Results of an european survey based on PBDEs analysis in household dust

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Introduction

Polybrominated diphenyl ethers (PBDE) are a group of bromine-containing organic compounds presented in a variety of polymer, textiles and other materials to prevent or retard of fire. Owing to the risks to the environment and health of penta- and octa BDEs commercial mixtures, the European Union has limited their use and commercialisation (76/769/EEC). Besides, the risk assessment on decaBDE is being performed to clear the uncertainties concerning possible negative effects. However, as the result of their widespread use and lipophility, PBDEs have become ubiquitous in environment and people[1].

Human exposure to persistent chemicals has traditionally been considered to be mainly through food consumption. Other exposure routes such inhalation and/or dermal exposure are only of quantitatively importance in the case of occupational exposures. However, this may be doubtful for PBDEs, which are being utilized in the modern indoor environment. These compounds are liberally applied to many common household items such as furniture, mattresses, computers and TVs to retard or hinder the outbreak of fire. Over time, these flame-retardants may leach out into the home environment, where they may be inhaled or ingested.

On the other hand, house dust is a repository for environmental pollutants that may accumulate indoors[2]. House dust may be a particularly significant source of chemicals exposure for children, even more important than food consumption. They spend a lot of time on floors and carpets where dust accumulates and, frequently, put their hands and other objects in their mouths “mouthing”, increasing the ingestion of dust. Recently, the PBDEs presence in household dust has produced an increasing interest in the international community, owing to data resulting from several articles published[3],[4],[5].

In the frame of an International collaboration, “Euroconsumers Organization” and “CIEMAT-POPs Group” have performed a study to evaluate the real dimension of this feasible problem. The investigation has been focused on using the contents of 100 vacuum cleaner bags coming from four European countries, as an indication of indoor PBDE contamination. Correlations with properties of the houses, such as location (urban or rural), square footage, indoor temperature and number of electrical appliances and computers have been also examined for any positive influences.

Materials and Method

A hundred of household dust samples were collected using domestic vacuum cleaners: 34 coming from Spain, 32 from Belgium, 22 from Portugal and 12 from Italy. Each dust sample was the result of cleaning a single home during 2003-2004 winter. Vacuum cleaner bags were covered with aluminum film for preserving from light, humidity and other external factors which might changes its chemical composition. All consumers, involved in the investigation, had to fill down a complete questionnaire for obtaining a detailed information about type and size of buildings, floor coverage, number of mattresses, carpets, electrical appliances or computers, indoor temperature and inhabitant habits. Samples were sent with their corresponding attached questionnaire to CIEMAT laboratory.

An amount of ˜ 1 g of dust was extracted with a mixture of hexane:acetone (3:1) as solvent in an ASE 100 system (Accelerated Solvent Extraction)[6]. Liquid extract was then concentrated to a small volume and futher subjected to purification in an automated cleanup system, including pre-packed sealed Teflon multi-layered silica gel and basic alumina columns. Analyses from mono- to hepta-BDE were carried out by GC/MS/MS in a Varian Saturn 2000 workstation equipped with a CP-3800 Gas Chromatograph. Complete details of this analytical methodology were published elsewhere[7]. DecaBDE 209 was studied by using a Gas Chromatograph Agilent 6890 connected to a Mass Spectrometer Agilent 5973 Network. A Factor Four VF-5ms (15m x 0.25 mm i.d., 0.1 µm film thickness)

containing 95% dimethyl-5% diphenylpolysiloxane capillary column was used for its determination. Identification and quantification of target species were carried out by following criteria of isotopic dilution technique, allowing high accuracy in calculation of final results. Limits of detection, LODs, were defined as the smaller concentration giving a signal with S/N > 3. LODs ranged from 0.006-0.220 ng/g d.w. (for BDE 3) to 0.550-20.900 ng/g d.w. (for BDE 209). As a routine procedure, a blank for subtracting the background level, was measured each ten samples batch.

Results and Discussion

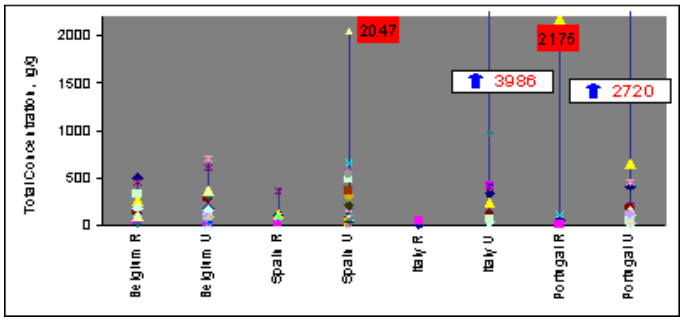
Table 1. Total concentration of PBDEs in dust (ng/g d.w.)

	Mean	Median	Lower	Higher
Spain	***238***	98	19	2047
Italy	***581***	286	11	3986
Portugal	***354***	91	15	2720
Belgium	***190***	125	18	711

The statistics values of total PBDE concentration in dust samples are depicted in Table 1. All samples analysed have PBDEs above limits of detection. Concentrations found in this study are in the same levels than those reported in other European investigations[5], and lower than the American ones[4]. Single high values have been observed in Spain (3% of samples), Italy (21 %) and Portugal (10%). In the concrete case of Spain, the highest value corresponds with dust sucked directly from the inner of a computer, corroborating the idea of PBDEs leach out from plastics and electronics to environment.

Correlation studies of house properties and inhabitant habits with PBDEs content have shown higher PBDEs values in urban samples than in those of a rural origin. Figure 1 shows as the highest values, are related to dusts coming from urban environment (mg/g levels). A mixture of penta (mainly TeBDE 47 and PeBDE 99) and deca compounds has been found, being more significant the contribution of decaBDE 209. Figure 2 shows the average isomer profiles corresponding with the four countries.

Figure 1. Influence of the location on the total PBDEs concentration



The mean percentage of decaBDE 209 is above 60 % in all cases.

We have also examined the BDE 47/BDE 99 ratio to compare it to the commercial DE-71 pentaBDE mixture, with 0.6 ratio. 24 % of samples have displayed a very similar ratio (0.5-0.7), suggesting those dust samples could be collected very close to a product that contained that mixture. A higher ratio has been found as the average (0.9), indicating both 47 and 99 congeners could come from other different pentaBDE mixtures or even that the degradation of BDE 99 may be leading to the formation of 47 BDE. Besides, as BDE 47 has a higher vapor pressure, this congener may be fluxing out of the treated products at a greater rate than BDE 99. Other authors have studied such ratio, publishing values higher than 0.6[8].

In summary, the results presented here corroborate the house dust is a PBDEs source and a possible human exposure route. It would be desirable to avoid their use in household products. These data could be considered as a first reference point for evaluating future PBDEs variations in the European indoor environment after the implementation of the 76/769/EEC European Directive.

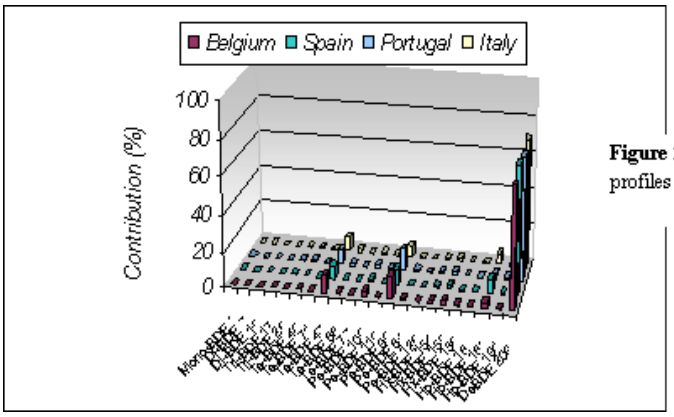


Figure 2. Average isomer profiles of dust samles

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