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Organochlorine Pesticides, HCB, and PCBs in Human Milk in Poland

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Persistent organochlorine compounds were used in agriculture and industry for years. Their ability to accumulate in organisms constituting links of the food chain (Dewailly *et al*. 1993, Ludwicki and Góralczyk 1994) and a unique chemical stability made them a hazardous environmental contaminants. The metabolism and excretion of chlorinated hydrocarbons is a very slow process. One of the most important means of elimination such compounds from the woman's body is lactation. Consequently, human milk has occasionally a significant concentration of organochlorine compounds (Dewailly *et al*. 1989). Such compounds are identified in women's milk all over the world.

The control of organochlorine compounds levels in the human milk is of particular importance as infants and little children fall into the high-risk category. This is due to the fact that they do not yet have fully developed detoxification mechanisms and their organs are in the process of rapid growth.

The objective of the present study was to identify organochlorine pesticides, HCB, and PCBs concentrations in human milk in Poland by comparing more and less industrialized regions. Subject to examination were also differences in the concentrations of these compounds in mature milk vs. that collected on the fourth day following delivery. This comparison seemed interesting since some authors use the transitory milk (up to one week after delivery) when mothers are still in the hospital. The concentrations of organochlorine compounds in such milk do not necessarily reflect the actual intake by children fed for considerably longer time.

MATERIALS AND METHODS

The study material was human milk. Samples were collected in a lactarium, and maternity clinic in Warsaw, and from donors in different regions of the country.

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The milk samples (10 ml) were kept frozen at -18°C until analysis. After defrosting, the samples were deproteinized with acetone and extracted with n-hexane, and the resulting extract was cleaned up using concentrated sulphuric acid. The purified extracts were then analyzed by GLC-ECD for organochlorine insecticides and HCB.

Following the detection of organochlorine insecticides and HCB, further analysis focused on quantitation of polychlorinated biphenyls. The hexane extracts collected were concentrated to a 1 ml volume and dehydrochlorinated using a 2.5% potassium hydroxide solution in a 96% ethanol. Following extraction with hexane, the samples were oxidized using an oxidizing agent (1.6 g of potassium bichromate, 18.5 ml of distilled water, and 100 g of concentrated H_2SO_4). PCBs were determined in the hexane layer by GLC-ECD.

Gas chromatography analysis was done on a Pye-Unicam 104, equipped with ^{63}Ni electron capture detector (ECD). Glass chromatographic column (1.8 m, 4 mm internal diameter) was packed with 7.5% QF-1 and 3% OV-17 (9: 1) on Gas Chrom Q (80-100 mesh). The operating conditions were: injection port and column temperature: 200°C, detector temperature 220°C; the carrier gas was nitrogen at a flow rate of 40 ml per minute and sample volume 5µl.

RESULTS AND DISCUSSION

97 milk samples were examined to compare mean concentrations of the compounds found in women's milk collected on the fourth day after delivery and in mature milk collected from the third week of lactation onwards. The findings are presented in Table 1. To eliminate variances stemming from uneven environmental pollution in various provinces, donors were selected from only one region, i.e. Warsaw.

In comparing the concentrations of organochlorine compounds in human breast milk at the beginning of lactation with the samples collected starting from the third week after delivery worthy of notice is the fact of higher concentrations of PCBs and β-HCB in mature milk. These variances were statistically significant ($p \leq 0.05$). The variance identified in the levels of other compounds examined in transitory and mature milk was not statistically significant.

The monitoring examination of human breast milk conducted in selected regions of the country covered 365 milk samples. The findings are shown in Table 2.

Table 1. Mean concentrations of organochlorine compounds in 4th day milk and mature milk (mg/l)

	HCB	α-HCH	β-HCH	γ-HCH	DDT	DDD	DDE	PCBs
4th day milk (N = 27)								
X	0,0013	0,0002	0,0014	0,0002	0,0050	0,0009	0,0211	0,0076
SD	0,0008	0,0002	0,0008	0,0001	0,0024	0,0003	0,0140	0,0041
Min	0,0005	0,0002	0,0004	0,0002	0,0019	0,0008	0,0066	0,0017
Max	0,0040	0,0013	0,0036	0,0004	0,0094	0,0025	0,0528	0,0165
Mature milk (N = 70)								
X	0,0016	0,0002	0,0023	0,0002	0,0046	0,0011	0,0201	0,0220
SD	0,0008	0,0001	0,0011	0,0001	0,0021	0,0005	0,0161	0,0444
Min	0,0002	0,0002	0,0004	0,0002	0,0008	0,0010	0,0050	0,0010
Max	0,0039	0,0009	0,0057	0,0008	0,0122	0,0051	0,1016	0,3199

X - mean concentration
SD - standard deviation

Table 2. Mean concentrations of organochlorine compounds in human breast milk collected from donors living in more and less industrialized areas of Poland (mg/l).

	HCB	α-HCH	β-HCH	γ-HCH	DDT	DDD	DDE	PCBs
More industrialized areas (N = 158)								
X	0.0016	0.0004	0.0019	0.0005	0.0055	0.0012	0.0254	0.0220
SD	0.0012	0.0010	0.0015	0.0014	0.0109	0.0033	0.0222	0.0447
Min	0.0002	0.0002	0.0004	0.0002	0.0008	0.0005	0.0009	0.0010
Max	0.0110	0.0108	0.0101	0.0156	0.1355	0.0381	0.1388	0.3200
Less industrialized areas (N = 199)								
X	0.0022	0.0007	0.0041	0.0004	0.0028	0.0006	0.0275	0.0131
SD	0.0028	0.0018	0.0080	0.0011	0.0050	0.0004	0.0251	0.0145
Min	0.0002	0.0002	0.0004	0.0002	0.0008	0.0005	0.0017	0.0010
Max	0.0152	0.0155	0.0835	0.0083	0.0409	0.0048	0.1850	0.0741

X - mean concentration
SD - standard deviation

Mean concentrations of the compounds examined in human breast milk as calculated on the basis of biological monitoring data did not differ from those found in human milk in other European countries (Bordet *et al.* 1993, Koopman-Esseboom *et al.* 1994, Vaz 1995). However, the concentrations were lower than those found in the milk of women from countries where such compounds are still being used, e.g., in India (Tanabe *et al.* 1990). At present, in Poland and other European countries alike, a trend has been reported toward decreasing concentrations of organochlorine pesticides in the biological material since the imposition of a ban on their usage (Greve and van Zoonen 1990, Noren and Lunden 1991, Vaz 1995).

Higher concentrations of industrial pollutants (PCBs and HCB) were reported for the milk of women from more industrialized areas of Poland, although this difference was not statistically significant for PCBs ($p \leq 0.05$).

Differences between mean concentrations of compounds in the milk of women living in less industrialized areas and those found in women living in more industrialized areas were statistically significant also for β-HCH and DDT ($p \leq 0.05$).

However, mean β-HCH concentrations were found to be higher by a statistically significant margin ($p \leq 0.05$) for less industrialized areas than for more industrialized ones.

Among eight chlorinated hydrocarbons from all milk samples examined, the concentrations of p,p'-DDE and PCBs were always the highest. Relatively high PCBs concentration in human breast milk is no surprise as PCBs were still widely used in many industries in not that distant past. Statistically significant differences were found between the mean concentrations of β–HCH, HCB, p,p'-DDT and PCBs examined in the milk samples of women from more and less industrialized areas ($p \leq 0.05$).

The biological monitoring covering various regions of the country indicated a pattern in that the concentrations of p,p'-DDT (a pesticide recalled from usage in Poland 20 years ago) were consistently lower than those of its metabolize p,p'-DDE. The DDT/DDE ratio in the biological material falls with the passage of time since the DDT introduction to the environment.

The study findings indicate that the concentrations of DDT and its metabolizes in human breast milk may be subject to regional fluctuations.

From the analysis of the findings it follows that among all HCH isomers, the β isomer showed highest concentrations although its fraction in commercial

Table 3. Mean concentrations of organochlorine compounds in rural and urban women's milk (mg/l).

	HCB	α-HCH	β-HCH	γ-HCH	DDT	DDD	DDE	PCBs
Rural women's milk (N = 143)								
X	0.0020	0.0005	0.0041	0.0004	0.0025	0.0005	0.0241	0.0128
SD	0.0026	0.0015	0.0088	0.0012	0.0061	0.0004	0.0210	0.0131
Min	0.0002	0.0002	0.0004	0.0002	0.0008	0.0005	0.0020	0.0010
Max	0.0152	0.0092	0.0835	0.0083	0.0409	0.0025	0.1117	0.0606
Urban women's milk (N = 110)								
X	0.0019	0.0008	0.0030	0.0005	0.0039	0.0008	0.0258	0.0131
SD	0.0023	0.0023	0.0041	0.0018	0.0133	0.0040	0.0213	0.0154
Min	0.0002	0.0002	0.0004	0.0002	0.0008	0.0005	0.0009	0.0010
Max	0.0143	0.0155	0.0260	0.0156	0.1355	0.0381	0.1033	0.0741

X - mean concentration
SD - standard deviation

lindane-containing pesticide products usually does not exceed 2 to 3%. The reason is that the excretion of the β isomer from the human body takes five times longer than it does for other HCH isomers, and the β-HCH ability to accumulate in tissue fat is from 10 to 30 times stronger than it is in other isomers (Jensen 1983). The amount of β isomer is usually reported to be the highest in human and animal tissue from different regions of the world (Greve and van Zoonen 1990, Sitarska *et al.* 1995). In general, the α– and γ-HCH levels approximated the detection limit of the method.

Mean hexachlorobenzene concentrations in human breast milk were reported to be relatively low despite the fact that it ranks among the most stable organochlorine compounds. Mean HCB concentrations in human breast milk amounted to approximately 0.002 mg/1 of milk (Table 2). Thus, they were comparable to those reported in Scandinavian countries (Jensen and Slorach 1991) and approximately 100 times lower than, for example, those found in the milk of women living in Spain (Conde *et al.* 1993).

The PCBs mean concentrations in human breast milk (Table 2) were lower than those found in other countries that amounted to the mean 1 mg PCBs/kg of fat (Environmental Health 1988).

Some countries, e.g., Japan, have witnessed a marked decrease in the PCBs

concentrations in human breast milk that is attributable to rigid limitations on the compounds' usage. In the Netherlands no perceptible change in the concentrations of these compounds in human tissue has been found for the last dozen or so years (Greve and van Zoonen 1990), while in Poland the concentrations of these compounds are still reported to be rising (Niewiadowska 1988).

143 rural donors and 110 urban donors were selected from a group of 253. The women did not change locations of their permanent residence for, at least, the last 10 years. The findings are presented in Table 3.

No significant differences were found between the compounds examined in the milk of rural and urban women ($p \leq 0.05$). Thus, the place of residence was concluded to have no effect on the concentrations of the substances in the human breast milk studied. This may be attributed to the fact that the diet being the basic source of these compounds for man does not differ much in rural and urban people.

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