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Assessment of the Levels of Polybrominated Diphenyl Ethers in Blood Samples from Guadalajara, Jalisco, Mexico

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Abstract The purpose of this study was to measure levels of polybrominated diphenyl ethers (PBDEs) in the blood of children (50 individuals) living in Guadalajara, Jalisco, Mexico. We analyzed six PBDE congeners by gas chromatography–mass spectrometry. Total PBDE levels ranged from not detectable (nd) to 15.2 μg/L on a whole-weight basis and from nd to 6,435 ng/g lipid on a lipid-weight basis. The dominant congener in our study was BDE-153, followed by BDE-154, BDE-99, BDE-100, and BDE-47. Levels of BDE-209 were below the detection limit. Our data indicate that children living in the areas studied in this work are exposed to high levels of PBDEs.

Keywords Brominated flame retardants · Children · Mexico · BDE47 · PBDEs · POPs

The Stockholm Convention on persistent organic pollutants (POPs), which came into force on the 17th May, 2004, initially outlawed the use of 12 industrial chemicals. However, in actuality, 22 chemicals are included in the Stockholm Convention on POPs (including polybrominated diphenyl ethers). The Stockholm Convention is a global treaty to protect human health and the environment from chemicals that remain intact in the environment for long periods, become widely distributed geographically, and accumulate in the fatty tissue of humans and wildlife. Moreover, this convention sought to determine baseline levels in environmental and biological samples. In developing countries, the levels of these chemicals in hot spots may be a public health issue because of their magnitude (UNEP 2012; Trejo-Acevedo et al. 2009).

Brominated flame retardants (BFRs) have routinely been added to consumer products for several decades in a successful effort to reduce fire-related injury and property damage (Costa et al. 2008). In this regard, polybrominated diphenyl ethers (PBDEs) are used as flame retardants in a wide range of products, for example textiles, automotive parts, construction materials, printed circuit boards, television, computer housings, and other electronic household equipment (Costa et al. 2008). In general, they are used as additives (not chemically bound to the material); they can, therefore, leak out of products into the environment (Costa et al. 2008). PBDEs have been marketed as one of three mixtures, known as pentabrominated BDE (PentaBDE), octabrominated BDE (OctaBDE), and decabrominated BDE (DecaBDE). DecaBDE is the most widely used PBDE globally, and is still produced in the USA and in Europe; pentaBDE and octaBDE have recently been banned in the European Union and in several states in the USA, and are no longer produced in these countries (Costa et al. 2008). Moreover, pentabrominated BDE and octabrominated BDE are included in Annex B of the Stockholm Convention (UNEP 2012).

In Mexico, two studies have been performed to determine exposure of the general population to PBDEs. The first, a preliminary study, assessed levels of PBDEs in the blood of women living in an urban area in San Luis Potosi

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city; in the same study, PBDE levels in the milk of women living in the rural Huasteca Region in San Luis Potosí were also evaluated (Lopez et al. 2006). PBDE blood levels in women living in the urban area were 21.5–37.5 ng/g lipid; levels in milk from women living in the rural area were 0.8–5.4 ng/g lipid (Lopez et al. 2006). In a second study PBDE levels were assessed in the blood of 173 Mexican children. Total PBDE blood levels ranged from not detectable (nd) to 43.4 ng/g lipid; the dominant PBDE congener was BDE-47. Of all the areas studied, PBDE levels were highest in children living in an industrial urban area (Ciudad Juarez, Chihuahua, Mexico) (Pérez-Maldonado et al. 2009).

Therefore, taking into account the scarcity of data for any matrix in Mexico, there is an urgent need to measure the concentrations of PBDEs in environmental and biological samples. In this context, the purpose of this study was to measure PBDE levels in the blood of children living in Guadalajara, Jalisco, Mexico.

Materials and Methods

The sampling sites were selected on the basis of previous knowledge of the economic activity in each area; sampling was performed in three different areas in Guadalajara, Jalisco, Mexico (Fig. 1; Table 1). Children of school age and residents of the study areas since birth were eligible for inclusion in this study. The parents of the participating children were informed about the study and all gave written informed consent before inclusion in the study. After informed consent agreements had been signed by all subjects, a questionnaire was circulated and blood and urine samples were obtained. During 2011, 50 blood samples

Fig. 1 Location of the communities studied

Table 1 Characteristics of the sites sampled

Site	n	Characteristics
Urban	11	Urban community located in Guadalajara, Jalisco city, with a low impact of agriculture, industrial, traffic, among other activities 20°41′36.11″N; 103°15′53.17″W
Industrial	28	Urban community located in Guadalajara, Jalisco city, with the largest industrial zone in that region. 20°31′03.15″N; 103°10′49.15″W
Agricultural	11	Urban community located in Guadalajara, Jalisco city, with a high impact of agriculture activity 20°34′06.14″N; 103°09′13.49″W

from eligible children (aged 6–12) were obtained. The questionnaire registered characteristics such as source of drinking water, occupational history of parents, age, weight, height, exposure to medicaments, environmental tobacco smoke exposure, and infectious diseases in the last month. The study was approved by the ethics committee of the School of Medicine, Universidad Autónoma de San Luis Potosí. Blood samples were drawn from a cubital vein into 10-mL vacuum tubes for serum collection. The tubes were then centrifuged at 1,200×*g* for 10 min and the serum was transferred with hexane-rinsed pasteur pipettes to hexane-rinsed brown glass bottles. Serum was stored at −20°C until analysis. All analysis of PBDEs in serum samples was performed in accordance with Pérez-Maldonado et al. (2009). In brief, the samples were denatured with 6 M hydrochloric acid (1 mL) and 2-propanol (6 mL), with mixing between each addition, and extracted with *n*-hexane–methyl *tert*-butyl ether (6 mL, 1:1 by volume). The organic layer was isolated and the water phase re-extracted with additional solvent. The organic phase was washed with aqueous potassium chloride (1 %). The solvent was evaporated and the lipid content determined gravimetrically for each sample. Co-extracted lipids were removed on a column of silica–sulfuric acid packed in a 3-mL solid-phase extraction cartridge. We performed final analytical determination of the target analytes by gas chromatography (GC-6890) coupled with mass spectrometry (MS-HP 5973). BDE-77 (2 ng/mL) was used as internal standard. Under these conditions and using data from nine replicate analyses near the lowest concentration attainable on the calibration curve, the method detection limits for all PBDEs were approximately 0.10 ng/mL. Internal controls at 2.0, 4.0, and 6.0 ng/mL for all compounds was included in all series of analyses. At these concentrations the between-assay variation coefficient was 3.0 % (±2.0 %) and average recovery for all compounds was between 93 % and 118 %. For quality control, organic contaminants in fortified human serum (National Institute of Standards and Technology (NIST) SRM 1958) were used; recovery was 95 ± 5 % for all the compounds tested. For all statistical

handling of the analytical data, all measurements below the limit of detection (LOD) were divided by the square root of two. We expressed our results on both whole-weight and lipid-weight basis. Results on a lipid-weight basis were calculated as reported elsewhere (Bergonzi et al. 2009). To satisfy normality criteria levels of all PBDEs were logarithm-transformed. Therefore, all the results are presented as geometric means. Mean levels for the three sites studied were compared by one-way analysis of variance (ANOVA) followed by the Tukey test. For all statistical analysis we used Jmpin Start Statistics Software 5.0 (SAS Institute).

Results and Discussion

Total PBDEs levels ranged from non-detectable (nd) to 29.2 ng/mL on a whole-weight basis and from nd to 9,080 ng/g lipid on a lipid-weight basis. Figures 2 and 3 show mean blood levels of total PBDEs for the three sites studied. The highest mean blood levels were found in children living in the agricultural area (approximately 6.3 ng/mL and 2,000 ng/g lipid; range: nd–29.2 ng/mL and nd–9,080 ng/g lipid), followed by children living in the industrial region (approximately 4.5 ng/mL and 995 ng/g lipid; range: nd–33.5 ng/mL and nd–6,750 ng/g lipid). The lowest mean blood levels of total PBDEs were found in the urban site (approximately 1.0 ng/mL and 130 ng/g lipid; range: nd–3.8 ng/mL and nd–770 ng/g lipid). The dominant congener in our study was BDE-153, followed by BDE-154, BDE-99, BDE-100, and BDE-47 (Fig. 4) in all the areas studied. The levels of BDE-209 were below the LOD (detection limit).

Comparison of PBDE levels in the children in this study with those found in other studies revealed that levels in this study were higher than those previously reported in Mexican children (mean levels of approximately 7.5 ng/g lipid

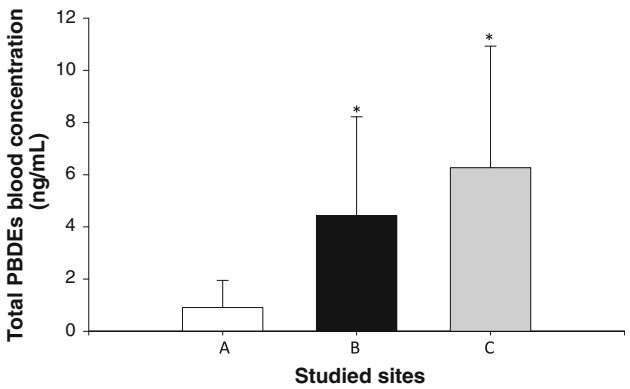

Fig. 2 Total PBDE blood levels (ng/mL) in samples from children living in Guadalajara, Jalisco, Mexico. Values are geometric means. *A* urban site; *B* industrial site; *C* agricultural site. *$p < 0.05$ compared with urban site

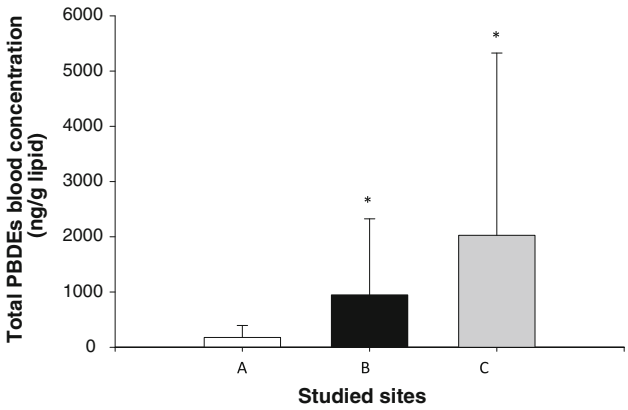

Fig. 3 Total PBDE blood levels (ng/g lipid) in samples from children living in Guadalajara, Jalisco, Mexico. Values are geometric means. *A* urban site; *B* industrial site; *C* agricultural site. *$p < 0.05$ compared with urban site

in a previous study of 173 Mexican children performed by our group; Pérez-Maldonado et al. 2009). Comparison of the levels found in this study with those found in children in NHANES IV (12–19 years old; approximately 53.0 ng/g lipid) revealed levels were much higher in the children assessed in this study – for children from agricultural, industrial, and urban sites in our study PBDE levels were approximately 40, 20, and 2 times higher, respectively, than in children in the USA (NHANES IV 2009). Moreover, mean PBDEs levels in our study (all studied sites) are higher than in other studies throughout the world (Rose et al. 2010; Toms et al. 2009; Link et al. 2012). For example, in a study performed in California, USA, mean PBDE levels in 2–5 year-old children were approximately 210 ng/g lipid (Rose et al. 2010); mean levels for children (aged 7–12 years) living in Australia were 26 ng/g lipid (Toms et al. 2009); in Germany mean levels of 7.8 ng/g lipid were found in blood samples of 9–11-year-old children (Link et al. 2012). Moreover, the levels in this study (for urban and industrial sites) are higher than those found in studies performed in places where computer e-waste is recycled (Han et al. 2011). In this regard, Han et al. (2011) studied children aged 6 to 8 from Luqiao, China, a computer e-waste recycling area. The PBDE content of blood samples from the children was 664.28 ± 262.38 ng/g lipid. An important finding was that BDE-153 was the predominant congener. This finding may be related to changes in environmental accumulation pattern over time, but it could also be affected by temporal changes. The notion of a temporal change is supported by a study performed on Faroe Island, Spain, in which Fängström et al. (2005a) assessed blood concentrations of polybrominated diphenyl ethers (PBDEs) in humans with a high seafood intake. The study was performed on pregnant women in 1994–1995 and in their children at seven years of age. Maternal serum

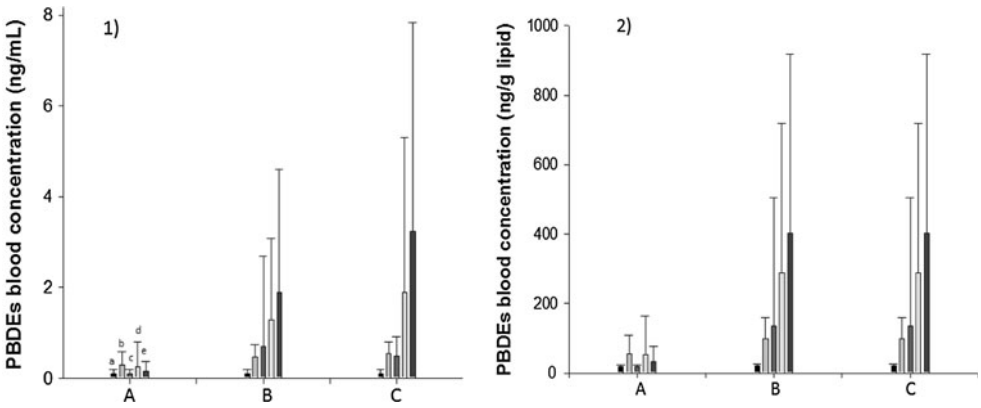

Fig. 4 Levels of PBDE congeners (ng/mL (**1**) and ng/g lipid (**2**)) in blood samples from children living in Guadalajara, Jalisco, Mexico. Values are geometric means. *A* urban site; *B* industrial site; *C* agricultural site. *a* BDE-47; *b* BDE-99; *c* BDE-100; *d* BDE-154; *e* BDE-153

was dominated by BDE-47, whereas BDE-153 was predominant in the children's serum seven years later. These results were also is supported by analysis of human milk from Faroe Island; the amount of BDE-153 increased dramatically between 1994–1995 and 1998–1999 (Fängström et al. 2005b). However, there is no definitive explanation of this change in the accumulation profile of PBDEs. Another possibility could be metabolism of BDE-209, leading to the formation of BDE-153 and BDE-154, or the greater persistence of BDE-153 than of the lower brominated congener BDE-47. Moreover, this result may indicate that sources of lower brominated diphenyl ethers are decreasing.

Our study has some limitations, for example lack of information regarding environmental media and dietary sources. However, our data are indicative of high levels of exposure of children living in the regions studied in this work. Therefore, further studies are needed to discover how people are exposed to PBDEs. In this regard, it would be important to consider diet (Carrizo and Grimalt 2007), dust inhalation, and dust ingestion (Tan et al. 2007; Athanasiasou et al. 2008). With regard to health risks, it is difficult to associate a specific health effect with the levels found in the children studied. However, structural resemblance to other well known environmental contaminants, for example polychlorinated biphenyls (PCBs) and studies of animals indicate toxic effects are two major reasons for environmental and health concern.

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