

## Dioxin and Dibenzofuran Concentrations in Human Milk Samples from Catalonia, Spain

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### Introduction

Because polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs) are lipid soluble, milk and adipose tissue, together with plasma, are frequently used as biomonitors of environmental exposure of humans to these organic pollutants (1,2). PCDD/Fs have been detected in milk from nonoccupationally exposed women at relatively high levels. Since the bioavailability of PCDD/Fs from breast milk is high, breast-fed infants can be potentially exposed to these highly toxic compounds. In relation to it, PCDD/F concentrations in breast milk were reported to decrease with duration of lactation and with the number of breast-fed children (1,2).

In recent years the levels of PCDD/Fs in human milk samples have been determined in a number of countries. However, information concerning the Spanish population is scarce (3,4) and there are no available data for the population of Catalonia (Spain). Nowadays, a hazardous waste incinerator (HWI) is being constructed in the area of Tarragona (southern Catalonia). To assess the impact of this new facility on public health, data about baseline levels of pollutants such as metals and PCDD/Fs are required. The present study was designed to determine the levels of PCDD/Fs in breast milk from women living in the area under potential influence of the new HWI.

### Materials and Methods

Fifteen milk pooled samples were obtained during 1996 from volunteer mothers living in the area of Tarragona (Catalonia, Spain). Six of them corresponded to women living in the vicinity of an industrial (petrochemical) zone, while the remaining 9 were pooled from women living downtown (urban area). All the participants in the study were primiparae mothers, between 25-35 years, who had lived in this area for at least the last 5 years. All mothers had a single child and infants were exclusively breast-feeding.

Pooled samples were homogenized and freeze-dried. Fat was extracted with a mixture of

diethyl ether and hexane after addition of sodium oxalate and ethanol. The solvent was exchanged to hexane and the fat content was determined. After addition of internal standards, aliquots of 1-1.5 g of fat were purified over a silica gel column. Samples were fractionated using activated carbon column with celite and cleaned with activated alumina columns. <sup>13</sup>C-labeled internal standards were used for quantification. A total of 16 congeners of PCDD/Fs were used to quantify PCDD/Fs. A gas-chromatography (Hewlett-Packard 5890) with fused silica capillary column, J&W Scientific DBDXN (60 m, ID 0.25 mm, film 0.15 μm) using SIR-mode and resolution of 10000, was used for PCDD/F analysis.

## Results and Discussion

Table 1 summarizes the individual levels (I-TEQ) of PCDD/Fs in the 15 human milk pooled samples, as well as the percentage of fat content in each sample. PCDD/F concentrations ranged between 5.9 and 17.1 pg I-TEQ/g fat (162-498 pg I-TEQ/l), with a mean value of 11.8 pg I-TEQ/g fat (310.8 pg I-TEQ/l).

Table 1: Individual levels of PCDD/Fs and percentages of fat in pooled samples of breast milk from mothers living in the area of Tarragona (southern Catalonia, Spain)

Sample code	pg I-TEQ/l	pg I-TEQ/g fat	Fat %
1	215	14.0	1.53
2	162	5.9	2.73
3	212	11.6	1.83
4	319	10.8	2.96
5	498	17.1	2.91
6	414	13.8	3.00
7	282	8.0	3.52
8	362	12.3	2.94
9	443	14.1	3.14
10	372	13.7	2.71
11	282	11.7	2.41
12	231	9.3	2.47
13	328	11.6	2.83
14	263	12.3	2.14
15	272	11.1	2.42

The levels of PCDD/F congeners, as well as the calculated mean I-I-TEQ and the maximum and minimum values for the 15 pooled milk samples are given in Table 2. It can be seen that PCDD levels diminished with the decreasing degree of chlorination. 2,3,7,8-TCDD was identified in all samples with a mean value of 1.04 pg/g fat. A different pattern was found for PCDFs. For these compounds, 2,3,4,7,8-PeCDF was the predominant congener, followed by 1,2,3,4,7,8-HxCDF.

Table 2: Levels (pg/g fat) of PCDD/F congeners in pooled samples of breast milk from mothers living in the Tarragona area (southern Catalonia, Spain)

CONGENER	Mean	SD	Minimum	Maximum
2,3,7,8-TCDD	1.04	0.07	0.59	1.53
1,2,3,7,8-PeCDD	4.02	0.92	1.93	5.62
1,2,3,4,7,8-HxCDD	2.84	0.57	1.27	3.32
1,2,3,6,7,8-HxCDD	27.88	1.54	15.10	38.00
1,2,3,7,8,9-HxCDD	4.55	1.34	2.26	7.53
1,2,3,4,6,7,8-HpCDD	38.26	24.54	9.36	118.00
OCDD	145.67	40.20	53.00	234.00
2,3,7,8-TCDF	0.68	0.22	0.33	1.11
1,2,3,7,8-PeCDF	0.31	0.12	0.12	0.61
2,3,4,7,8-PeCDF	7.95	2.11	3.85	11.00
1,2,3,4,7,8-HxCDF	3.06	0.50	1.91	3.87
1,2,3,6,7,8-HxCDF	2.49	0.48	1.49	3.44
1,2,3,7,8,9-HxCDF	0.04	0.02	ND	0.08
2,3,4,6,7,8-HxCDF	1.00	0.28	0.36	1.47
1,2,3,4,6,7,8-HpCDF	2.00	0.61	0.90	3.19
1,2,3,4,7,8,9-HpCDF	0.14	0.07	0.02	0.30
OCDF	0.34	0.16	ND	0.66
Total I-TEQ (PCDD/Fs)	11.82	2.71	5.93	17.10
Fat content (%)	2.63	0.52	1.53	3.52

PCDD/F levels in milk from mothers living in the industrial area were found to be slightly higher than those in women living downtown (12.7 and 11.2 pg/g fat, respectively). However, the differences were not statistically significant with the exception of 1,2,3,7,8,9-HxCDD, whose concentrations were significantly higher ( $p < 0.05$ ) in milk from mothers living in the industrial area than in those living downtown. Although a number of factors, especially dietary habits, can play an important role in the levels of PCDD/Fs in human milk, the highest PCDD/F concentrations in breast milk levels are generally found in polluted industrial areas.

While PCDD/F levels in milk from women living in the area of Tarragona are rather similar to those found in Norway (5), Germany (6) or Madrid (Spain) (4), they are lower than those obtained in Finland (7), France (4), The Netherlands (8), USA (9), or the Basque Country (Spain) (3), and higher than those found in Jordan (10) and Palestina (11).

According to the present results, the average PCDD/F intake of an infant of 5 kg ingesting 700 ml/day of breast milk would be 43.4 pg I-TEQ/kg/day. This value is 4.3 times greater than the tolerable daily intake of 10 pg I-TEQ/kg established by the WHO for the general

population. However, because of the relatively short nursing period and the rapidly increasing amount of adipose tissue of the infant, it has been pointed out that tolerable intake values established for lifetime exposures are not applicable for breast-fed infants.

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