Levels of Polybrominated Diethyl Ether (PBDEs) in some foods commonly consumed in Nigeria

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Introduction

Brominated flame retardants (BFRs) are high volume industrial chemicals usually incorporated into a wide range of textiles and polymers used in several consumer products [1]. They are frequently applied to combustible materials to reduce their flammability, to delay ignition and to meet fire safety requirements [2] in the production of electric cable insulation, plastic, furniture (carpeting and drapery), office equipment, textiles, water and sewage pipes and electronic devices such as televisions, computers, and copier [3]. Polybrominated diphenyl ethers (PBDEs) are non-reactive BFRs, which are not chemically bound and may escape during production, use, disposal and recycling processes [4]. There are 209 congeners of PBDEs which can get into the environment. The Stockholm Convention has listed PBDEs and some other persistent organic pollutants (POPs) for strict regulation because of their persistence and toxicity [5]. Adverse effects on humans have been reported for PBDEs and other POPs. They have been linked to cancer, reproductive disturbances, metabolic syndrome, obesity and neurodevelopmental toxicity [6] and have been found in different environmental matrices and human organs such as outdoor environment, indoor environment [7] and biota [8], human serum [9], breast milk [6] and urine [10].

Food has been reported as the main source of human exposure to PBDEs in several studies [11]. The occurrence of PBDEs in food is probably as a result of their high persistence and hydrophobicity, leading to bioaccumulation in the food chain. There is limited information on PBDEs in different environmental matrices and foods in Nigeria [12]. Thus, this study assesses the levels of PBDEs in some foods commonly consumed in Nigeria so as to ascertain the potential human exposure to this chemical through diets.

Materials and method

All chemicals and reagents used were of analytical grade and of high purity. HPLC grade acetone and n-hexane used for the extraction were obtained from Merck (Germany). The magnesium sulphate and sodium chloride were obtained from BDH Laboratories (England). A mixture of 8 PBDEs congeners (BDE 17, 28, 47, 99, 100, 153, 154, 183) was obtained from AccuStandard Inc. (New Haven, CT06513, USA). All glassware were washed thoroughly and rinsed with solvents prior to use. A total of composite food samples of 7 different food categories were collected in 2 superstores and 3 local markets in Lagos and Ibadan. The samples collected include: meat (6 raw beef, 8 raw chicken, 4 canned beef, 4 canned chicken), fish (6 frozen fish, 8 seafood, 6 dried fish, 4 canned fish), dairy (2 raw cheese, 4 processed cheese, 4 evaporated milk), edible oil (10 vegetable oil, 6 palm oil), 6 eggs, fruits and vegetables (4 apples, 2 canned apple juice, 4 tomato, 2 canned tomato paste) and cereals (6 beans and 6 rice). Each composite sample was made up of 3 different samples collected at three different sampling points. The raw beef, raw chicken, frozen fish, raw cheese and eggs were boiled in a beaker and homogenised in an electric blender prior to extraction. Canned beef, canned chicken, seafood, dried fish, canned fish, processed cheese, evaporated milk, vegetable oil, palm oil, apples, canned apple juice, tomato, canned tomato paste, beans and rice were homogenized without

processing prior to extraction. The food samples were kept in an air tight glass containers and refrigerated until further analyses. 5 g of the homogenized food samples were placed in 50 mL centrifuge tubes, 20 mL of 1:1 acetone: hexane (v/v) was added, and then shaken for 1 min. 1 g of NaCl and 4 g of $MgSO_4$ were added and was shaken for 3 min. Samples were centrifuged for 5 minutes at 3400 rpm, and 1 mL of each extracts were transferred into a Supel QuE PSA/C18/ENVI-Carb (AC) tubes (QueChERS kit), shaken for 1 min and centrifuged for another 3 minutes at 3400 rpm. Supernatants were separated into a 2 mL glass vials and analysed using Agilent model 6890A gas chromatograph equipped with a ^{63}Ni microelectron capture detector (μECD). An Agilent DB-XLB fused silica column (30 m x 250 mm x 0.25 um i.d) was used. The operating conditions was: injector temperature 250°C, detector temperature 300°C, oven temperature was initially at 100°C (1 min hold) and finally increased to 300°C at 25°C/min (10 min hold) to give the total run time of 19 mins. The carrier gas was nitrogen (99.99 % purity) and flow rate was 1 ml/min. One microliter of the extract was injected. Calibration curve was obtained using PBDEs working standards (500, 250, 125, 62.5, 31.25 and 15.63 ng/g). Recovery ranged between 81–105% for all the congeners. The LODs ranged from 0.005 pg/g- 0.01 pg/g while the LOQs ranged from 0.02 pg/g to 0.03 pg/g.

A questionnaire-based dietary survey was conducted in the two cities. Dietary data were collected through interviews of 250 residents. Information on the age, weight and gender of each interviewee and their consumption rate of the food categories were obtained and analysed statistically using Statistical Package for the Social Sciences (SPSS). Human exposure to PBDEs via consumption of the studied foodstuffs was estimated based on the average daily intake (EDI) and health risk index (HRI) [13]:

$$\text{Estimated Daily Intake (EDI)} = \frac{C \times IR}{BW}$$

$$\text{Health risk index} = \frac{\text{Estimated Daily Intake}}{R_{fd}}$$

Where, C is the mean PBDE concentration (pg/g) in foods obtained in this study and IR is the mean intake rate of meat, fish, dairy, edible oil, vegetable and fruit, egg and cereal, which was 50, 100, 30, 100, 120, 50 and 1000 g/day, respectively. The average body weight (BW) for an adult used was 68 kg. R_{fd} is the Reference dose.

Results and Discussion

The concentrations of PBDEs in all the food samples ranged from 5.78-35.9, 6.73-113, 3.66-11.6, 1.08-40.6, 2.08-6.31, 2.42-7.94 and 1.59-2.70 pg/g in the meat, fish, dairy, edible oil, vegetable oil, egg and cereal, respectively. The mean concentrations of the PBDEs in all the samples are shown in Table 1. When compared with what was reported in the literature, the levels of ΣPBDEs in foods commonly consumed in Lagos and Ibadan, Nigeria was lower than what was reported in the USA [14], Spain [15], and Hong Kong [16]. The estimated daily intake (EDI) of PBDEs in meat, fish, dairy, edible oil, vegetables and fruits, egg and cereals were 10.7, 132, 2.84, 19.5, 3.85, 7.57 and 29.0 pg/kg/day, respectively (Table 2). Fish had the highest concentrations of all the congeners followed by edible oil and meat while the lowest was in dairy products. The EDI values of the congeners ranged from 0.01 - 132 pg/kg/day and were very low except BDE-28 and BDE-17 in most foods (Figure 1). Hazard risk indices (HRI) must be less than 1 for it not to pose any health hazard [17]. The HR of the congeners, BDE-47, BDE-99 and BDE-153, which reference doses were available obtained in this study ranged from 1.85E-10-6.46E-8. The total HRI values for the three congeners was lower than 1, which indicated that there was low risk of exposure to

PDBEs in foods commonly consumed in the two cities (Table 3). This study showed that the estimated dietary exposure to PBDEs is unlikely to be a significant health concern in Lagos and Ibadan but there is need for routine monitoring of PBDEs and other toxic chemicals in foods commonly consumed in Nigeria to safeguard public health. Also, considering other food categories and animal feeds is imperative and recommended.

Table 1: Mean concentrations in pg/g of PBDEs in different food samples commonly consumed in Lagos and Ibadan, Nigeria

Food samples	Minimum concentration (pg/g)	Maximum concentration (pg/g)	Mean±SD (pg/g)
Meat (n = 22)	5.78	35.9	14.5±10
Fish (n = 24)	6.73	113	90.1±108
Dairy (n = 10)	3.66	11.6	6.44±3.6
Edible oil (n = 16)	1.08	40.6	13.3±7.2
Eggs (n = 6)	2.42	7.94	5.23±1.5
Vegetables and fruits (n = 12)	2.08	6.31	4.29±0.4
Cereal (n = 12)	1.59	2.70	1.97±0.4

Table 2: Estimated daily intake of PBDEs (pg/kg/day) in the different food samples commonly consumed in Lagos and Ibadan, Nigeria

Food categories	BDE 17	BDE 28	BDE 47	BDE 99	BDE 100	BDE 153	BDE 154	BDE 183	PBDE
Meat	1.17	4.92	1.09	0.45	1.11	1.69	0.16	0.08	10.7
Fish	28.9	47.5	10.6	16.1	12.4	9.79	6.30	0.82	132
Dairy	0.19	0.71	0.18	0.20	0.31	1.11	0.01	0.13	2.84
oil	6.61	7.62	1.14	0.54	1.82	1.20	0.54	0.06	19.5
egg	0.30	0.53	0.14	0.19	1.18	0.76	0.23	0.52	3.85
veg	0.76	1.32	0.44	0.86	0.68	1.96	0.77	0.77	7.57
cereal	3.15	6.39	2.48	1.36	6.10	3.59	0.59	5.32	29.0

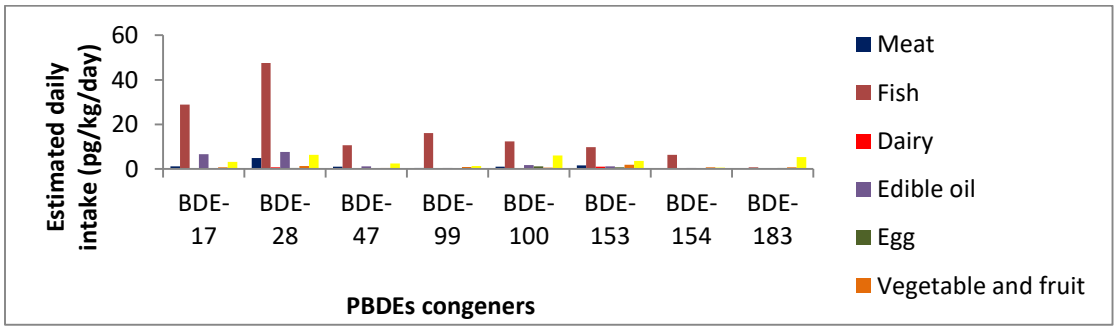

Figure 1: Estimated daily intake of PBDE congeners in foods commonly consumed in Lagos and Ibadan, Nigeria

Table 3: HRI of PBDEs in foods commonly consumed in Lagos and Ibadan, Nigeria

Food categories	BDE-47	BDE-99	BDE-153	ΣPBDE
Meat	1.17E-09	4.49E-10	8.43E-10	2.46E-09
Fish	2.89E-08	1.61E-08	1.96E-08	6.46E-08
Dairy	1.91E-10	1.95E-10	2.23E-09	2.62E-09
Edible oil	6.61E-09	5.38E-10	2.40E-09	9.55E-09
Egg	3.02E-10	1.85E-10	1.53E-09	2.01E-09
Vegetables and fruits	7.62E-10	8.58E-10	3.92E-09	5.54E-09
Cereal	3.15E-09	1.36E-09	7.18E-09	1.17E-08

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