Concentrations of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in breast milk of women from Catalonia, Spain

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Introduction

Polychlorinated biphenyls (PCBs) are widely distributed environmental contaminants derived from their extensive use as coolants and lubricants in transformers, capacitors, and other electrical equipment. Polybrominated diphenyl ethers (PBDEs) are widely used as flame retardant in plastics, textiles and are also persistent organic pollutants (POPs). These compounds are lipophilic and difficult to metabolize. Therefore, environmental exposure of living organism results their accumulation in fat tissues via food.

Although incineration has demonstrated to be a commercially available technology for hazardous waste (HW) disposal, the stack emission from HW incinerators (HWI) of a number of inorganic and organic substances has raised an important concern about the environmental and health consequences of this process of treating HW. In 1996, the construction of the first and to date only HWI in Spain was initiated in Constantí (Catalonia, Spain). Regular operations started in 1999. In order to establish the potential health risks of exposure on the general population living near the new HWI, a biological monitoring program was designed[1-3]. To establish baseline levels of dioxins and furans (PCDD/Fs) , and PCBs in human milk, samples were collected from women living in the neighborhood of the facility before starting regular operations.

The purpose of the present study was to determine the concentrations of PCBs in breast milk of women living in the vicinity of the HWI after four years of regular operations in the facility. The levels of PBDEs were also measured.

Experimental

Sampling

Information about the HWI, as well as the characteristics of the area in which it is placed were previously reported[1]. Breast milk samples were collected during 2002 from mothers living in Tarragona County, in zones under potential influence of the HWI. The participants in the study were 25-35 years of age, who had lived in Tarragona County for at least the last 5 years. Only healthy primiparae mothers were included in this survey. Questionnaires about age, current and former area of residence, dietary and toxic habits to mothers donating breast-milk were completed by the participants. Fifteen pooled samples from several individuals were obtained. Seven samples corresponded to women living in urban zones, while the remaining 8 samples belonged to women living near an industrial (petrochemical complex) zone. A questionnaire completed by the participants prior to sampling did not indicate any occupational exposure to PCDD/Fs, PCBs or PBDEs. All mothers had a single child, and infants were exclusively breast-feeding. The time elapsed between delivery and sampling was between one and two months. The milk samples of each participant were pooled (50-100 ml) and were stored frozen (-20^{o}C) until chemical analyses.

Analytical procedure

Fat from milk sample was extracted with a mixture of diethyl ether and hexane after addition of sodium oxalate and ethanol. Fat content was determined gravimetrically. An amount of 1.5 g of the fat was spiked with four ^{13}C-labeled non-*ortho* PCBs (PCB 77, 81, 126, and 169), fourteen ^{13}C-labeled other PCBs (PCB 30 [^{12}C-labeled], 52, 80, 101, 105, 118, 138, 153, 156, 157, 170, 180, 194 and 209), and with six ^{13}C-labeled PBDEs (PBDE 28, 47, 77, 99, 153 and 183). The sample was defeated in a silica gel column with sulfuric acid and purified on an activated carbon column (Carbopack C, 60/80 mesh) containing Celite (Merck 2693). Further clean-up of the sample was achieved with an activated aluminum column (Merck 1097, standardized, activity level II-III). Another activated carbon column (without Celite) was used to separate the non-*ortho* PCBs from other PCBs and PBDEs. The quantification of analytes was performed by selective ion recording using a VG 70-250 SE (VG Analytical) mass spectrometer (resolution 10,000) equipped with a HP 6890 gas chromatograph with fused silica capillary column (DB-DIOXIN, 60 m, 0.25 mm, 0.15 µm). Limits of quantification (LOQ) for the different PCB congeners ranged between 0.07 pg/g fat and 0.01 ng/g fat, and for PBDE congeners between 0.002 and 3.8 ng/g fat . Recoveries for all internal standards were more than 60%. Set

of toxic equivalency factors (TEF), recommended by WHO in 1998, was used for PCB toxic equivalents (WHO-TEQ)[4].

Results and discussion

Tables 1and 2 summarize individual concentrations of planar and total PCBs (33 congeners), as well as fat percentages in the 15 samples of breast milk from mothers living in Tarragona County. Results show data corresponding to the current and the baseline studies. The current PBDE concentrations (sum 15 congeners) are also given. In the present study, the most toxic PCB congeners, PCBs 126 and 169, showed mean concentrations of 35.4 and 27.4 pg/g, respectively. With respect to the total PCBs, congeners 153 and 180 reached the highest levels (70.7 and 51.08 pg/g fat, respectively) (data not shown). In turn, planar PCBs ranged from 1.3 to 6.3 pg WHO-TEQ/g fat with a mean value of 3.8 pg WHO-TEQ/g fat. Total PCB concentrations ranged from 3.8 to 13.3 pg WHO-TEQ/g fat (mean value: 8.7 pg WHO-TEQ/g fat). In the baseline survey, planar PCBs ranged from 2.4 to 11.5 pg WHO-TEQ/g fat, with a mean value of 7.3 WHO-TEQ/g fat, while total PCB concentrations ranged from 5.9 to 24.4 pg WHO-TEQ/g fat (mean value: 15.7 pg WHO-TEQ/g fat). The comparison of the current data with those from other studies is very difficult considering the specific congeners that are included in each study. However, the concentrations of PCBs and PBDEs found in this study are lower than those recently reported for countries such as Belgium and Japan[5,6] .

A comparison of the current mean, standard deviation, and minimum and maximum PCB concentrations with the baseline levels is summarized in Table 3. Significant decreases ($p < 0.001$, Mann-Whitney U-test) were noted for both planar and total WHO-TEQ PCBs: 47.9% and 44.6%, respectively. This reduction is in agreement with the decline observed during the same period in the levels of PCBs in plasma of workers of the HWI under current evaluation.[7]

Table 4 shows the current PCB and PBDE concentrations in human milk samples classified according to the specific place of residence of the participants in the study: urban or industrial zones. Concentrations in women living in the urban area were higher than those in the industrial area (10.1 and 7.4 pg WHO-TEQ/g fat, respectively). Although no significant differences were found between both groups, a trend for a greater decline of concentrations in the industrial area could be observed. A 60% reduction in PCB concentrations in human milk from women living in the industrial area was noted, whereas the decrease reached only a 30% in women living in urban areas.

Taking into account the results of this study in human milk samples, as well as the concentrations of PCBs recently found in plasma of workers who live in the same area[7], together with the recent results concerning the environmental monitoring program (soil and herbage samples)[8], we can conclude that living in the vicinity of the new HWI should not mean any additional exposure to PCBs for the general population of the area.

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Table 1. Individual levels of planar and total PCBs , PBDEs and percentages of fat in samples of breast milk from women living near the HWI in 2002 (Constantí, Catalonia, Spain). Current data.

	Planar PCBs		Total PCBs		PBDEs	
Sample Code	Sum of planar PCBs (pg/g fat)	WHO-TEQ (pg/g fat)	Sum of all PCBs (ng/g fat)	WHO-TEQ pg/g fat	Sum of PBDEs (ng/g fat)	Fat %
1	54.8	2.5	185.9	7.1	5.6	3.2
2	28.8	1.3	134.3	3.8	2.5	3.1
3	91.4	4.4	295.6	9.9	3.7	1.6
4	96.7	5.2	330.2	10.7	2.6	1.6
5	115.6	6.3	402.3	13.3	1.8	3.0
6	85.1	4.8	120.6	8.3	1.5	1.7
7	73.4	3.8	305.1	9.0	1.2	4.1
8	96.2	5.1	230.5	12.7	1.5	1.6
9	66.8	4.3	253.0	8.4	1.4	3.0
10	59.1	3.3	253.2	7.6	1.4	4.7
11	61.0	3.0	177.6	6.6	1.0	3.6
12	63.3	3.7	275.9	8.0	6.6	2.5
13	42.8	2.0	171.6	4.9	2.3	4.1
14	71.5	2.8	219.1	6.9	1.3	4.3
15	102.8	4.7	471.0	13.0	1.7	3.4

Table 2. Individual levels of planar and total PCBs and percentages of fat in samples of breast milk from women living near the HWI (Constantí, Catalonia, Spain). Baseline data.

Sample code	Planar PCBs		Total PCBs		Fat %
	Sum of planar PCBs (pg/g fat)	WHO-TEQ (pg/g fat)	Sum of all PCBs (ng/g fat)	WHO-TEQ pg/g fat	
1	113.7	5.1	337.2	11.6	1.5
2	50.3	2.4	184.4	5.9	2.7
3	138.8	8.3	494.3	16.8	1.8
4	111.5	6.7	425.9	14.3	3.0
5	191.2	11.5	694.1	24.4	2.9
6	164.5	9.0	611.3	19.2	3.0
7	73.7	3.9	266.0	8.3	3.5
8	145.3	8.5	604.1	18.8	2.9
9	176.2	10.8	636.8	23.0	3.1
10	158.1	8.7	580.8	18.1	2.7
11	125.5	7.2	477.9	15.6	2.4
12	99.7	5.0	305.3	10.4	2.5
13	124.1	7.5	490.4	17.0	2.8
14	130.6	7.9	518.0	16.9	2.1
15	112.7	6.2	440.0	14.9	2.4

Table 3. Comparison of the current PCB levels with the baseline PCB concentrations, and current PBDE levels

	Current	Baseline	
	X ± SD	X ± SD	P
Planar PCBs			
Sum of planar PCBs (pg/g fat)	74.0 ± 23.8	127.7 ± 37.3	<0.001
pg WHO-TEQ/g fat	3.8 ± 1.3	7.3 ± 2.5	<0.001
Total PCBs			
Sum of all PCBs (pg/g fat)	255.1 ± 96.7	471.1 ± 147.0	<0.001
pg WHO-TEQ/g fat	8.7 ± 2.8	15.7 ± 5.1	<0.001
Total PBDEs (ng/g fat)	2.40 ± 1.67	*	

* No measured

Table 4. Concentrations of PCBs (pg WHO-TEQ/g fat) and PBDEs (ng/g fat) in milk from women living in Tarragona County. Current and baseline data

	Current		Baseline	
	Urban (n = 7)	Industrial (n = 8)	Urban (n = 9)	Industrial (n = 6)
Planar PCBs				
Mean	4.5	3.2	6.5	8.3
SD	1.1	1.3	2.3	2.4
Minimum	2.8	1.3	2.4	5.0
Maximum	6.3	5.2	9.0	11.5
Total PCBs				
Mean	10.1	7.4	14.2	17.9
SD	2.8	2.4	4.7	5.1
Minimum	6.9	3.8	5.9	10.4
Maximum	13.3	10.7	19.2	24.4
Sum of PBDEs				
Mean	2.2	2.5	*	*
SD	1.9	1.5	*	*
Minimum	1.3	1.0	*	*
Maximum	6.6	5.6	*	*

*No measured

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