Human Exposure P9

Levels of PCDD/Fs and PCBs in Human Milk in 1994 in Finland: Decrease in Concentrations from 1987 to 1994

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Introduction

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/F) are globally distributed toxic chemicals in the environment and were found in human milk in the 1980s (1,2). Food is the main source of PCDD/Fs and polychlorinated biphenyls (PCB) in humans (3). The decrease of concentrations of PCDD/Fs between 1986 and 1993 in human blood and milk has been reported from Germany and The Netherlands (4,5). The studies conclude that measures taken to reduce the PCDD/F and PCB emissions to the environment have resulted in a reduction of human body burdens of these compounds.

WHO/EURO has coordinated two rounds of follow-up studies on levels of PCDDs, PCDFs and PCBs in human milk. Finland participated in both of them (6,7). We are now reporting results of the second round randomly sampled primiparae human milk samples (42 samples) from Southern (14) and Eastern (28) Finland. Another objective in this study is to evaluate the trend of the concentrations of PCDD/Fs and PCBs in primiparae mother milk between 1987 and 1994.

Methods

The concentrations of 17 toxic PCDD/Fs, of three non-*ortho* (IUPAC 77, 126, and 169) PCB congeners, of five mono-*ortho* (IUPAC 105, 114, 118, 156 and 157) PCB congeners, and of 28 di-*ortho* (IUPAC 18, 28, 33, 47, 49, 51, 52, 60, 66, 74, 99, 101, 110, 122, 123, 128, 138, 141, 153, 167, 170, 180, 183, 187, 189, 194, 206 and 209) PCB congeners, the total sum of PCDD/F (\sum PCDD/F) and PCB (\sum PCB) congeners, and toxic equivalents, I-TEqs (TEqs, for PCBs) of them were determined from human milk samples.

ORGANOHALOGEN COMPOUNDS Vol. 38 (1998) About 40 ml of each human milk sample was analysed as described previously (8). Our laboratory has participated successfully in international quality control studies for the analysis of PCDDs and PCDFs in cow milk samples organized by EU/BCR-project in 1993 (9,10). Laboratory is also an accredited testing laboratory (No T77) in Finland (SFS-EN 45001 and ISO/IEC Guide 25).

Results and Discussion

Average ages of the primiparae mothers (27.9 years in the urban area and 27.0 years in the rural area) and average milk fat contents (3.75 % in the urban area and 3.88 % in the rural area) did not differ statistically significantly from each other in 1992-94. Furthermore, ages and milk fat contents did not differ statistically from those in our previous study (8).

Table 1. Selected PCDD/F concentrations, \sum PCDD/Fs and I-TEqs (mean \pm standard deviation as pg/g fat) in the primiparae mother's milk from the urban and rural area in Finland, in 1992-94 and in 1987. Asterisks indicate a statistically significant difference between urban and rural area in 1992-94 (*p<0.01, **p<0.005, ***p<0.001), and * indicate a statistically significant difference between 1987 and 1992-94 results (*p<0.01, **p<0.005, ***p<0.001).

Selected	Conc. pg/g fat in 1992-94		Conc. pg/g fat in 1987	
congeners	Urban area n=14	Rural area n=28	Urban area n=47	Rural area n=37
2,3,7,8-Cl ₄ DF	1.93 ± 0.74***	0.49 ± 0.44	2.98 ± 2.89	6.75 ± 4.29^{xxx}
2,3,7,8-Cl₄DD	2.66 ± 1.46	1.71 ± 0.68	3.37 ± 1.85	2.50 ± 1.25 ×
2,3,4,7,8-Cl ₅ DF	16.3 ± 7.0*	10.4 ± 4.65	20.1 ± 12.5	13.1 ± 5.41
1,2,3,7,8-Cl,DD	6.22 ± 2.16*	4.36 ± 1.56	9.78 ± 4.87 ***	7.53 ± 3.25 ***
1,2,3,6,7,8-Cl ₆ DD	33.2 ± 8.94	26.9 ± 8.16	48.2 ± 15.8 ***	41.5 ± 15.3^{xxx}
OCDD	230 ± 80.9***	126 ± 55.7	187 ± 83.6	171 ± 71.5 ***
∑ PCDD/F	381 ± 120***	217 ± 76.6	375 ± 132	339 ± 108 ^{xxx}
I-TEq	19.9 ± 7.42*	13.6 ± 4.57	26.3 ± 11.9	20.1 ± 6.54 xxx

Selected PCDD/F concentrations, \sum PCDD/F concentrations and I-TEqs in the primiparae mother's milk from the urban and rural area in 1992-94 and in 1987 are presented in Table 1 (8). The average \sum PCDD/F were in the urban area 381 pg/g fat (375 in 1987) and in the rural area 217 pg/g (339 in 1987). The average I-TEqs of primiparae mother milks were in the urban area 19.9 pg/g fat (26.3 in 1987) and in the rural area 13.6 pg/g (20.1 in 1987).

Primiparae mother milk I-TEq values of Helsinki (19.9 pg I-TEq/g fat) were similar to those measured in Europe (Belgium, Germany, The Netherlands and Spain), while values of Kuopio area (13.6 pg I-TEq/g fat) were similar to values measured in Norway, Austria

and eastern parts of Europe (7).

In Table 2 there are marker PCBs, \sum PCB concentrations and TEqs in the primiparae mother's milk from the urban and rural area in 1992-94 and in 1987. The average \sum PCB were in the urban area 296 ng/g fat (496 in 1987) and in the rural area 198 ng/g (396 in 1987). The average TEqs of primiparae mother milks were in the urban area 18.5 pg/g fat (37.0 in 1987) and in the rural area 11.6 pg/g (26.5 in 1987).

Sums of marker PCBs in urban area (192 ng/g fat) were similar to those measured in Croatia, Denmark, the Netherlands and Norway, while values of rural area (127 ng/g fat) were similar to values measured in the United Kingdom (7).

Table 2. Marker PCB concentrations, \sum PCB and TEqs (mean ± standard deviation as pg/g fat) in the primiparae mother's milk from the urban and rural area in Finland, in 1992-94 and in 1987. Asterisks indicate a statistically significant difference between urban and rural area in 1992-94 (*p<0.01, **p<0.005, ***p<0.001), and * indicate a statistically significant difference between 1987 and 1992-94 results (*p<0.01, **p<0.005, ***p<0.001).

Selected	Conc. ng/g fat	in 1992-94	Conc. ng/g fat	in 1987
congeners	Urban area n=14	Rural area n=28	Urban arean=47	Rural area n=37
PCB 28	2.39 ± 1.77	4.04 ± 4.09	9.92 ± 8.44 ***	6.40 ± 6.35
PCB 52	0.52 ± 1.60*	0.29 ± 0.19	0.81 ± 0.75^{xxx}	0.41 ± 0.32
PCB 101	1.20 ± 0.67***	0.38 ± 0.36	1.60 ± 1.39	0.52 ± 0.94
PCB 138	56.8 ± 23.0***	32.5 ± 12.7	82.8 ± 32.3 ^{xx}	74.4 ± 42.8 ***
PCB 153	92.3 ± 33.7**	57.7 ± 24.9	122 ± 52.1	102 ± 45.6 ***
PCB 180	39.2 ± 11.8	31.9 ± 17.5	61.3 ± 31.2 ***	45.6 ± 18.6 ^{xx}
∑ PCB	296 ± 108**	198 ± 80.8	496 ± 220 ***	396 ± 167 ***
TEq*	18.5 ± 7.48**	11.6 ± 5.03	37.0 ± 25.1 ***	26.5 ± 10.0 ***

[•]given as pg/g fat

When expressed as \sum PCDD/F, primiparae human milk concentrations were significantly higher in the urban area than in the rural area. The PCDD/F I-TEqs concentrations were higher in the urban than in rural area but statistically less significantly than \sum PCDD/F. \sum PCB and TEq concentrations were significantly higher among urban than rural primiparae.

The difference between urban and rural concentrations of PCDD/Fs and PCBs in primiparae mother milk remained when comparing current results with 1987 results (8). In fact, the differences in concentrations between areas have expanded. When the rural concentrations of

ORGANOHALOGEN COMPOUNDS Vol. 38 (1998) \sum PCDD/F, I-TEq, \sum PCB and TEq were 90, 76, 80, and 73 %, respectively, of the concentrations in urban area in 1987, the current percentages are 57, 68, 67 and 63, respectively.

The decrease in concentrations of PCDD/Fs between 1986 and 1993 in human blood and milk has been reported from Germany and The Netherlands (4,5). In this study, the decrease in I-TEqs of PCDD/Fs and TEqs of PCBs in primiparae mothers' milk was found to be 28 % and 53 %, respectively, when compared to the concentrations of the primiparae mothers' milk in 1987 (8). The declining of I-TEqs of PCDD/Fs and TEqs of PCBs seems to be greater in rural (32 and 56 %, respectively) than in urban (24 and 50 %, respectively) area. The estimation of 4-7 % for annual declining of PCDD/Fs and PCBs in this study is based on the results of the same laboratory with similar methods. There is not data available for the time trends of PCDD/Fs and PCBs in Finnish food between 1987-1994 and therefore the cause of the decrease of concentrations of PCDD/Fs and PCBs in human milk is unclear.

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References

1. Fürst P, Meenken H.A, Krüger Chr and Groebel W; *Chemosphere* 1987, 16, 1983 - 1988. 2. Lindström, G. (*Thesis*), Polychlorinated dibenzo-p-dioxins and dibenzofurans: Analysis of and occurrence in milk., University of Umeå, 1988, ISBN 91-7174-355-3.

3. Rappe C; Chemosphere 1992, 25, 231-234.

4. Päpke O, Ball M and Lis A; Chemosphere 1994, 29, 2355-2360.

5. Liem A.K.D, Albers J.M.C, Baumann R.A, van Beuzekom A.C, den Hartog R.S, Hoogerbrugge R, de Jong A.P.J.M and Marsman J.A; *Organohalogen Compounds* 1995, 26, 69-74.

6. Yrjänheikki, E.J., Ed. Levels of PCBs, PCDDs and PCDFs in breast milk: results of WHOcoordinated interlaboratory quality control studies and analytical field studies.

Copenhagen, FADL Publishers, 1989 (published on behalf of the WHO Regional Office for Europe, Environmental Series No. 34).

7. WHO/ECEH. Levels of PCBs, PCDDs and PCDFs in human milk. Second round od WHOcoordinated exposure study. Environmental Health in Europe 3. 1996, WHO, European Centre for Environment and Health.

8. Vartiainen T, Saarikoski S, Jaakkola J.J and Tuomisto J; Chemosphere 1997, 34, 2571-2583.

9. Rymen, T; Fresenius J Anal. Chem. 1994, 348, 9-22.

10. Schimmel H, Griepink B, Maier E.A, Kramer G.N, Roos A.H and Tuinstra L.G.M.T; Fresenius J Anal Chem. 1994, 348, 37-46