143] reported that 61% of the cathode ray tube (CRT) casings in televisions and computer monitors in waste dumps in Nigeria contained more than 10,000 ppm bromine from BFRs which has led to contamination at dumpsites and landfills.

# 6.2 Bioaccumulation in aquatic and terrestrial environments

previous study reported high average А concentrations of PBDEs in landfill leachates in South Africa which ranged from 8392-54751 pgL-<sup>1</sup> [144]. This is a concern because it can leach and pollute the groundwater below. Alternative flame retardants such as NBFRs have also been detected in sediments (below detection to 310 ngg-1) and leachates (below detection to142 pgL<sup>-1</sup>) at landfills in South Africa. Detection frequencies were higher in leachates (75-100%) than sediments (20-50%) of municipal solid waste landfill [145].

Landfills are also a source of contamination to birds such as African sacred ibis who feed on dumping sites. PBDEs were detected in nine bird species studied while HBCDs were detected in only four bird species. Highest PBDE and HBCD concentrations (Table 7) were found in the African sacred ibis in South Africa. PBDEs correlated with PCBs indicating similar sources [146].

Discharges into rivers from wastewater treatment plants (WWTP) are also another source of contamination in Africa [147]. A previous study reported the highest

Country	Sample	Flame retardant	Concentration	References
Antarctica	Seal blubber	PCB	1058	Schiavone et al. 2009
	Seal blubber	PBDE	11	
	South polar skua	PBDE	146	Yogui et al. 2009b
	Gentoo penguin	PBDE	8.12	
South Africa	African penguin eggs	PCB	42-64	Bouwman et al. 2015
	African penguin eggs	PBDE	0.31-0.34	
	Sacred ibis B	PBDE	396	Polder et al. 2008
	Sacred ibis B	HBCD	71	

 Table 7. Bioaccumulation of flame retardants (ng/g lw) in Antarctica and Africa.

concentrations of these pollutants from discharge into the Vaal River, South Africa. Significant concentrations of BFRs like PBB, PBDE, and HBCDs were detected in the river water, sediment and fish [148]. Extremely high  $\Sigma$ PBDE concentrations ranging from 1850 to 25 400 ngg<sup>-1</sup> (median, 3240 ngg<sup>-1</sup>) were also reported in Durban Bay sediments, South Africa which is similar to polluted Pearl River Delta, China [149]. This may be due to weak regulations and noncompliance with environmental laws.

Total PBDE concentrations of sewage sludge samples from WWTP in Cape Town, South Africa ranged between 13.1 and 652 ngg<sup>-1</sup> dry weight (dw). The reuse of the sewage sludge for agricultural purposes could enhance the possibility of these contaminants entering into the food chain [150]. Humans and animals are exposed to these organic contaminants through dust ingestion, inhalation of polluted air and diet,

Asante et al. [151] analyzed cow milk samples from rural and urban locations in Ghana. PCB concentrations in cow milk from Ghana were higher than PBDE concentrations ranging between 2.5-87 ngg<sup>-1</sup> lw (mean, 27 ngg<sup>-1</sup> lw) in urban areas and from 2.1-45 ngg<sup>-1</sup> lw (mean, 14 ngg<sup>-1</sup> lw) in rural areas. Total PBDE concentrations in cow milk from Ghana ranged from 0.47-11 ngg<sup>-1</sup> lw (mean, 2.3 ngg<sup>-1</sup> lw) in urban locations and from 0.047-2.8 ngg<sup>-1</sup> lw (mean, 1 ngg<sup>-1</sup> lw) in rural areas. HBCDs were not detected in cow milk samples from Ghana.

However, PCBs, PBDEs and HBCDs were detected in human milk from Ghana [152]. The average concentrations of total PCB (62 ngg<sup>-1</sup> lw) and PBDE (45 ngg<sup>-1</sup> lw) concentrations in human milk were higher than what was obtained from cow milk samples from Ghana suggesting higher exposure of humans to flame retardants. Total PBDEs in human milk from Ghana was higher than some countries in Asia and Europe but lower than North America while HBCD concentrations (mean, 0.54 ngg<sup>-1</sup> lw) were lower than Japan, USA and Belgium [152]. Another study in South Africa reported relatively low total PBDEs (1.7 ngg<sup>-1</sup> lw) but similar total HBCDs (0.55 ngg<sup>-1</sup> lw) in human milk [153].

# 6.3 Regulation

Even though most countries in Africa may have signed the Stockholm Convention, there is relatively little or no data on regulation of flame retardants in African countries. Only one African country (Ghana) was reported to have signed and adopted the Stockholm Convention in 2001 and ratified it in 2003 [152]. But the international restrictions on these persistent chemicals seem to have significant effect in Africa. A four-fold decline in total PCB concentrations of penguin eggs in South Africa has been reported after 30 years [154].

# 7. Flame Retardants in Antarctica

The Antarctic continent is vulnerable to contamination

by POPs such as PCBs and PBDEs due to the ability of these chemicals to undergo long range atmospheric transport and condense in cold climates despite its remoteness [155]. DP was detected in the remote areas of the Arctic and Antarctica suggesting that DP is susceptible to long-range atmospheric transport [156].

Low concentrations of PBDEs were also detected in lichens and mosses living on King George Island, maritime Antarctica due most probably to absorption of PBDEs directly from the atmosphere [157]. Lichen contamination on King George Island, maritime Antarctica was positively correlated with local precipitation suggesting that wet deposition may play a role in bioaccumulation of PBDEs [158].

A biomonitoring study using seal blubber from Antarctica reported PCB concentrations (Table 7) ranging from 106-3660 ngg<sup>-1</sup> with an average of 1058 ngg<sup>-1</sup> while PBDE had a concentration of 11 ngg<sup>-1</sup> [159]. This shows higher usage, bioaccumulation, and persistence of PCB relative to PBDEs. Another biomonitoring study using Trematomusbernacchii showed that PCB concentrations decreased from 1980-2010 in circumantarctic seawater. PBDE concentrations increased up to 2005 and then decreased from 2005-2011 probably due to restricrions on the use of PBDEs [160].

Research stations have been identified as one of the main sources of HBCD contamination in Antarctica [161]. Antarctica does not have permanent human residents but there are 30 countries with 80 research stations across the continent which produces wastewater discharged into surface waters. High concentrations of HBCDs were found in indoor dust (109-226 ngg<sup>-1</sup> dw), sewage sludge (45-69 ngg<sup>-1</sup> dw), sediments (2350 ngg<sup>-1</sup>) and aquatic biota (554 ngg<sup>-1</sup> lw) in Antarctica [161-181].

The presence of HBCD in Antarctica suggests it was used as a replacement for the banned PBDEs. A recent study reported other alternatives detected in Antarctica. Fresh snow samples were collected from East Antarctica and analyzed for PFRs. PFRs were detected showing evidences for long range transport of alternative flame retardants such as PFRs in Antarctica. Among the PFRs detected, TCEP had the most frequent quantification [16].

# 8. Global PBDE levels in the Environment

PBDE was studied in about 21 out of the 29 countries selected for this paper while NBFR was the lowest studied (Figure 1). Though PBDEs are often associated with soils and sediments, air and water particulate phases are also important transport media for their dispersion on local, regional and global scales [28]. Table 8 shows levels of PBDEs in dust, sediment, wastewater and human milk from different countries representing different continents in the world.



Country	Dust	Sediment	Human milk	Waste water/Sewage sludge
USA	7957 ng/g Bradman et al.,	49 ng/g dw	56 μg/kg lw	1530-2120 μg/kg
	2014	Yun et al., 2008	Ryan & Rawn, 2014	Hale et al., 2012
Canada	5000 ng/g	1160-1610 ng/g dw	48 µg/kg lw	29-1000 ng/L
	Shoeib et al., 2012	Law et al., 2006	Ryan & Rawn, 2014	Kim et al., 2013
UK	37-540 ng/g Muenhor & Harrad 2012	287 µg/kg Vane et al., 2010	1.3-21 ng/g lw Bramwell et al., 2014	NA
Germany	386 ng/g	77-205 pg/g dw	1.9-2.03 ng/g lw	97.1-2217 ng/g dw
	Fromme et al., 2009	Suhring et al., 2016	Raab et al., 2008	Knoth et al., 2007
China	3520 ng/g	17.1-588 ng/g dw	1.5-17 ng/g	1-254 ng/L
	Zhu et al., 2015	Chen et al., 2013	Bi et al., 2006	Deng et al., 2015
Korea	4623-6650 ng/g	27.8 ng/g dw	2.73 ng/g lw	298-48000 ng/g dw
	Lim et al., 2014	Moon et al., 2007	Lee et al., 2013b	Lee et al., 2014
Australia	87-733 ng/g Toms et al., 2009b	0.65-2.5 ng/g dw Drage et al., 2015	11.1 ng/g lw Toms et al., 2007	NA
South Africa	NA	10-24 ng/g ww Chokwe et al., 2015	1.3 ng/g lw Darnerud et al., 2011	13.1-652 ng/g dw Daso et al., 2012

lw-lipid weight; dw-dry weight; ww-wet weight

#### Table 8. Global levels of PBDE in the environment.

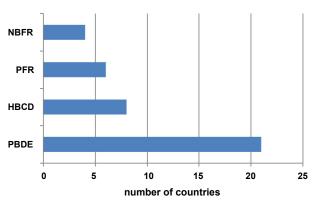


Figure 1. Number of countries studying flame retardant.

PBDE concentrations in indoor dust are highest in North America (Table 8) and follows the trend; USA > Korea > China> Australia> UK/Germany. This may be due to flammability standards for furniture and electrical appliances in USA. Low concentrations of PBDE in dust from Europe suggest low usage and strict regulations in the continent. However, PBDE concentrations in USA sediments are relatively low though Australia has the lowest concentrations followed by South Africa.

Regulations and enforcement of environmental laws may be responsible for the low PBDE concentrations in USA sediments. It may be difficult for the USA to regulate indoor pollution by flame retardants compared with regulations on discharge of effluents into surface waters. UK and Canada had the two highest PBDE sediment concentrations suggesting high usage and uncontrolled discharge of polluted effluents into surface water in these countries.

South Africa had the lowest PBDE concentration in human milk while Germany had the second lowest consistent with concentrations in other environmental samples. North America had extremely high concentrations which was the highest on a global scale reflecting higher exposure of humans to toxic flame retardants in the region despite the ban and restrictions. Higher exposure and bioaccumulation in humans may also lead to higher concentrations in domestic sewage which eventually leads to higher concentrations in waste water and sewage sludge.

South Africa had lowest PBDE concentrations (Table 8) while USA and Korea had extremely high concentrations in the sewage sludge from waste water treatment plants reflecting higher usage and disposal in North America and Asia. Waste water from Canada had higher PBDE concentrations than China. These are potential sources of PBDE in the environment.

#### 9. Research Gaps

There are very few studies on flame retardants in Africa, so more work is needed in this region. There is specifically paucity of data on bioaccumulation of flame retardants in marine environments in Africa. Even though few studies have shown that PBDE concentrations are low in Africa, there is still the need to address the e-waste recycling problem which is the major source of these compounds in Africa. Similarly, the few countries in Oceania seem to have very low levels on a global scale but it was difficult to find studies determining PBDE concentrations in waste water and sewage sludge from waste water treatment plants. This was the same observation in UK though Germany has researched extensively in this area. There are lots of studies on flame retardants in human tissues from Europe which was lacking in North and South America. We only came across one study from Africa (Ghana) that determined flame retardants in cow milk which is very commonly consumed in developed countries. This was lacking in other continents though human milk is very



extensively studied in developed countries. There are clear and sound regulations for flame retardant use in developed countries which is lacking in developing continents such as Asia and Africa. This may be due to financial and institutional constraints.

#### 10. Conclusion

Flame retardants have been detected in every continent including remote regions in Antarctica providing evidence for long range transport of these toxic chemicals. Despite the ban and restrictions, PCB and PBDEs are still being detected at high concentrations in the environment. High levels have been reported in dust from homes, offices and cars; water, sediment and aquatic organisms; terrestrial animals and human tissues showing that these chemicals are ubiquitous and persistent in the environment. However, several studies in North America have reported decreasing trends in the levels of these chemicals. The regulation of these chemicals is a big challenge because it's difficult to find alternatives without similar toxicity, persistent and bioaccumulative properties. Alternative flame retardants such as NBFRs and PFRs are also now being detected at elevated concentrations in the environment. Studies assessing the effects of these alternatives on environmental health are few but necessary especially in North America and Europe. There is a need to monitor the usage of these toxic chemicals in developing countries that are not doing so currently in order to reduce their levels in the global environment.

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