

## **Levels of Organochlorine Pesticides and Polychlorinated Biphenyls (PCBs) in Human Milk and Serum Collected from Lactating Mothers in the Northern Adriatic Area of Yugoslavia**

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Organochlorine pesticides and polychlorinated biphenyls (PCB) are persistent contaminants of the environment. Owing to their lipophilic properties they are primarily stored in fat-rich tissues and fluids of humans and animals. The results of a monitoring study on levels of the DDT-complex, HCH-group of isomers, hexachlorobenzene (HCB) and PCB in human milk and serum are presented in this paper. Samples were collected in the northern Adriatic area for which no data have so far been available. Sample donors were chosen from rural and small urban locations where mediterranean eating habits are still maintained. Our previous studies were conducted in an industrialized continental town of Croatia (Krauthacker et al. 1980, Krauthacker et al. 1986, Krauthacker et al. 1989).

### **MATERIALS AND METHODS**

Samples were collected from two groups of mothers: one living on the island Krk and the other living in Labin, a small town on the peninsula Istria. In Krk milk samples came from 33 primiparae and multiparae (age: 20-40 yr) during winter 1986/87. In Labin milk and serum samples were collected from 10 primiparae (age: 21-34 yr) in winter 1989; serum samples were taken once, and milk samples twice (seven days apart). Milk sampling was done by manual expression into glass containers and samples were kept frozen until analysis. Blood was sampled by venipuncture, and the serum (separated by centrifugation) was kept frozen until analysis. For extraction of milk samples a chloroform/methanol mixture (1:1) was used and after fat content determination the hexane fat solution was cleaned with conc. sulphuric acid (Krauthacker et al. 1986). Serum samples were extracted with the same procedure as described previously (Krauthacker et al. 1980), but the hexane

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extracts were cleaned with conc. sulphuric acid instead of with florisil. Gas chromatographic analysis was done on a Varian 1440 with a  $^3\text{H}$  Sc electron capture detector, a Pye Unicam 204 with a  $^{63}\text{Ni}$  electron capture detector or a Tracor 550 with  $^{63}\text{Ni}$  electron capture detector. The following packed columns were used: a) 1.3% SF-96 + 5.3% QF-1 on Supelcoport 0.131-0.157 mm, b) 5% OV-101 on Chromosorb W DMCS/AW 0.157-0.197 mm, c) 3% OV-1 on Chromosorb W HP 0.157- 0.197 mm and d) 1.5% OV-17 + 1.95 % OV-210 on Gas Chrom Q 0.157-0.197 mm. All samples were chromatographed on two different columns. Only the compounds identified on both columns were evaluated. The compounds were identified by their retention times as compared to known standards. Total PCB were measured on columns a and b, and compared to Aroclor 1260 standard. Quantitation was done comparing the peak heights in the sample with those in corresponding standards. The PCB were quantitated by summation of peak heights from seven major peaks when samples were chromatographed on column a, and from twelve major peaks on column b. All concentrations for each individual sample were the mean values of those determined on two columns. The recoveries were 62-92 % for milk and 40-89 % for serum depending on the compound determined. All results were corrected for recoveries. Determination limits were 1  $\mu\text{g/L}$  for HCB, HCH-isomers or DDT complex, and 2  $\mu\text{g/L}$  for PCB.

## RESULTS AND DISCUSSION

Milk and serum samples were analysed for HCB, alpha-, beta- and gamma-HCH, p,p'-DDE, p,p'-DDD, p,p'-DDT and PCB. The results are given in Table 1. Concentrations are expressed per volume of serum, on whole milk basis and on milk fat basis. All concentrations are given as medians and ranges, because they are not normally distributed. All milk and serum samples contained p,p'-DDE and PCB. The frequency of HCB and beta-HCH was also high. Alpha-HCH was not found in any milk sample, but it was found in eight (out of ten) serum samples. The presence of p,p'-DDT and p,p'-DDD was not detected in any serum sample. The frequency of HCB and p,p'-DDT in the milk samples from Krk was higher than in those from Labin. There was no correlation for p,p'-DDE and PCB concentrations in serum vs milk collected in Labin ( $r = 0.29$  for p,p'-DDE,  $r = 0.55$  for PCB).

The literature data concerning concentrations of compounds in milk samples from primiparae as against multiparae are contradictory. Slorach and Vaz (1983) claimed primiparae to have higher concentrations of organochlorine compounds in milk than multiparae, whereas no difference was reported by Drijver and co-workers (1988). Our data are evaluated in Table 2.

Table 1. Concentrations of organochlorine compounds in human milk and serum determined in two population groups. Results are corrected for recoveries.

COMPOUND	MILK ( $\mu\text{g/kg}$ )		SERUM ( $\mu\text{g/L}$ )		
	Krk (N=33)	Labin (N=20)	Labin (N=10)		
	Milk basis	Fat basis	Milk basis	Fat basis	
HCb					
-Median	2	100	ND	ND	2
-Range	1-4	50-170	1-3	40-110	1-4
-n	30	30	5	5	10
alpha-HCH					
-Median	ND	ND	ND	ND	2
-Range	-	-	-	-	1-2
-n	0	0	0	0	8
beta-HCH					
-Median	3	100	2	50	18
-Range	1-11	50-420	1-5	30-200	13-31
-n	28	28	14	14	8
gamma-HCH					
-Median	ND	ND	ND	ND	ND
-Range	1-2	30-200	1-2	30-40	21
-n	3	3	4	4	1
p,p'-DDE					
-Median	32	1080	17	550	6
-Range	6-124	520-6610	5-46	240-1740	4-13
-n	33	33	20	20	10
p,p'-DDD					
-Median	ND	ND	ND	ND	ND
-Range	2-5	110-210	-	-	-
-n	2	2	0	0	0
p,p'-DDT					
-Median	ND	ND	ND	ND	ND
-Range	1-13	40-410	-	-	-
-n	13	13	0	0	0
PCB					
-Median	15	500	9	270	7
-Range	4-44	210-1620	3-14	80-580	6-1
-n	33	33	20	20	10
Fat content (%)					
-Median	3.0		2.9		Not
-Range	0.6-7.0		2.2-4.2		deter.

ND=below detection limit, N=number of analysed samples, n=number of positive samples. Ranges apply to positive samples.

No difference concerning parity was found for p,p'-DDE, PCB, HCB and beta-HCH, which occurred most frequently in the studied population. Likewise, no difference was found between concentrations from two samplings; this

applied to individual samples and to a group of twenty samples from ten mothers (Table 2). Noren (1983) found that concentrations decrease very little even up to six months lactation.

Table 2. Concentrations of compounds found in primiparae vs multiparae, and in samples collected in first sampling vs second sampling (seven days later). Results are expressed in  $\mu\text{g/kg}$  milk fat and are corrected for recoveries.

COMPOUND	KRK		LABIN	
	Primiparae N=15	Multiparae N=18	1st Sample N=10	2nd Sample N=10
HCB				
-Median	80	100	ND	ND
-Range	50-140	60-170	40-110	40-70
-n	14	16	2	3
beta-HCH				
-Median	80	110	50	60
-Range	50-420	50-200	50-200	30-140
-n	13	15	6	8
p,p'-DDE				
-Median	1050	1080	520	580
-Range	550-6610	520-3320	240-1740	290-1210
-n	15	18	10	10
PCB				
-Median	460	570	260	290
-Range	210-650	330-1620	120-410	80-580
-n	15	18	10	10

ND=below detection limit, N=number of analysed samples, n=number of positive samples. Ranges apply to positive samples.

Jensen (1983) calculated the maximum tolerable concentrations for PCB, DDT complex, HCB and gamma-HCH in human milk from acceptable daily intakes (ADI) for these compounds. This calculations were based on the assumption that a 5-kg infant consumes 800 g milk per day. If Jensen's values are compared with our median concentrations (Table 1) it follows that our milk samples contained 2.5-times higher PCB than the calculated "tolerance" values, while the concentrations of the other compounds were equal to or smaller than the "tolerance" concentrations.

The concentrations and the incidence of all compounds analysed in this study in milk and serum were within the same range as those reported recently for different Yugoslav population groups and for populations in industrialized countries (Bush et al. 1984; Jan and Tratnik 1988; Jensen 1983; Jensen 1989; Krauthacker et al. 1986; Krauthacker et al. 1989; Krawinkel et al.

1989; Pavkov et al. 1987; Rončević et al. 1987; Skaare et al. 1988; WHO 1989)

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