PBDEs and PCBs in European occupational environments and their health effects

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Abstract

Flame retardants such as polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) have been widely used in numerous applications for the retardation of fires. In this effort the indoor air concentrations of PBDEs and PCBs in European environments, obtained from various research studies, are gathered, analysed and evaluated. Specific micro-environments and materials used in indoor buildings appear to influence the concentration of flame retardants. Even though PBDEs and PCBs in Europe were found at low concentrations, there are some indoor environments presenting elevated levels of halogenated flame retardants (HFRs). Congener PBDE 209 is the most abundant in every studied environment. The extensive use of electrical devices increases the PBDEs concentration. High PBDE and PCB concentrations were found in the UK due to the strict fire regulations in this country. High PCB concentrations in indoor air were detected in buildings reinforced with concrete, as well as in schools, industrial and public buildings and in recycling plants. HFRs have shown that they are linked with various diseases including cancer, immune, neurological, endocrine, and reproductive effects and chlorance. Limitation and/or banning of HFRs is ongoing by many organisations and countries and the need for a universal approach is required.
Keywords: halogenated flame retardants, PBDEs, PCBs, emission sources, indoor air concentrations, health effects.

1 Introduction

Since the 1960s, flame retardants have been used as additives in products to reduce the danger of fire and consequently the risk of life when fires occur in

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various indoor environments, where people spend most of their time. The annual world production of flame retardants is approximately 600,000 metric tons, of which about 60,000 are PCBs and 150,000 PBDEs (Darnerud *et al* [1]). HFRs are added to compartments in cars, trucks, trains and airplanes. Flame retardants are used in products such as insulating materials, electronic and electrical goods, upholstered furniture, textiles, sealants, plastics, building materials and carpets. However, there are potential risks from the toxicity and eco-toxicity of the usage of flame retardants. During environmental analyses, flame retardants were present in a wide range of environmental samples, including marine biota, mother's milk and sediments. (Kemmlein *et al* [2], McDonald [3])

As of 1st of July 2006 Directive 2002/95/EC (European Commission, 2003) (the "RoHS" Directive) restricts the use of flame retardant chemicals belonging to the group of PBDEs in electrical and electronic equipment. The maximum tolerated concentration for these chemicals is 0.1%, as set in Commission Decision 2005/618/EC. Certain applications, materials and components can be exempted from the restrictions if their elimination or substitution via design changes or materials and components is technically impracticable, or if the negative environmental, health and/or consumer safety impacts caused by substitution outweigh the environmental, health and/or consumer safety benefits thereof. Even though DecaBDE is no longer produced in the EU, 7,600 tonnes of it are imported each year in addition to 1,300 tonnes that are included in articles (Pakalin *et al* [4]).

PCBs have been domestically manufactured since 1929 but because of possible health implications and environmental impacts, their use and production were severely restricted in many countries. Sweden and UK restricted their use and production in 1972, the USA in 1977, Norway in 1980, Finland in 1985, and Denmark in 1986 ([5]). Due to their non-flammability, chemical stability, high boiling point, and electrical insulating properties, PCBs were used in numerous industrial and commercial applications. Japan banned the production and use of PCBs in 1972 (WHO [6]). The 24 countries of the Organization for Economic Co-operation and Development (OECD) adopted a Decision in 1973, limiting the use of PCBs to certain specific applications and asking for the control of the manufacture, import, and export of bulk PCBs, for adequate waste treatment and for a special labelling system for PCBs and PCB-containing products [7].

PCBs have been classified in the European Union as R33, N; R50-53 [8]. R33 has been assigned as danger of cumulative effects. When a preparation contains at least one substance assigned the phrase R33, the label of the preparation must carry the wording of this phrase as set out in Annex III to Directive 67/548/EEC, when the concentration of this substance present in the preparation is equal to or higher than 1%, unless different values are set in Annex I to Directive 67/548/EEC. In addition R50 has been assigned as very toxic to aquatic organisms and R53 that may cause long-term adverse effects in the aquatic environment [9].

Even though flame retardants are subjects to several restrictions in use they can still be found in a wide-range of indoor environments in the EU. In the

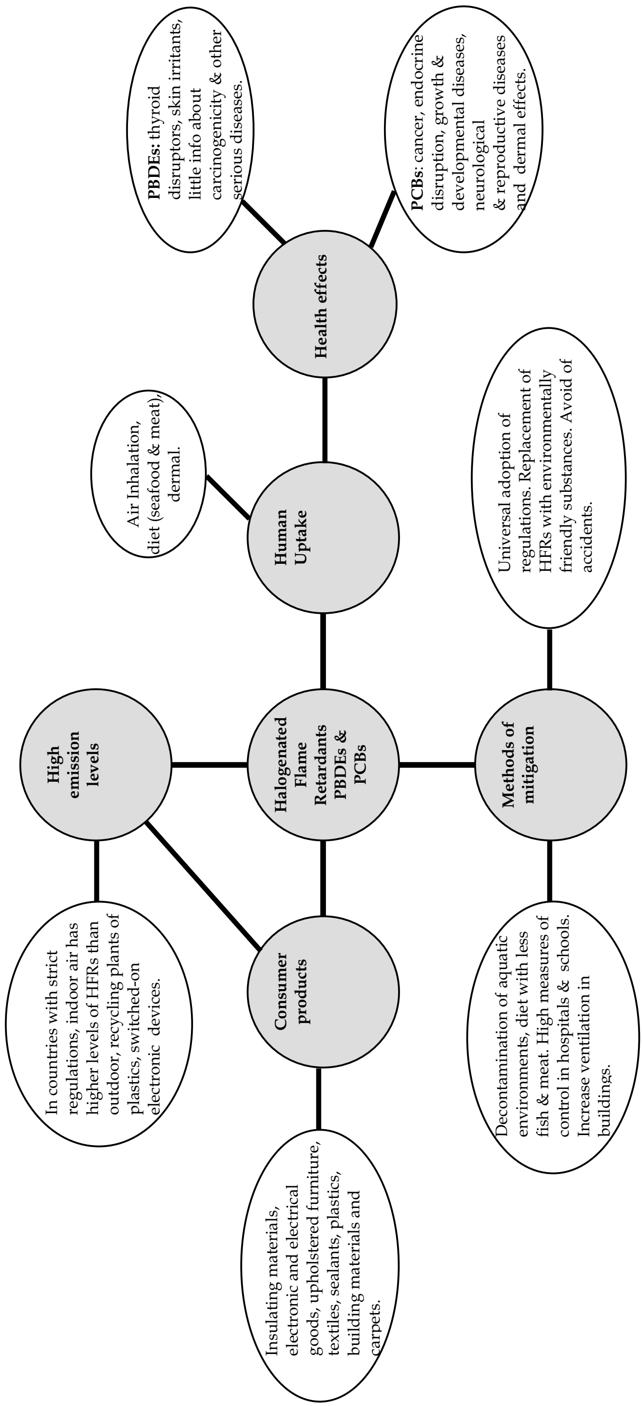

Figure 1: Graph summarising the current knowledge on the HFRs given in this paper.

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present work, a study of the concentrations of HFRs in indoor air in Europe, their possible sources as well as their effects of those in human health is presented. A summary - histogram prior to analysis of the results and conclusions is illustrated in Figure 1.

2 Methodology

Firstly, a literature review was acquired. Studies that included indoor air measurements of PBDEs and PCBs in Europe were collected and their results were analysed. The data contained the specific congener, the sum of many congeners, the values of the measurements, the environment where the measurements took place, the literature reference of which the data were derived, the location and the number of the measurements. Then, the data were compared between each other in the same location to determine possible differences from various products and detect potential emission sources. Afterwards, the data obtained from different literature sources and thus different measurements were compared to distinguish variations between locations and possible trends of flame retardants concentrations in the region of Europe were investigated. In addition, the PBDEs and PCBs indoor air concentrations were used as indications of human exposure and possible health effects. This was done by comparing the measured data with toxicological studies and threshold limit values describing the connectivity of various flame retardants levels to certain disease or generally ill health. Those toxicological data were obtained from the literature and described later on. Finally, conclusions were completed on the current presence of PBDEs and PCBs in European indoor air and channels for mitigation of their health effects were proposed.

3 Results and discussion

PBDE congeners in different indoor locations were observed in the UK and Belgium (Harrad *et al* [10]). Samples taken from the University of Birmingham seem to contain high amount of congeners of PBDE in dust compared to the same congeners found in other cities in UK. In addition, the PBDE 209 congener found in all indoor locations was the highest among the other congeners for both the UK and Belgium. Its value was exceptionally high (mean values range: 2,177 – 45,000 ng/g & maximum: 520,000 ng/g). The reason for this is its high use during manufacturing of products since it has high stability and low-degree-burning properties. PBDE 209 is a DecaBDE and possesses these aforementioned properties due to its fully-brominated diphenyl ether (Darnerud *et al* [1]).

The content of brominated hydrocarbon flame retardants in air samples from a plant recycling electronics goods, a factory assembling printed circuit boards, a computer repair facility, offices equipped with computers and outdoor air have been measured (Sjodin *et al* [11]). Elevated values of the higher PBDEs, i.e. PBDE 183 and 209 as well as of 1,2-bis(2,4,6-tribromophenoxy)-ethane and tetrabromobisphenol A were detected in all the studied areas in the recycling plant (max: 44 – 200 ng/m^3). During the recycling process release of PBDEs and other flame retardant chemicals is observed, since those flame retardants were found at much lower concentrations inside a room where were being assembled circuit boards and in an office with computers (max. of PBDE 209 was 0.32 ng/m^3).

Measurements of PBDEs and PCBs were acquired in air in 31 homes, 33 offices, 25 cars, and 3 public microenvironments (Harrad *et al* [12]). Cars found to be the most contaminated microenvironment for ΣBDE (average) 0.709 ng/m^3, but the least for ΣPCB (average) 1.391 ng/m^3.

The high levels of PBDEs in internet cafes/computer rooms (mean: 0.17 ng/m^3), in public buildings/offices (mean: 2.25 ng/m^3) and in computers/electronics shops (mean: 0.117 ng/m^3) could be attributed to the presence and/or usage of electronic devices, such as personal computers, monitors, televisions, etc. (Mandalakis *et al* [13]). Lower PBDE levels were observed in furniture stores (mean: 0.012 ng/m^3), houses (mean: 0.009 ng/m^3) and in a laboratory (mean: 0.003 ng/m^3). The relatively limited operation of electronic devices might be the reason for the slightly lower PBDE levels in computers/electronics shops compared to offices, internet cafes/computer rooms where electronic devices are in use.

By comparing the sum of six of the most frequently and reliably monitored congeners (Σ_6PBDE, sum of BDE 28, 47, 99, 100, 154 and 153), results show that workplaces in UK (mean: 0.166 – 2.787 ng/m^3) and Sweden (mean: 1.231 ng/m^3) contain higher Σ_6PBDE levels than workplaces in Greece (mean: 0.047 ng/m^3) (Mandalakis *et al* [13]). Moreover, the levels of PBDEs measured in Greek homes (mean: 0.007 ng/m^3) were among the lowest reported in the literature due to the possible low-level usage of PBDE-containing material in residential homes in Greece and/or due to enhanced natural air ventilation (opening of windows) of houses in Greece due to warm climatic conditions.

Measurements of polychlorinated biphenyls (PCBs) were acquired in air in 31 homes, 33 offices, 25 cars, and 3 public microenvironments (Harrad *et al* [12]). The high ΣPCBs concentrations in public microenvironments (MEs) (mean: 30.73 ng/m^3) and offices (mean: 18.15 ng/m^3) were possibly due to the high content of PCB-containing materials that are used inside those buildings. Inversely, lower PCBs concentrations were detected at homes in Birmingham (mean: 2.82 ng/m^3) and in cars (mean: 1.39 ng/m^3).

PCB-containing materials were found to still release PCBs in homes since the indoor PCB concentrations were constantly higher than outdoors, approximately by a factor of 2–50 circa (Menichini *et al* [14]). The indoor concentrations of Σ_{62} PCBs varied between 6.5 and 33 ng/m^3. The windows were closed during the measurements for almost all the sampling time; thus, the contribution of the indoor sources was close to the highest achievable under these experimental conditions. No apparent association was originated linking concentrations and height of the floor: the high/low floor ratios for both Σ_{62} PCBs (sum of: PCBs17, 18, 28, 30, 41, 44, 47+48, 49, 52, 64, 66+80, 70, 74, 77, 81, 85, 87, 91, 95, 97, 99, 100, 101, 105, 110, 114, 118, 123, 128, 135, 136, 137, 138+163, 141, 146, 149, 151, 153, 155, 156, 157, 167, 170, 171, 172, 174, 176, 177, 180, 183, 187, 189, 194, 195, 200, 201, 202, 203+196) and Σ_6 PCBs (sum of: PCBs 28, 52, 101, 138 (+coeluting 163), 153, 180) were close to one at three buildings and 0.3–0.5 in the fourth one.

The PCB concentrations at most of the 181 public buildings in the state of Schleswig-Holstein were below the German guideline-level that has been set at 300 ng/m^3 (Heinzow *et al* [15]). Buildings reinforced with concrete, erected between 1968 and 1972, had elevated PCB concentrations in indoor air. Contrarily, the lowest levels were found in brickwork-constructed buildings. High concentration of the Σ_{15}PCB (sum of: PCBs 28, 52, 101, 138, 153, 180, 8, 18, 22, 31, 44, 49, 70, 99, 118) was marked (maximum: 1071 ng/m^3), generating doubts about its potential health effects.

Lower chlorinated PCBs were the prevailing congeners in a contaminated school building in Nuremberg, Germany (Liebl *et al* [16]). The concentrations of PCB 28, 52, 101 ranged between 4-600, 38-2300, 3-1100 ng/m^3 respectively. Concentrations of the three higher chlorinated indicator congeners were less than 80 ng/m^3. Total concentrations of all PCB congeners (sum of indicator congeners times 5) ranged between 690-20800 ng/m^3 (median 2044 ng/m^3). These levels were much higher than those in the previous study environments in Europe and it would be very important to distinguish the sources of materials that release such high levels of PCBs.

Total PCB levels in indoor air up to 4200 ng/m^3 were observed in various industrial and public buildings in Switzerland emitted from joint sealings. In an industrial building contaminated with PCB (former production of transformers), total PCB levels up to 13000 ng/m^3 were detected. Public buildings had lower PCBs than industrial buildings but in a few of those values close to the maximum value that have been imposed by a range of countries were reached. Depending on the quantity of joint sealings in a room, on the PCB type and content in the materials, and on the air exchange rate, total PCB concentrations in indoor air up to several 1000 ng/m^3 may take place. (Kohler *et al* [17])

3.1 Health effects of PBDEs and PCBs

PBDEs have a propensity to bio-accumulate and have similar chemical structure to thyroid hormones and polychlorinated biphenyls (PCBs). Bio-accumulation of PBDE congeners in human tissue and other biota could cause disruption of thyroid hormones, neurobehavioral deficits and possibly cancer (McDonald [3]). Hydroxylated metabolites of PBDEs structurally resemble thyroid hormones and therefore compete for binding to thyroid hormone receptors and transporter proteins and thus bind to thyroid hormone transport protein (i.e. transthyretin) (Meerts *et al* [18]) and to the thyroid hormone receptors TR-α1 and TR-β (Marsh *et al* [19]). Studies at newborn mice point out that PBDEs like PCBs, cause learning and motor deficits as well as brain development that worsen by ageing (Eriksson *et al* [20]). Workers exposed to polybrominated biphenyls and polybromodiphenyl ethers, e.g., DeBDE, during manufacture revealed a higher than normal prevalence of primary hypothyroidism and a noteworthy decline in sensory and fibula motor velocities (WHO [21]).The fully-brominated deca-BDE has been accused for inducing carcinogenicity in male and female rats [22]. High levels of BDE-47 in adipose tissue in persons has been correlated to non-Hodgkin lymphoma (NHL) disease (Hardell *et al.* [23]). Skin irritation is also caused from PBDE exposure (WHO [21]).

Inversely to the PBDEs, PCBs have been classified from the International Agency for Research on Cancer (IARC) for their carcinogenicity. Limited evidence for carcinogenicity has been found in humans and sufficient in animals. Overall summary evaluation of carcinogenic risk to humans is Group 2A. Therefore, the agent is probably carcinogenic to humans (IARC [24]). PCBs akin to PBDEs are toxic substances, stable in the environment with propensity to bio-accumulate. As a result they are present in the aquatic as well as in the terrestrial food chain (Jensen [25]). The health effects attributable to PCBs exposure include: (a) cancer, including increases in liver cancer and malignant melanoma; (b) immune effects such as suppress immune responses and decrease host resistance; (c) reproductive effects, including possible reduction of birth weight, conception rates and live birth rates as well as reduced sperm counts; (d) neurological effects including possible deficits in visual recognition, short-term memory and learning and possible peripheral neuropathy; (e) endocrine effects, such as exert effects on thyroid hormone levels and suppress thyroid hormone receptor (TR)-mediated transcription ; (f) elevations in blood pressure, serum triglyceride and serum cholesterol, chlorance, eye and skin irritation, headache, dizziness, depression, increased eye discharge, increased sweating at the palms and feeling of weakness, hyperpigmentation of skin and mucous membranes. (Faroon *et al* [26])

3.2 Threshold or safe level/restrictions

PentaBDE and octaBDE are candidates for inclusion in the United Nation's Stockholm Convention on Persistent Organic Pollutants (POPs). DecaBDE, remains in use today in North America, but in the EU it was banned on 1 April 2008 by the European Court of Justice.

The National Institute of Occupational Health (NIOSH) of the US considers chlorodiphenyl containing 54% chlorine to be a potential occupational carcinogen (Aroclor 1254). NIOSH usually recommends that occupational exposures to carcinogens should be limited to the lowest feasible concentration. The recommended exposure limit for 10 hours time-weighted average is set to 0.001 mg/m^3 (or 1000 ng/m^3) (NIOSH [27]). The guideline-level of total PCB indoor air concentrations in buildings in Germany has been set at 300 ng/m^3 (tolerable concentration) and 3000 ng/m^3 (action level) [28]. In addition, based on a tolerable daily intake (TDI) of 1 μg of PCB per kg body weight per day, a tentative guideline value (maximum tolerable concentration) for PCB in indoor air of 6000 ng/m^3 was communicated by the Swiss authorities. This limit was set for buildings such as schools or offices, where people spend an average of 8 hours per day. For buildings such as residences, where permanent exposure can be assumed (24 h per day), the maximum tolerable concentration is 2000 ng/m^3 [29].

4 Conclusions

High concentrations of PBDE congeners 99 and 209 were found in various locations in the UK and also in Sweden in a recycling plant. Those high concentrations are attributed to particularly stringent fire safety regulations existing in the UK and to the fact that Deca-PBDE (PBDE 209) had not been banned in the EU during these measurements. PBDEs in Athens were higher in places were computers or electrical devices were in use than places with electrical devices switched-off. Moreover, comparison among diverse places in Europe showed that dismantling halls, PC rooms in Sweden and offices in UK had the highest Σ_6BDEs (sum of BDE 28, 47, 99, 100, 154 and 153) compared to other indoor environments.

The indoor air concentration of PCBs is independent from the height of the apartments and is mainly related to indoor materials containing PCBs. High PCB concentrations in indoor air were detected in buildings reinforced with concrete. Lower PCB levels were found at brickwork constructed buildings. Very high PCB concentrations were monitored in a school in Nuremberg, Germany, where the maximum values of the congeners PCB 28, 52 and 101 were higher than the proposed tolerable concentration limit (300 ng/m^3).

Limited data on the human health effects of PBDEs exist in literature. PBDEs seem to act as thyroid disruptors and as skin irritants in humans. Carcinogenic, reproductive and neurobehavioral diseases have been observed in animal studies as reviewed in the work of (Darnerud *et al* [30]). Inversely PCBs have been accountable for serious diseases including cancer, endocrine disruption, neurological and reproductive diseases and dermal effects. Food, especially from the aquatic environment as well as mother's milk seems to increase the human risk in such diseases. In addition environments where high air concentrations of HFRs exist should be avoided.

4.1 Suggestions

Recycling plants, schools and hospitals should be designed in such a way, so they do not impose risk for occupational environment. Increase of the ventilation inside those buildings, and removal of products or materials that are possible sources of PBDEs and PCBs could provide healthier exposed air. The exposure to flame retardants is not simply a national problem, since products are used in countries other than the place of manufacture, whereas migration of such chemicals through air is inevitable. Therefore harmonised guidelines of PBDEs and PCBs are needed with the co-operation of all the countries. Accidents of additions of PBDEs and PCBs to either food or animal food have to be strictly avoided. Methods of decontamination of aquatic environments will decrease the current levels of HFRs. Alternatives to flame retardants are needed. Those alternatives should have a satisfactory protection against fire but at the same time they have to be environmental friendly as well as not causing health problems. Some include magnesium dioxide and mixtures of it with antimony oxides, boron, melamine, melamine salts, silicon dioxide, and silicones as well as a newer class of materials known as "nano-additives" such as layered clay minerals [31].

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