TREND STUDIES OF POLYCHLORINATED BIPENYLS, DIBENZO-p-DIOXINS AND DIBENZOFURANS IN HUMAN MILK

K. Norén and Å. Lundén
Department of Physiological Chemistry,
Karolinska Institutet,
Box 60 400, S-104 01 Stockholm, Sweden

ABSTRACT

Human milk has been used in studies of long-term changes in the concentrations of organochlorine contaminants in mothers from the Stockholm region. The previously observed decline (1972 to 1985) in the levels of pesticides, polychlorinated biphenyls, dibenzo-p-dioxins and dibenzofurans seems to have ceased for certain compounds since the levels were about the same in samples collected in 1985 and 1989.

INTRODUCTION

Several countries have taken measures to decrease the environmental contamination with organochlorine compounds. However, the persistent contaminants may circulate in the environment for long times after the usages are stopped or the discharges disminished. Also, the composition of the contaminants may change by decomposition and metabolism. Therefore, it was considered of interest to investigate the time related changes in the concentrations of specific congeners of polychlorinated biphenyls (PCBs), dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs) in human milk fat, which reflect the body burden of such contaminants.

SAMPLES

Milk was collected at the Mothers' Milk Centre in Stockholm in 1972, 1976, 1980, 1984-85 and 1988-89. The samples from 1972 to 1985 have been analysed previously (1,2) and reanalysed (3). The average age of the mothers delivering milk to the centre was 27-28 years in the different periods 1972 to 1985. Sixty per cent of the mothers nursed their first and most of the others nursed their second infant. In the period 1988-89 the average age of the mothers was 30 years and 55 per cent of the mothers were nursing their first infant. Equal amounts of milk from different mothers were mixed and the pooled samples were analysed.

METHODS

Ten ml of milk were extracted with the solid sorbent, Lipidex 5000 (4). Clean-up

and separations were made with aluminium oxide, silica gel and activated charcoal (4,5). The concentrations of pesticides and PCBs were determined by electron capture GC. Total PCBs were calculated according to original method using a packed column (6), otherwise capillary columns were used. Certain coplanar PCBs, polychlorinated dibenzo-p-dioxins and dibenzofurans were analysed by selected ion monitoring GC/MS at a resolution of 9000 (4,5).

RESULTS AND DISCUSSION

Milk from the Mothers' Milk Centre in Stockholm has been collected in different time periods since 1967 for analysis of organochlorine contaminants. A continuous decline in the levels of PCBs in the milk was observed from 1977 to 1985 (3,4). The average levels from this period were 0.93 (1976/77), 0.88 (1978), 0.79 (1979), 0.76 (1980), and 0.60 µg/g fat (1985). However, this trend seems to have ceased and the levels in samples collected in 1988-89 were about the same (0.65 µg/g fat) as in samples from 1984-85 (0.60 µg/g fat). Comparisons of the relative composition of PCBs indicate that the percentage of PCB 118 (2,3',4,4',5-pentachlorobiphenyl) is decreasing and the percentages of PCB 153 (2,2',4,4',5,5'-hexachlorobiphenyl) and 180 (2,2',3,4,4',5,5'-heptachlorobiphenyl) are increasing with the year of collection (Table 1). This may indicate that the profile of PCBs to which the mothers are exposed may have changed during the years studied. Bühler et al. (7) have determined the half-life times of the congeners 108/118, 138, 153 and 180 in man and found them to be 100-300, 321, 338 and 124 days, respectively. The compounds with the longest half-life times predominate in the samples analysed.

The levels of the coplanar PCBs 77 (3,3',4,4'-tetrachlorobiphenyl), 126 (3,3',4,4'5-pentachlorobiphenyl) and 169 (3,3',4,4',5,5'-hexachlorobiphenyl) have declined from 1972 to 1985, but this trend is no longer seen from 1985 to 1989 (Table 2). Similarly, the decline in the levels of PCDDs and PCDFs from 1972 to 1985 (3) has been discontinued and the samples from 1988/89 have levels similar to those in the samples from 1985 or higher (Table 3). The results of these studies indicate that the exposure of mothers to PCBs, PCDDs and PCDFs in the Stockholm area has not decreased since 1985.

ACKNOWLEDGEMENTS

This study was supported by grants from the Swedish Environmental Protection Agency, contract Nr 5326188-9, the Swedish Medical Research Council (03X-219), Stiftelsen Hiertas Minne and Karolinska Institutet.

Table 1. Relative concentrations of selected PCB congeners (per cent of total PCBs) in human milk.

Year	1972	1976	1980	1988/85	1988/89
Number of mothers	135	153	431	102	140
PCB congener					
28	3.1	2.2	3.8	3.8	1.3
52*	2.1	2.2	2.5	2.7	3.7
118	5.2	4.4	4.0	4.0	3.9
153	18.7	19.0	20.0	20.7	23.2
105	1.3	1.6	1.0	1.2	1.0
138	16.6	17.2	17.2	17.9	17.8
156	1.7	1.8	1.7	2.1	2.2
180	7.6	8.1	8.3	8.8	9.9

* Interference possible.

Table 2. Average levels, pg/g fat, of non-ortho substituted coplanar PCBs in human milk. Ranges are given in parentheses.

Year	1972	1976	1980	1984/85	1988/89
Number of mothers	195	204	431	102	140
Number of samples	3	3	4	2	7
Fat concentration g/100g	2.7 (2.6-2.8)	2.7 (2.7-2.8)	2.9 (2.9-3.1)	2.8 (2.6-2.9)	2.6 (2.2-2.9)
PCB congener					
77	76 (65-88)	41 (27-62)	29 (27-33)	38 (36-40)	27 (19-47)
126	298 (260-355)	253 (226-297)	166 (141-183)	104 (94-114)	98 (78-122)
169	67 (66-69)	74 (71-78)	65 (59-77)	44 (38-50)	47 (40-54)

Table 3. Average levels, pg/g fat, and ranges of PCDDs and PCDFs in human milk.

Year	1972	1976	1980	1985	1989
Number of mothers	227	245	340	102	100
Number of samples	4	4	4	7	5
Fat concentration g/100g	2.9 (2.7-3.2)	2.8 (2.7-3.0)	3.0 (2.9-3.1)	2.6 (2.2-2.9)	2.6 (2.2-2.9)
2,3,7,8-TCDD	5 (4-6)	5 (4-5)	3 (2-3)	1 (1-2)	3 (2-3)
1,2,3,7,8-PeCDD	9 (7-11)	7 (6-11)	6 (5-8)	5 (2-8)	7 (6-9)
1,2,3,6,7,8-HxCDD	45 (35-52)	40 (35-52)	31 (26-34)	30 (20-47)	38 (24-51)
1,2,3,4,6,7,8-HpCDD	119 (109-139)	96 (80-107)	70 (64-78)	69 (42-91)	57 (35-73)
OCDD	458 (356-520)	371 (325-405)	338 (327-344)	244 (217-285)	268 (249-313)
2,3,7,8-TCDF	4 (3-6)	3 (2-4)	3 (2-4)	2 (1-2)	2 (1-2)
2,3,4,7,8-PeCDF	32 (29-34)	29 (26-34)	17 (13-20)	14 (10-22)	17 (16-18)
1,2,3,6,7,8-HxCDF	14 (12-16)	14 (12-17)	8 (8-8)	8 (4-10)	7 (6-10)
1,2,3,4,6,7,8-HpCDF	24 (16-32)	21 (13-28)	7 (6-12)	8 (5-16)	8 (6-11)
OCDF	6 (3-7)	4 (3-5)	5 (3-6)	5 (2-7)	2 (1-3)

REFERENCES

1. Westöö G. and Norén K., Ambio, 7, 62-64 (1978).
2. Norén K., Acta Paediatr. Scand., 72, 259-264 (1983).
3. Norén K., Chemosphere, 17, 39-49 (1988).
4. Norén K. and Sjövall J., J. Chromatogr. Biomed. Appl., 422, 103-115 (1987).
5. Norén K., Lundén Å., Sjövall J. and Bergman Å., Chemosphere, to be published.
6. Norén K., Arch. Environm. Contam. Toxicol., 12, 277-283 (1983).
7. Bühler F., Schmid P. and Schlatter Ch., Chemosphere, 17, 1717-1726 (1988).