Bull. Environ. Contam. Toxicol. (1993) 50:425–432

Environmental Contamination and Toxicology

Organochlorine Pesticide and PCB Congener Content of French Human Milk

F. Bordet, J. Mallet, L. Maurice, S. Borrel, and A. Venant

Centre National d'Etudes Veterinaires et Alimentaires, Laboratoire Central d'Hygiène Alimentaire, 43 Rue de Dantzig 75015, Paris, France

There are no recent data concerning the contamination of human milk by organochlorinated (O.C.) compounds in France. In Europe most of foodstuffs are subject to control in order to limit the presence of organochlorine compounds and particularly the pesticides, in conformity with European Economic Community recommendations. The Maximal Levels of pesticide Residues (MLR) in dairy products are given in a Directive (24 July 1986) of the Commission of European Communities.

As the organochlorine pesticides have not been used in France for twenty years, their presence in dairy products has decreased gradually to the actual level which is near the detection limit (Venant et al., 1991). However, the O.C. pesticides are still used by some countries for agricultural and sanitary purposes. As these compounds are hardly degraded in the environnement and are easily soluble in fat, they tend to be bioaccumulated. They are transmitted to humans by consumption of contaminated products or by direct contact. In fact, several studies have shown that milk secretion is the most important means of excretion of O.C. in women. Consequently the infants being breast fed are exposed to these residues present in their mother's milk. (Coilins et al. 1982, Noren 1983, Mes et al. 1984, Safe et al. 1985, Mes et al. 1986).

A first French study (Auber 1986) indicates that human milk is highly contaminated with O.C. pesticides. In a second study realized in our laboratory (results presented in this paper), we have determined the levels of 13 O.C. pesticides and total PCBs in 30 samples of human milk. As expected and in accordance with similar studies in other countries (Mes et al. 1987, Galetin-Smith et al. 1990, Krauthacker et al. 1991, Sasaki et al 1991), we have found positive results. These findings prompted us to look more precisely at this question and to determine the level of individual PCBs in human milk. 20 other samples of human milk were tested for the presence of 17 congeners of PCB : 28, 52, 44, 66, 101, 99, 77, 118, 153, 105, 138, 126, 187, 156, 180, 169, 203 (IUPAC nomenclature) and 13 O.C. pesticides : α, β and γ HCH, HCB, aldrin, heptachlore-epoxide, α and γ chlordane , dieldrin, endrin, DDE, TDE, DDT.

MATERIALS AND METHODS

30 samples of human milk were analysed for total PCBs and O.C. pesticides in 1990 and 20 other samples were analysed for PCB congeners and O.C. pesticides

Send reprint requests to F. Bordet at the above address.

in 1991. The samples were collected manually in glass bottles and covered with aluminium foil, both previously tested for contamination and stored at -20°C until analysed. Milk was collected from 2 to 90 days after parturition, and each donor answered a questionnaire about age, time of collect, number of infants fed and diet.

Gas chromatographic analysis has been performed using a Varian Model 3400. Chromatograph determination of OCs and PCBs was carried out using a 50 m, 0,32mm, 0,25 µm glass capilary column CP SIL 8 CB from Chrompack. The initial column temperature was 100°C, held for 2mn after the injection, programmed from 100°C to 220°C at 20°C/min, held during 10 min, programmed from 220°C to 260°C at 3°C/min and held during 15 min. The mobile phase was Helium (C quality) at 2,5 ml/min. The on-column injector was programmed from 50°C to 200°C at 150°C/min. The 63-Ni electron capture detector's temperature was 300°C with a make up of Nitrogen flow at 30 ml/min.

All solvents used were pesticide analytical grade reagents free of interfering residues as tested by gas chromatography (GC). All glasswares used for the analysis were rinsed with n-hexane before use.

Standards of PCBs and organochlorine pesticides were obtained from Accustandard and Reidel de Haenn. One standard solution was prepared for 13 pesticides (α, β, γ HCH, HCB, aldrin, heptachlore-epoxide, α and γ chlordane, dieldrin, endrin, DDE, TDE, DDT) and another for 17 PCBs (28, 52, 44, 66, 101, 99, 77, 118, 153, 105, 138, 126, 187, 156, 180, 169, 203, IUPAC nomenclature).

Table 1. Recoveries of O.C. pesticide and PCB solutions used as standard.

O.C. Pesticides	% of recovery	PCB	% of recovery
αHCH	79	28	106
HCB	69	52	92
βHCH	97	44	90
γHCH	95	66	115
Aldrin	79	101	94
Heptachlore epoxide	68	99	93
γChlordane	90	77	93
αChlordane	88	118	97
PP' DDE	81	153	95
Dieldrin	55	105	96
Endrin	37	138	94
PP' TDE	95	126	115
PP' DDT	90	187	91
		156	95
		180	93
		169	106
		203	90

The extraction and purification procedures are already described (Venant et al., 1986). Percentage of recovery for each molecule of O.C. pesticides and PCBs is presented in table 1.

Table 2. Concentration (ng/g of milk fat) of PCB congeners in 20 human milk samples .

PCB	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	M1
28	*	*	*	*	*	*	*	*	11	84	45	20	84	42	nd	nd	35	30	7	16	31
52	80	217	12	230	40	nd	170	40	17	nd	31	48	63	nd	397	nd	nd	20	nd	nd	68
44	nd	20	90	60	50	nd	nd	90	12	nd	26	nd	175	nd	nd	210	15	25	13	19	40
66	nd	60	10	nd	30	nd	110	nd	76	470	30	130	326	196	772	317	465	90	58	55	159
101	nd	30	10	nd	40	9	20	30	33	131	50	25	nd	73	nd	98	nd	nd	48	6	30
99	nd	nd	6	nd	30	30	20	30	15	230	18	20	*	57	452	118	20	5	10	nd	56
77	nd	nd	nd	nd	nd	nd	nd	nd	40	49	57	16	nd	40	258	48	85	70	82	29	39
118	nd	nd	nd	nd	nd	nd	nd	nd	43	140	78	32	22	89	95	130	20	10	79	15	37
153	10	nd	10	20	nd	nd	nd	nd	26	145	32	92	127	86	684	234	70	35	4	22	80
105	*	*	*	*	*	*	*	*	nd	nd	nd	nd	37	nd	nd	nd	30	20	nd	nd	7
138	100	nd	30	20	50	nd	30	nd	94	22	397	148	330	281	45	331	50	25	9	20	99
126	70	70	80	140	10	140	150	130	nd	nd	4	nd	63	42	nd	nd	nd	nd	nd	nd	45
187	10	190	90	20	10	20	nd	nd	5	33	9	11	nd	18	33	30	7	5	16	1	25
156	10	nd	nd	20	10	20	20	20	6	27	2	16	5	20	39	35	5	2	2	1	13
180	10	250	30	110	70	130	120	100	31	164	2	143	nd	152	234	227	30	15	237	5	103
169	10	*	nd	140	*	nd	80	*	3	nd	nd	nd	187	37	nd	nd	20	3	4	3	29
203	nd	200	nd	30	20	nd	80	10	3	28	12	13	nd	18	43	24	nd	1	4	4	24
M2	300	837	368	790	370	349	800	440	414	1968	793	714	4090	1150	3052	1802	852	356	573	196	→1010
F%	2	.5	2.5	1.5	1.5	.5	.5	1	2.31	.33	1.98	1.5	.95	1.2	.2	.45	.8	1.98	3.3	3.3	
M3	600	419	920	1185	555	**175**	400	440	956	649	1570	1071	**3885**	1380	610	811	682	705	1891	647	**→978**

M1 : Average level per congener in ng/g of milk fat
M2 : Average level per sample in ng/g of milk fat
M3 : Average level per sample in ng/100 ml of milk
F% : % of milk fat/milk volume extracted x 100
* : No value nd : Below detection limit

Table 3. Concentration (ng/g of milk fat) of O.C. pesticides in 20 human milk samples.

O.C.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	M1
αHCH	42	20	55	153	25	15	107	122	5	13	4	6	453	10	nd	nd	1	nd	3	2	52
HCB	84	49	15	59	36	3	10	56	107	*	451	449	577	9	217	322	51	55	223	12	147
βHCH	57	159	7	310	121	159	22	106	144	665	718	403	19	59	335	282	18	1323	695	142	287
γHCH	8	62	25	27	17	73	49	47	24	69	23	40	12	22	132	55	17	19	16	11	37
Aldrin	0.4	nd	80	51	nd	39	82	39	4	nd	8	6	59	nd	32	70	6	8	nd	1	24
H.E.	1	27	5	52	8	13	11	23	34	575	86	83	208	15	441	317	24	nd	3	6	97
γChlordane	nd	nd	nd	nd	27	nd	nd	nd	7	nd	15	nd	27	6	28	nd	nd	nd	8	1	6
αChlordane	nd	112	66	69	7	nd	100	10	37	116	3	53	27	32	266	606	8	37	14	2	78
DDE	530	359	1117	252	170	1332	959	372	790	9556	1181	1625	81	2006	18531	1915	262	782	1110	743	2183
Dieldrin	nd	nd	nd	nd	nd	nd	nd	nd	184	1357	429	143	nd	103	650	267	169	143	271	79	190
Endrin	nd	nd	nd	nd	nd	nd	nd	nd	36	nd	33	nd	842	28	nd	122	nd	17	65	20	58
TDE	nd	nd	nd	nd	nd	nd	nd	nd	5	24	5	7	55	10	59	15	192	3	3	38	21
DDT	44	nd	1	1	8	61	nd	nd	8	246	26	49	784	13	75	239	9	3	8	14	79
M2	770	676	1342	648	448	1708	1340	775	1385	12621	2982	2864	3144	2313	20766	4210	757	2390	2419	1071	→3231
F%	2	.5	2.5	1.5	1.5	.5	.5	1	2.31	.33	1.98	1.5	.95	1.2	.2	.45	.8	1.98	3.3	3.3	
M3	1540	**338**	3355	972	672	854	670	775	3199	4165	5904	4296	2987	2776	4153	1895	606	4732	**7983**	3534	**→2770**

M1 : Average level per congener in ng/g of milk fat
M2 : Average level per sample in ng/g of milk fat
M3 : Average level per sample in ng/100 ml of milk
F% : % of milk fat/milk volume extracted x 100
H.E. : Heptachlore epoxyde nd : Below detection limit * : No value

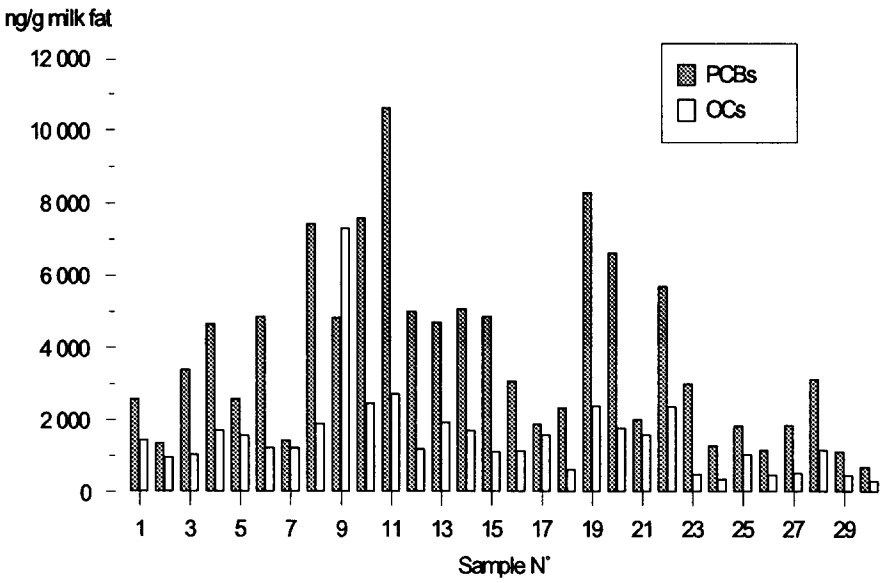

Figure n° 1 : Individual variation for total PCBs and total OCs level in breast milk (1990).

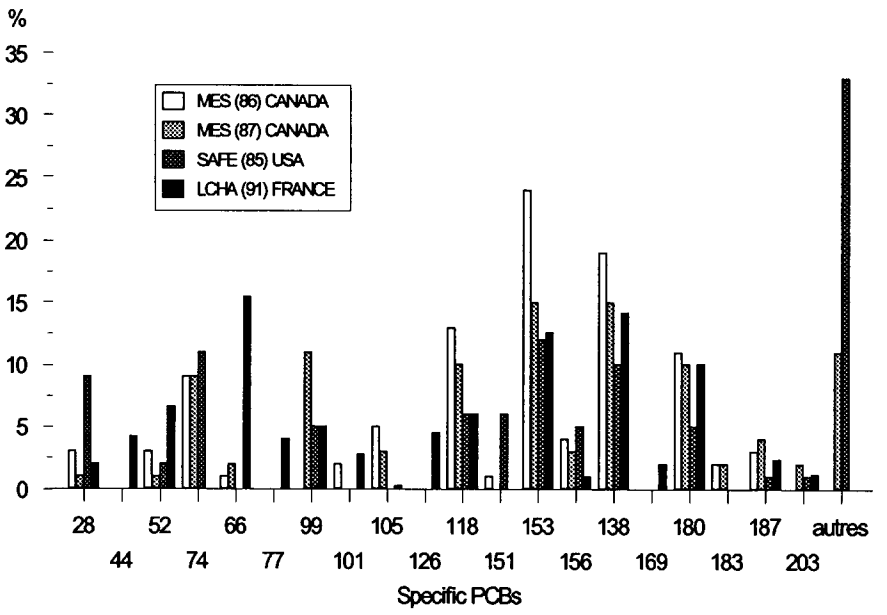

Figure n° 2 : A comparison of specific PCB relative contents in human milk from different surveys

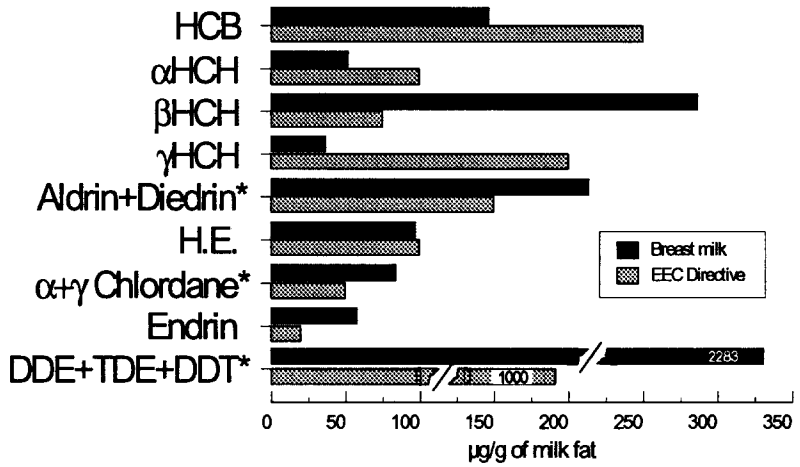

Figure n° 3 : Comparison between pesticide content in 20 breast milk samples and EEC Directive level in milk products

H.E. : Heptachlore epoxyde

* : Level products montionned together in EEC Directive :

- Aldrin + Dieldrin (150 µg/g)
- α + γchlordane (50 µg/g)
- DDE + TDE + DDT (1000 µg/g)

RESULTS AND DISCUSSION

Figure n° 1 shows the level of contamination of 30 samples of milk by total PCBs and total O.C. pesticides. The raw data concerning the amount of PCB congeners and O.C. pesticides in 20 samples are presented in table 2 and 3. Usually the authors express the level of contamination in ng/g of milk's fat. As the milk's fat varies from 0.2 to 3%, it would be better to give the rate of contamination as ng/100 ml of milk, in order to have a more precise appreciation of the contamination specially when the milk has a low percentage of fat and a high level of contamination (samples n°10 or 15 table 3).

At present time there is no European recommendation about the minimal level of individual or total PCBs in food, but we can compare our results with the maximal level of PCBs tolerated by swiss legislation (0,5 ppm) and notice that most of our samples exceed these values (Figure 1 and Table 2).

Comparing our results with published data on the subject (figure n° 2), it can be seen that some congeners show a high rate of contamination in several countries. The most frequent congeners are PCBs 118, 153, 138 and 180. Each of them represents 10 to 20% of all PCBs. We have also found PCB 66 at a rate of 15 %. The two PCBs 153 and 138 are mentioned in the International Dairy Federation standard 130:1986 for their analytical and toxicological significance. Among the 209 PCBs, the most toxic ones are 77, 126 and 169 which are present in French human milk at a level of 39, 45 and 29 ppb respectively (table 2).

O.C. pesticide levels in human milk could be compared with the Maximal Admissible Levels recommended in EEC Directive for cow's milk and dairy products (Figure 3). Only HCB, α HCH and γ HCH are within the acceptable range and DDE contamination alone is often present above the level of 1 ppm authorized by the Directive for the total DDE, DDT and TDE together. Table 2 shows that 60 % of our samples are contaminated by PCBs (level = 0.5 ppm) and table 3 shows that 70 % of them are contaminated by betta HCH. All of the samples have at least one pesticide residue above the maximal autorized level. Only 25 % of the samples are contaminated by DDE, but the level of sample contamination by this residue is high.

We were not able to find a direct correlation between the time of milk collection and the rate of contamination. Samples n°5, n°9 and n°17, collected at 5 days, 30 days and 90 days after birth, are contaminated at the same level of total PCBs.

Table 4. Infant intake of PCBs and OC pesticidesduring the first three months of lactation

	mg / 3 months			µg / day / infant
	mini	maxi	average	average
OCs	0.18	4.30	1.50	16.7
PCBs	0.09	2.10	0.53	5.9

It is difficult to estimate accurately the amount of intake of each of these compounds by an infant, as it depends on the body burden of the mother, the percentage of milk fat, the quantity of milk intake and so on. According to Galetin-Smith et al. (1991) the daily intake of PCBs for one infant varies from 7,5 µg/day to 17 µg/day, and the daily intake of total DDTs varies from 8,7 to 11,4 µg/day. Residues are not constant during the whole lactation. Since we have collected samples at different times of lactation over a period of 3 months, the average amount of residues (PCBs and pesticides separately) in these samples could be taken as an estimate of the quantity ingested by each infant. We also calculated the highest and lowest levels on the basis of daily intake of 600 g of milk (table 4).

Acknowledgments. The authors thank Beaudeloque's Hospital, Paris-Brune Lactarium and all mothers for the supply of human milk samples.

REFERENCES

Auber C, (1987) Enquête de "Terre vivante" : Pollution du lait maternel. Les quatre saisons, N°42 : 33-39

Coilins GB, Holmes DC, Hoodless RA, (1982) Organochlorine pesticide residues in human milk in Great Britain, 1979-80. Human Toxicol 1, 425-431

Galetin-Smith R, Pavkov R, Roncevic N, (1990) DDT and PCBs in human milk : implication for breast feeding infants. Bull Environ Contam Toxicl 45 : 811-818

Krauthacker B, (1991) Levels of organochlorine pesticides and polychlorinated biphenyls (PCBs) in human milk and serum collected from lactating mothers in the northern Adriatic area of Yugoslavi. Bull Environ Contam Toxicol 46 : 797-802

Mes J, Marchand L, (1987) Comparison of specific polychlorinated biphenyl isomers in human and monkey milk. Bull Environ Contam Toxicol 39 : 736-742

Mes J, Doyle JA, Adams BR, Davies DJ, Turton D, (1984) Polychlorinated biphenyls and organochlorine pesticides in milk and blood of canadian women during lactation. Arch Environ Contam Toxicol 13 : 217-223

Mes J, Davies DJ, Turton D, Sun WF, (1986) Levels and trends of chlorinated hydrocarbon contaminants in the breast milk of canadian women. Food Add Contam 3 : 313-322

Mes J, Turton D, Davies DJ, Sun WF, Lau PY, Weber D, (1987) The routine analysis of some specific isomers of polychlorinated biphenyl congeners in human milk. J Environ Anal Chem 28 : 197-205

Noren K, (1983) Levels of organochlorine contaminants in human milk in relation to the dietary habits of the mothers. Acta Paediatr Scand 72 : 811-816

Safe S, Safe L, Mullin M, (1985) Polychlorinated biphenyls : congener-specific analysis of a commercial mixture and human milk extract. J Agr Fd Chem 33 : 24-29

Venant A, Borrel S (1987) Protocole de préparation d'échantillons de graisse végétales et animales pour la détermination de résidus de composés organo-chlorés. Analusis 3 :145-147

Venant A, Borrel S, Mallet J (1991) Evolution de la contamination du lait et des produits laitiers par les résidus de composé organo-chlorés. Lait 71 : 107-116

Received June 4, 1992; accepted September 22, 1992.