POLYBROMINATED DIPHENYL ETHERS (PBDEs) IN PLACENTA AND HUMAN MILK

Teija Strandman[1], Jaana Koistinen[1], Terttu Vartiainen[1,2]

[1] National Public Health Institute, Division of Enviromental Health, P.O.Box 95, FIN-70701 Kuopio, Finland
[2] Department of Environmental Sciences, University of Kuopio, P.O. Box 1627, FIN-70211 Kuopio Finland

Introduction

Polybrominated diphenyl ethers (PBDEs) are added to a wide range of products such as electronics, plastics and textiles to protect them from catching fire[1]. DecaBDE is the mostly used PBDE. The annual global use of decaBDE was estimated to be 30 000 tons in 1990. The other commercial products are mainly penta- and octabromodiphenyl ethers. Their global use in 1990 was 4,000 tons and 6,000 tons, respectively[2]. Since PBDEs are not covalently bound to the materials, they can leach to environment during the lifetime of the products.

PBDEs have been found to accumulate in lipids. The accumulation of lower brominated PBDEs is greater than that of higher brominated ones[3,4]. Several investigations of PBDEs in human adipose tissue[5,6,7], blood[8] and milk[9] have been reported recently. 22'44'-TeBDE (BDE 47) seems to be the predominant congener in the human samples, excluding the blood samples from the electronics dismantlers[8]. A time-related trend study shows that the levels of PBDEs in human milk have increased distinctly[9]. In 1972 the concentration of BDE 47 was 0.06 ng/g lipid and in 1997 the concentration had reached the value of 2.28 ng/g lipid.

The health effects of PBDEs are not well known, but an important factor in PBDE toxicity might be their effects on hormonal functions[10]. It has been reported that the oral exposure to PBDEs decreases the serum thyroxin concentration in mice and rats[11]. A nonsignificant elevated risk for Non-Hodgkin's lymphoma connected with PBDEs has been reported in humans[6]. Neonatal exposure of mice to BDE 47 and BDE 99 may cause neurotoxic effects in adult animals [12,13].

The main objectives of this study were to find out if PBDEs can be found in human placenta and to compare the levels in placenta with the levels in human milk.

Materials and Methods

The human milk and placenta samples from eleven donors were analyzed for the content of 2,4,4'-TriBDE (BDE 28), 22'44'-TeBDE (BDE 47), 22'44'5-PeBDE (BDE 99) and 22'44'55'-HxBDE (BDE 153). The samples were selected from an epidemiological study, which is ongoing at the National Public Health Institute in Finland. The collection of samples was performed during 1994-1998. The age of the donors varied from 25 to 42 years (median 34 years). Six samples were from donors of their first childbirth, one from a donor of her second, one from a donor of her third and

two samples from donors of their fourth childbirth. The number of childbirth from one donor is unknown. The PBDE standards were purchased from Promochem.

PBDEs were extracted and isolated from the samples similarly to PCBs and dioxins. The human milk samples were treated in a separation funnel with sodium oxalate solution and ethanol and PBDEs were extracted with a mixture of diethyl ether and hexane. The placenta samples were homogenized (Mixer B-400, Büchi) and freeze dried. A subsample, equivalent to 1 g lipids from milk or 10-14 g freeze dried placenta, was spiked with internal standard mixtures of ^{13}C-labeled PCDD, PCDF and PCB standards. The clean-up procedure included a silica gel column, an activated carbon column containing Celite, an activated alumina column and an activated carbon column[5, 14].

PBDEs were analyzed with a high resolution mass spectrometer combined with a high resolution gas chromatograph which was equipped with a fused silica capillary column (DB5MS, 60 m, 0.25 mm, 0.25 μm). The quantitation of PBDEs was performed by selective ion recording using a Autospec Ultima (Micromass, Altrincham, UK) mass spectrometer (resolution 10,000). The results were calculated using ^{13}C-labeled PCB 180 as an internal standard. The samples were spiked with recovery standards (^{13}C-labeled BDE 47 and BDE 99) before analysis to determine the recovery of PCB 180. The laboratory reagent and equipment blank samples were treated and analyzed by the same method as the actual samples, one blank for every set of samples. Detection limits for the different PBDE congeners were 0.04 ng/g in lipid.

Results and Discussion

The mean, median, minimum and maximum concentrations of BDEs 28, 47, 99 and 153 per lipid weight in human milk and placenta samples are given in Table 1. Since the lipid contents of placentas were not determined, average lipid content 0.5% was used to calculate the concentrations per lipids in placenta.

Table 1. The mean concentration, standard deviation, median, minimum and maximum concentration of BDEs 28, 47, 99 and 153 as ng/g lipid in human milk and placenta (recoveries for theinternal standard ranged from 60 to 114 %).

	BDE 28		BDE 47		BDE 99		BDE 153	
	placenta	milk	placenta	milk	Placenta	milk	placenta	milk
Mean	0.14	0.16	1.09	1.31	0.42	0.39	0.42	0.39
Stdev	0.08	0.15	0.83	1.15	0.18	0.23	0.17	0.20
Median	0.12	0.13	0.77	0.85	0.41	0.35	0.40	0.29
Min	0.06	0.04	0.36	0.30	0.21	0.14	0.22	0.19
Max	0.34	0.59	2.80	4.25	0.89	0.94	0.86	0.72

The concentrations of four studied PBDEs were at the same level in human milk and placenta (Table 1). BDE 47 was the predominant congener in all samples. The percentage of BDE 47 from the sum concentrations of PBDEs varied from 34% to 73% in human milk samples and from 42% to 67% in placentas. The concentrations of PBDEs in human milk reported here are at a similar level as in a Swedish study[9].

The sum concentrations of PBDEs (Sum PBDE) in individuals are presented in Figure 1. Sum PBDE ranged from 0.88 ng/g lipid to 5.89 ng/g lipid in human milk and from 1.00 ng/g lipid to 4.40 ng/g lipid in placenta. The four highest sum concentrations were from donors of their first childbirth.

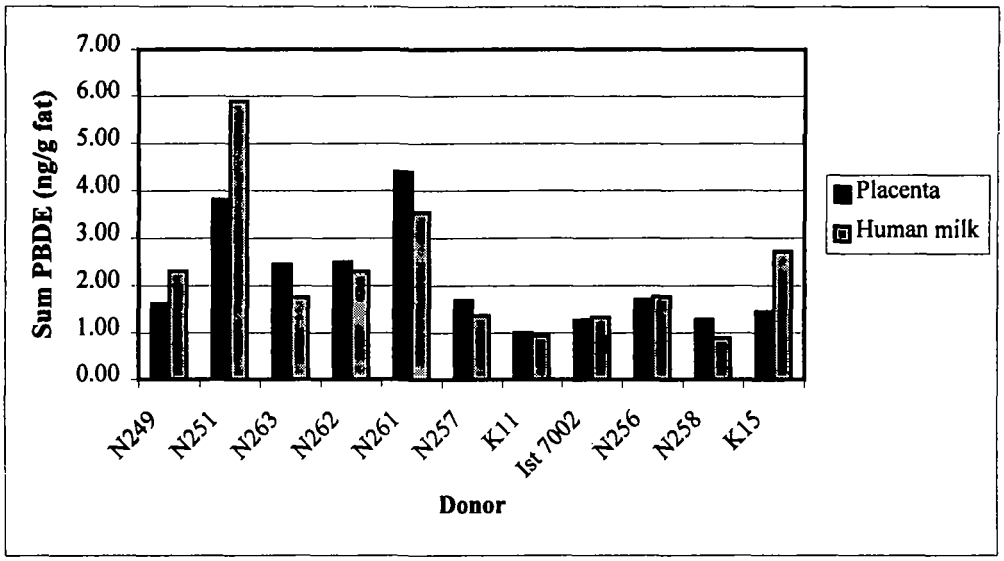

Figure 1. The sum concentration of four PBDE congeners (ng/g lipid) in human milk and placenta from eleven donors.

As far as we know, the occurrence of PBDEs in placenta have not been reported before. According to this study, similar concentrations of PBDEs per lipid can be found in human milk and placenta. If the concentrations in placenta represent the concentrations in fetus, there is a risk that humans can be exposed to PBDEs already in their mothers' uterus and undesirable effects are possible.

Acknowledgements
We thank Ms. Tuula Rissanen and Ms. Katri Mehtonen for technical assistance with sample treatment and Ms. Eija Mehtonen ja Mr. Tuomo Korhonen for HRGC/HRMS analyses.

References

(1) World Health Organization (1997) Flame retardants: A general introduction. Environmental Health Criteria 192, Geneva: WHO

(2) World Health Organization (1994) Brominated diphenyl ethers. Environmental Health Criteria 162, Geneva: WHO

(3) Sellström U., Jansson B., Nylund K., Odsjö T., Olsson M. (1990) Conference abstract, Organohalogen compounds 2, 357.

(4) Pijnenburg, A.M.C.M., Everts, J.W., de Boer, J., Boon, J.P. (1995) Rev. Eviron. Contam. Toxicol. 141, 1

(5) Strandman, T., Koistinen, J., Kiviranta, H., Vuorinen, P.J., Tuomisto, J., Tuomisto, J., Vartiainen, T. (1999) Conference abstract, Organohalogen compounds 40, 355

(6) Hardell, L., Lindström, G., van Bavel, B., Wingfors, H., Sundelin, E., Liljegren, G. (1998) Oncology Research 10, 429

(7) Meneses, M., Wingfors, H., Schuhmacher, M., Domingo, J.L., Lindström, G., van Bavel, B. (1999) Chemosphere 39, 2271

(8) Sjödin, A., Hagmar, L., Klasson-Wehler, E., Kronholm-Diab, K., Jakobsson, E., Bergman, Å. (1999) Environ. Health Perspect. 107, 643

(9) Meironyté, D., Norén, K., Bergman, Å. (1999) J. Toxicol. Environ. Health Part A 58, 329

(10) TemaNord (1998) Polybrominated Diphenyl Ethers: Food Contamination and Potential Risks, no505, ISBN 92-893-0150-3

(11) Darnerud, P.O., Sinjari, T. (1996) Conference abstract, Organohalogen compounds 29, 316

(12) Eriksson, P., Jakobsson, E., Fredriksson, A. (1998) Conference abstract, Organohalogen compounds 35, 375

(13) Eriksson, P., Viberg, H., Jakobsson, E., Örn, U., Fredriksson, A. (1999) Conference abstract, Organohalogen compounds 40, 333

(14) Vartiainen, T., Saarikoski, S., Jaakkola, J.J. and Tuomisto, J. (1997) Chemosphere 34, 2571