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**THE STUDY OF HUMAN EXPOSURE
TO PCBs AND OCPs IN THE REGION III – EUROPE**

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INTRODUCTION

Over almost 40 years of continuous determination of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in environmental samples, the analytical procedures have been changed according to the world trends in the analysis of trace substances.

Unfortunately, comparisons between the present analytical results and older data can be questionable because of differences in the analytical procedures such as: packed column versus capillary column separation, lower versus higher precision and accuracy, total PCBs versus congener specific analysis (**Deconing and Karmaus 2000**). The authors of various reports and papers have used different formats for the presentation of their material and often such important data as the number of samples, date of sampling and limit of detection, have not been reported (**Schutz et al. 1998**).

Consequently, some of the data presented in this study should be treated as indicating general levels and trends in environmental contamination and human exposure to PCBs and certain OCPs. When possible trends and other evaluations given in this report will include qualifying information to allow a better insight into the validity and comparability of the data.

SOURCES OF OCPs AND PCBs IN POLAND

Persistent organic pollutants (POPs), such as OCPs and PCBs are typically hydrophobic and lipophilic. In aquatic systems and soil they partition strongly to solids, notably organic matter, avoiding the aqueous phase. They also tend to incorporate into lipids in the organism, rather than entering the aqueous milieu of cells, and become stored in fatty tissue.

On the one hand, a combination of stability and propensity to form gas under appropriate environmental conditions means that POPs are subject to long-range atmospheric transportation, and a combination of their resistance to metabolism and lipophilicity means that they will accumulate in food chains (**Jones and de Voogt, 1999**).

On a local scale, among others in Poland, the risk of exposure to POPs significantly depends on the amounts and facility of their distribution from local sources, often correlated with the size of production. As follows from the data in Table 1, from among OCPs included

in the Dirty Dozen UNEP list, DDT was produced for the longest time (~ 33 years) and in the greatest amounts (~ 48,125 kT).

Contrary to Sweden, where the use of DDT was restricted in 1970, and this substance was totally banned in 1975 (**Norén and Meironyté, 2000**), in Poland the production and use of DDT were restricted only in 1980 (**Falandysz, 1996**).

Table 1. Production and use of certain OCPs and PCB formulas in Poland.

Formulas	Manufacture time	Quantity of production/usage [kT]
DDT	1947 - 80	48,152
Aldrin	1959 - 63	0,015
Dieldrin	1958 - 71	0,082
γ - HCH (lindane)	1956 - 92	7,510
HCB (hexachlorobenzene)	1962 - 72	0,188
Chlorofen (PCBs)	1966 - 70	1,000 ?
Tarnol (PCBs)	1974 - 77	0,679 ?

Data from references **Falandysz 1996; Falandysz and Szymczyk 2001; Kania-Korwel and Sulkowski, 2002**

Taking into account, that the time needed for p,p'-DDT and p,p'-DDE concentration decrease to half of the initial value ($t_{\text{dec.1/2}}$), (**Norén and Meironyté, 2000**), is 4 and 6 years, respectively, it should be expected that at present, the environmental levels of these xenobiotics in Poland can be relatively high.

Total quantity of HCB imported to Poland in 1962-72 was about 0,194 kT and the amount of the imported fungicide ranged from 0,0045 kT in 1968 to 0,008 kT in 1972 (**Falandysz et al. 2001a**). Fortunately, in the area of Poland there are no point sources able to produce large amounts of environmental pollution with HCB. Additionally HCB has favorable physico-chemical characteristics enabling its relatively easy volatilization from land surfaces and is prone for atmospheric transportation. Therefore, it should be expected that the levels of HCB residue in food chain would be constant showing no time-trend.

Although the production of PCBs in Poland did not bring a major contribution to the world market (Fig. 1), the amount of total Polish PCB formulas – Chlorofen (Ząbkowice Śląskie) and Tarnol (Tarnów) was estimated to about 1.679 kT (Table 1).

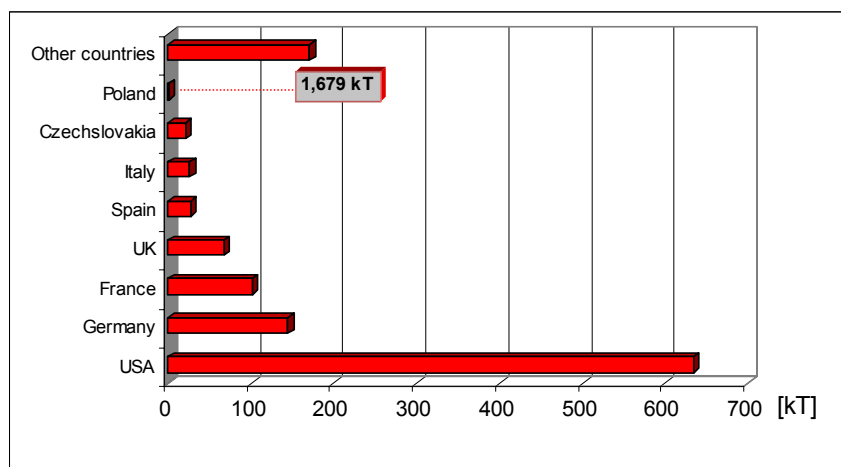


Figure 1. Production of PCB formulas [kT] in different countries in 1929-1985.

Data from references **(Holoubek, 2000; Kania-Korwel and Sulkowski, 2002)**

It results from Fig.1, the total amount of PCB mixtures produced in Poland was approximately 13 times and 86 times lower than those produced in Czechoslovakia and Germany, respectively.

Not identified quantities of PCB formulas from Soviet Union (Sovol) and Czechoslovakia (Delor) were imported to Poland before 1971 (**Falandysz and Szymczyk, 2001**). The presence of commercial PCB mixtures made in Germany (Clophen), France (Pyralen), and Italy (Phenaclor) in Polish installations cannot be excluded. It is also difficult to estimate the number of capacitors imported from different European countries (East Germany, Romania, Hungary, Sweden etc.)

The results of a detailed stock-taking of technical installation witch could be using products containing PCBs, made in southern western Poland, in 1995-97, proved, that the high voltage transformers, will few exception (~ 1.0%) are filled with Poland made oils which do not contain PCBs (**Rutkowski et al. 1997**).

Taking into consideration the data mentioned above and the introduction of national regulations regarding safe use of electrical equipment containing PCB (**Falandysz and Szymczyk 2001**), it can be supposed that exposure of Polish people to PCBs could be a minor problem that in other industrial countries of Europe.

PCBs AND OCPs OCCURANCE IN POLISH ENVIRONMENT

Ambient air and surface water (Vistula and Oder rivers) in Poland have been showed low concentration of PCBs which not exceed 0.12 ng/ m³ for air (**Falandysz et al. 1998**) and 14ng/l for water (**Lulek 2001; Galer et al. 2000**). In the precipitation and rainwater samples collected over period ten months in 1998 in Gdańsk region, among the OCPs examined (α HCH, γ HCH, aldrin, heptachlor epoxide, o,p'-DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD and p,p'-DDT), γ HCH, aldrin and heptachlor epoxide were detected most often. The highest concentration was recorded for γ HCH (5,1 ng/l). The presence of OCPs in deposition samples is most probably related to their persistence in the environment and/or to inflow with air masses from regions where they are still used. Similar concentrations of OCPs in rain were observed in Sierra Nevada Mountains (USA) in 1996 (γ HCH < 0,018 –1,87 ng/l) (**Gryniewicz et al, 2001**).

The contamination of soil and sediments with OCPs and PCBs may be related to point sources e.g. industrial discharges and waste plant effluents, but more frequently, as in the case of pesticides pollution, it is attributed to diffuse sources: precipitation, agriculture runoff, particle transport (**Falandysz et al. 2001b**). Due to their similar physicochemical properties, the fate of OCPs and PCBs is expected to be largely similar in the sediment and soil, but the characteristic of the medium, including its biological activity, has to be considered.

The mean PCBs concentration in agriculture and forest soils in Poland, collected in 1990-94 were relatively low (0 – 28 ng/ g dry weight) (**Lulek, 2001**). They are comparable with the results obtained for agriculture soil in Iassy rural sites in Romania (4.5 $\pm$ 2.0 ng/g dry weight) (**Covaci et al. 2001**).

The PCB concentration levels in the soil samples collected near their potential sources (transformer stations, spill sites) and from the area of the former Soviet army bases in the north-western and south-western regions of Poland were by one to two order of magnitude higher than in the agriculture soil (**Lulek, 2001**).

The highest level of OCPs and PCBs was detected in the soil in Silesia region (**Falandysz et al. 2001b**). Data presented in table 2 show that Silesia region soils are more polluted by organochlorines when compared to some other parts of Poland. This situation reflects the differences in population density and levels of industrialization.

Table 2. OCPs and PCBs levels in soils from southern industrial region of Poland

Sample	Katowice		Kraków	
Year of collection	1994		1994	
Sample number	9		15	
Analyte	Concentration[ng/g dry weight]		Concentration[ng/g dry weight]	
	Mean	Range	Mean	Range
HCB	6.4 ± 9.6	0.46 – 30	1.7 ± 2.6	0.19 - 9.9
Σ HCH ¹⁾	5.9 ± 3.3	1.1 – 11	11 ± 29	0.36 - 110
Σ DDT ²⁾	110 ± 89	23 – 260	260 ± 620	4.3 - 2400
Total PCB ³⁾	380 ± 300	67 – 870	53 ± 34	4.6 – 110

Σ HCH¹⁾= α HCH+ β HCH+ γ HCH; Σ DDT²⁾=p,p'DDT+p,p'DDE+p,p'DDD;
Total PCB³⁾=sum of resolved peaks on HRGC chromatogram (quantification standard - Kenachlor formulas, with known PCBs composition and content)

Data from reference (**Falandysz et al. 2001**)

The detected residual concentrations do indicate on relatively high use of PCBs. The p,p'-DDT/p,p'-DDE ratio equal 1: 62 is much greater than this observed by the same author in sediments (0.8 : 4.3) from the same geographic region of Poland. Proportion of HCHs were in order γ HCH> β HCH> α HCH. The α HCH/ γ HCH ratio indicates use of lindane at a much higher rates than of the technical BHC (benzene hexachloride) in Poland.

PCBs have been determined in recent [0-1(2), 1(2)-5 and 5-10 cm deep layers] sediments from different sites of southern Baltic Sea, including the Szczecin Lagoon, collected from May 1996 to October 1999 (**Konat and Kowalewska, 2001**). The sum of seven PCB congeners (28, 52, 101, 118, 138, 153 and 180) ranged between 1 to 149 ng/g dry weight, on mean this was rather low (< 40 ng/g dry weight). The highest content of PCBs was observed not in the vicinity of the most probable sources of pollution, but in areas of intensive sedimentation. There are the zones where mixing of river and marine waters occurred. Correlation of particular PCBs in all samples studies with sum of PCBs was linear and high ($r \geq 0.75$ -0.97), which implies a common source for all of them. However, an occurrence of PCB 28 is evidently different from that of other congeners. Most reasonably seems, that its better solubility in water and/ or decomposition of higher chlorinated biphenyls by e.i. microbiological attack are responsible for the observed PCB 28 contribution.

HUMAN EXPOSURE TO OCPs AND PCBs IN POLAND

Food chain

Taking into the consideration the PCB levels, recently detected in Polish environment (soil, air, water, sediments) (**Lulek, 2001, Konat and Kowalewska, 2001**) and the fact that water presents 3.4% and air 0.04% (**Hansen, 2001**) of the total daily intake of 23 PCBs congeners, it can be supposed that food chain could be a primary pathway of human exposure to PCBs in Poland.

Since 1995, a national monitoring program has been launched to assess the quality of plants, agriculture and foods products in Poland (**Report, 2001**).

Among others the contents of OCPs (HCB, p,p'- DDE, p,p'-DDD, p,p'-DDT and α,β,γ -HCH) as well as 7 PCB congeners (28, 52, 101, 118, 138, 153, 180) in raw and processed products of animal and plant origin have been determined. Fig.3 presents the percent contribution of cow milk, bovine, pork, game and carp adipose tissue samples in which the compounds of interest were detected in the years 1998-2000.

Low levels of OCPs were detected in 96% of the analysed samples. From among the compounds studied p,p'- DDE was found in the greatest percent of samples (96%), followed by p,p'-DDT in 35.6% of the samples and p,p'- DDD in 21.9% of the samples. The presence of p,p'-DDT was most often detected in pork and game adipose tissue, while p,p'-DDE mainly in carp adipose tissue.

In the years 1998-2000, the highest concentration of Σ DDT (table 3) was found in the adipose tissue of game, lower in carp and the lowest in pork and bovine adipose and milk.

From among the samples of animal raw products in 1.1% (26/ 2382) the total concentration of Σ DDT was higher than admissible by the Polish standards specified in the Maximal Acceptable Concentration of Σ DDT in meat and milk –1 μ /g lipids. In samples of cow milk and bovine adipose the concentrations of none of the pesticides studied exceeded the admissible level.

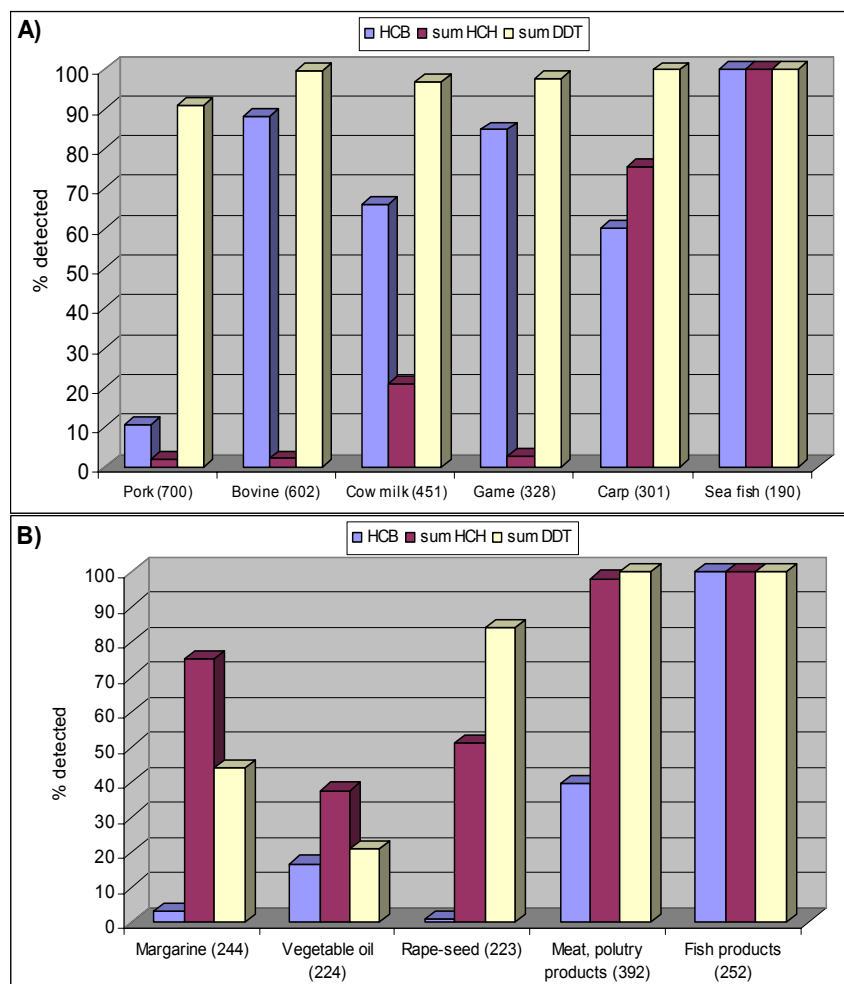


Figure 2. Contribution [%] of samples with detected residues of selected chlorinated pesticides in A) animal and fish tissues B) different processed food products. Data from reference (Rapport, 2001).

Table 3. Mean concentrations of selected OCPs in Polish raw and processed food products.

Matrix	Year of collection	No of samples	Analyte	Concentration [ng/g lipids]		Range [ng/g lipids]
				Mean	Median	
Swine (adipose tissue)	1998-2000	700	HCB	< 1,0	0	0 – 30
			Σ HCH ¹⁾	< 1,0	0	0 – 200
			Σ DDT ²⁾	79,0	24	0 – 7125
Bovine (adipose tissue)	1998-2000	602	HCB	4,0	4	0 – 18
			Σ HCH	< 1,0	0	0 – 15
			Σ DDT	62,0	44	0 – 786
Cow milk	1998-2000	451	HCB	3,0	3	0 – 12
			Σ HCH	1,0	0	0 – 62
			Σ DDT	39	32	0 – 201
Wild boar and Roe deer (adipose tissue)	1998-2000	328	HCB	5,0	4	0 – 28
			Σ HCH	<1,0	0	0 – 49
			Σ DDT	262,0	83	0 – 3824
Carp (muscle)	1998-2000	301	HCB	4,0	3	0 – 60
			Σ HCH	12,0	9	0 – 82
			Σ DDT	194,0	118	0 – 4033
Meat products	2000	392	HCB	4,9	3,0	0 – 18,0
			Σ HCH	54,0*	-	-
			Σ DDT	54,8	47,0	0 – 501,0
Vegetable oils	2000	392	HCB	3,3	3,0	0 – 6,0
			Σ HCH	11,9*	-	-
			Σ DDT	23,0	22,0	0 – 30,0
Margarine	2000	244	HCB	3,0	3,0	0 – 3,4
			Σ HCH	24,1*	-	-
			Σ DDT	28,0	22,0	0 – 65,0
Rape-seeds	2000	233	HCB	3,0	3,0	0 – 4,0
			Σ HCH	75,5*	-	-
			Σ DDT	52,0	50,0	0 – 679,0
Fish: cod, herring, flounder, sprat	2000	174	HCB	17,0	15,0	1,1 – 84,0
			Σ HCH	33,0*	-	-
			Σ DDT	600,0	550,0	48,0 – 2210,0
Canned cod liver	2000	16	HCB	36,0	37,4	16,3 – 53,9
			Σ HCH	32,5*	-	-
			Σ DDT	1191,0	856,0	221,0 – 2830,0
Canned fish	2000	51	HCB	9,1	8,9	0,2 – 9,0
			Σ HCH	12,2*	-	-
			Σ DDT	185,0	154,0	10,7 – 818,0

Σ HCH¹⁾= α HCH+ β HCH+ γ HCH; Σ DDT²⁾=p,p'DDT+p,p'DDE+p,p'DDD;

* - sum of mean concentration of α -HCH, β -HCH and γ - HCH

Data from reference (**Report, 2001**)

In all samples mentioned above HCB was determined in low concentrations (0 – 60 ng/g lipids), which corresponds with the low levels of this fungicide in Polish soils, reported by Falandysz et al. (**Falandysz et al. 2001b**), HCH isomers were detected in trace amounts (0 – 200 ng/g lipids).

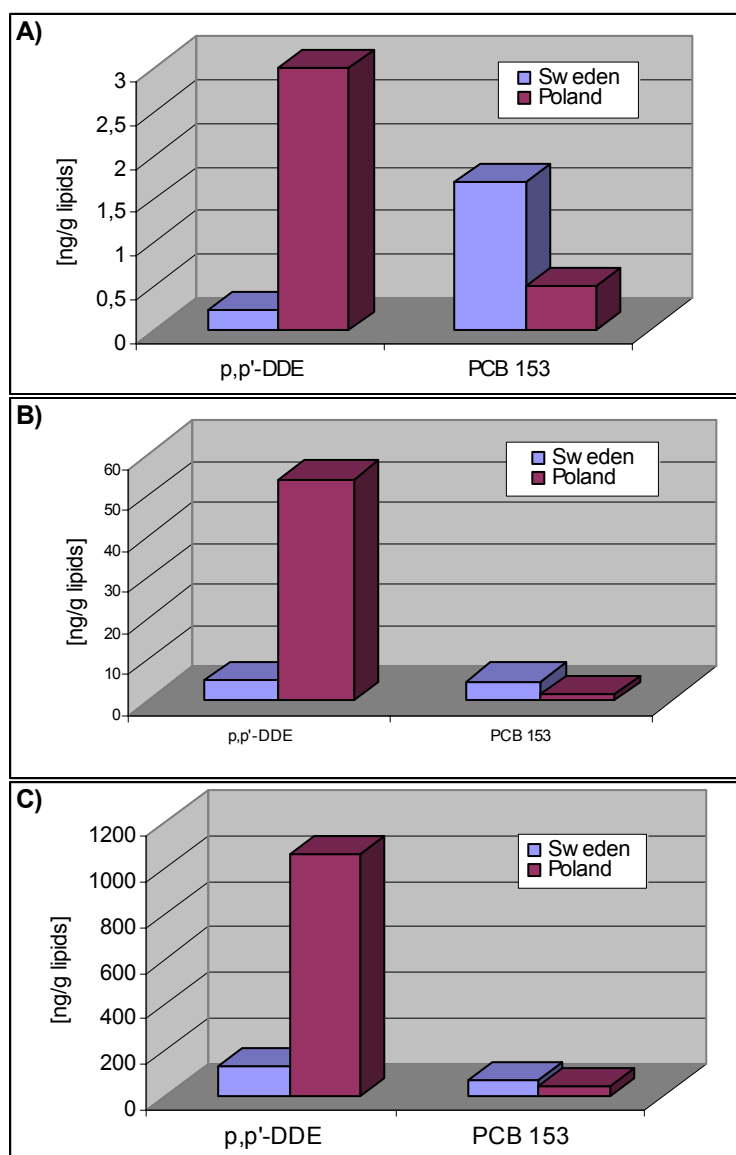


Figure 3. Mean concentration of A) p,p'-DDE (1997, n=79?) and PCB 153 (1997, n=79?) in Swedish and p,p'-DDE (1999, n=200) and PCB 153 (2000, n=150) in Polish adipose pork tissue, B) p,p'-DDE (1997, n=71?) and PCB 153 (1997, n=71?) in Swedish and p,p'-DDE (1999, n=152) and PCB 153 (2000, n=150) in Polish adipose bovine tissue, C) p,p'-DDE and PCB 153 in human milk from Sweden (1997, n=40) and Wielkopolska region in Poland (2001, n= 28). Data from references **(Report, 2001; Glynn et al. 2000)**

As follows from the data in Figs 3 a and b, significant differences in the concentration of p,p'-DDE were found in the Polish and Swedish samples of bovine and pork adipose the **(Report, 2001, Glynn et al. 2000)**. The levels of p,p'-DDE in Polish samples are about 10 times higher in pork and bovine adipose tissues. .

Within the monitoring study in 2000, also samples of rape seeds, margarine and processed meat and poultry products. As follows from Fig. 2 only in poultry and meat products and rape seeds DDT was found in over 80% of samples. The presence of DDT was detected in less than 50% of the margarine samples and less than 20% of plant oil samples.

From among the pesticides under detection the lowest was the concentration of HCB (0-18 ng/g lipids) in all samples analysed (Table 3). It was also established that lower levels of p,p'-

DDE and p,p'-DDT occur in the raw and processed products of plant origin (rape seeds, margarine, oils) than in those of animal origin. Only in 1 of 1261 samples analysed the concentration of β -HCH exceeded 0,1 μ g/ g lipids - the Maximal Acceptable Concentration, and in none of the samples the concentration of Σ DDT and Σ HCH was higher than admissible in Polish and EU standards.

A separate group of matrices analysed within the monitoring were the raw and processed products of Baltic fish. The contents of OCP was determined in 190 samples of raw fish products and 252 samples of processed fish products (table 3).

The residues of OCPs were detected in practically all samples analysed. Only in 2.1% (4/190) samples of raw fish products and 1.2 % (3/252) samples of processed fish, the Σ DDT exceeded 2 μ g/g lipids, that is the maximum admissible value according to the French standards of 1998 (**Report, 2001**).

The levels of OCP concentrations determined in raw and processed fish in 2000 are much lower than those reported by other authors for the Baltic fish determined in the beginning of the 90s –examples -. HCB- 41 ng/g lipids; Σ HCH – 75 ng/g lipids; Σ DDT – 1257 ng/lipids in herring (1) from Gulf Gdańsk collected in 1992 (**Strandberg et al. 1998**); HCB- 86 ng/g lipids; Σ HCH – 75,4 ng/g lipids; Σ DDT – 1098 in herring from Baltic sea (8) collected in 1992/3 (**Atuma et al. 1996**) This decrease confirms the tendency of a gradual decrease of the OCPs level in bottom sediments and organisms coming from the Baltic sea reported for the last decades (**Monitor 16, 1998, Konat and Kowalewska 2000**).

A separate study within this monitoring scheme involved determination of 7 indicator PCB congeners (28, 52, 101, 118, 138, 153 and 180) in raw and processed products of plant and animal origin. From among 651 of the samples analysed (cow milk, bovine, pork, game and carp adipose tissues) in 71 % the presence of PCBs at low levels was detected (median of the Σ 7 PCB – 0 – 4 ng/ g lipids). The least frequently PCB was found in pork tissue (21% of the samples analysed).

As follows from the data shown in table 4, the mean concentrations of the total PCB varied between 1.2 ng/ g lipids in pork adipose tissue to 27.8 ng/g lipids in carp adipose tissue.

Table 4. Mean concentrations of specified PCB congeners [ng/g lipids] in the raw and processed food products in Poland in 2000.

Matrix	n	PCB 28	PCB 52	PCB 101	PCB 118	PCB 138	PCB 153	PCB 180	Σ PCB
Cow milk	101	0,200	0,10	n. d.*	0,100	0,700	0,90	0,30	2,30
Vegetable oil	224	28,50	46,70	30,1	1,00	8,30	13,80	2,50	130,90
Margarine	244	33,40	46,10	26,2	3,30	7,90	15,10	3,40	135,40
Rape-seed	223	35,10	55,10	21,10	5,10	3,20	14,60	4,60	138,80
Meat and poultry products	392	52,80	56,10	18,20	4,80	11,10	7,30	2,10	152,40
Carp	100	1,60	1,60	3,70	3,00	7,90	5,90	4,10	27,80
Bovine adipose tissue	150	0,10	<0,1	<0,1	0,30	1,40	1,10	0,70	3,60
Pork adipose tissue	200	n.d.	0,10	<0,1	<0,1	0,50	0,40	0,20	1,20
Σ PCB = PCB28+PCB52+PCB101+PCB118+PCB138+PCB153+PCB180 ; n.d.* - not detected									

Data from reference: **Report, 2001**

It is worth noting that the mean levels of PCB 153 determined in bovine and pork adipose tissue from Poland (Fig.3a and 3b) were about 3 times lower than those detected in 1997 in the animal tissues from Sweden (**Report, 2000, Glynn et al., 2000**). It confirms the data concerning relatively low levels of PCB in Polish environment (**Lulek, 1998; Lulek, 2001; Falandysz and Szymczyk, 2001**).

Another group of products tested for the presence of PCB were rape seeds, plant oils, margarine and processed poultry products. The limit of the determination (LOD) of congener 52 was 5 ng/ g lipids and of the other congeners – 1 ng/g lipids.

In each sample the presence of at least two PCB congeners was detected. The concentration of PCB 153 did not exceed the value 100 ng/ g lipids (table 4), assumed as indicating the presence of dioxins (**Report, 2001**). In 14% of the samples of meat products (n=392) and in 6.6 % samples of rape seeds (n=223) the concentration of Σ PCB exceeded the second value indicating the presence of dioxins, it means - the Σ PCB > 250 ng/g lipids.

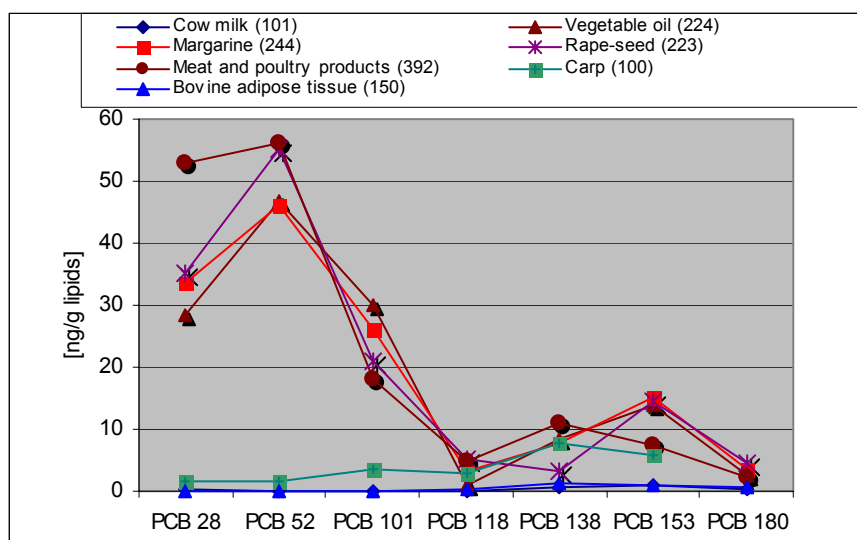


Figure 4. The concentration profiles of seven indicator PCB congeners in the raw and processed food products. Data from reference: (**Report, 2001**)

As shown in Fig. 4 the concentration profiles of seven PCBs in the raw and processed food products analysed, Baltic sea fish excluded, differ across the matrix. The highest concentration of PCB 52 and PCB 28 has been determined in rape seeds, vegetable oil and margarine as well as in the samples of meat and poultry products. In the samples of cow milk and carp tissue the highest concentrations of PCB 153 and 138 have been found.

Similarly as for OCPs, all analysed samples of raw fish (n=190) and fish products (n=252) have been contained PCBs. Only in one sample of raw fish (n=190) no presence of congener 101 was detected, and in one sample of fish products no PCB 101, 153 and 180 were detected. The mean content of the Σ PCB (28, 52, 101, 118, 138, 151, 153 and 180) in all raw fish products, including the cod livers was 460 ng/ g lipids, which corresponds to 31 ng/ g tissue.

The content of PCB in tissues of the main species of Baltic sea fish (cod, herring, sprat, flounder) was rather low, the total PCB ranged between 270-420 ng/g lipids and was significantly lower than reported in samples collected in 1992 (herring from Gulf Gdańsk – 1333 ng/g lipids) (**Strandberg et al. 1998**). The highest mean concentration of the total PCB was determined in the samples of cod species (660 ng/ g lipids) and it was about 6 times higher than the mean concentration of the total PCB in the other tinned fish products. It is worth noting that in none of the fish product samples the concentrations admissible according to the Dutch standards, i.e. 2 µg of total PCBs/g lipids, were exceeded.

The concentrations of PCBs determined in 2000 both in the raw and processed fish products are much lower than those reported by other authors as determined in the end of the 80s and beginning of the 90s (**Lulek 2001**).

The above discussed data and the results of the monitoring study of raw and processed food products performed in 1995-2000 (**Report, 1996; Report, 1997, Report, 2000**) confirm a tendency of decreasing levels of OCPs and PCBs in Polish food products observed for the last 30 years.

On the basis of the results of the national monitoring, the daily intake of selected POPs has been estimated (**Report, 2001**). The dietary daily intake of Σ DDT, γ -HCH and total PCBs has calculated from the average concentration of specifies pesticides and total PCBs detected in row and processed food products in 2000 as well as from the average daily intake of these products reported by GUS (Central Statistical Office), in the same year. The estimated dietary intakes (EDI) were compared to the values of acceptable daily intakes (ADI) from Codex Alimentarius (20 μg total DDT/kg body weight/day; 1 μg γ -HCH /kg body weight/day) (**Report, 2001**) and from USA/FDA recommendation (**Ludwicki et al. 1996**) (1 μg total PCBs /kg body weight/day). It is worth noting, that the estimated daily intakes (EDI) in pork, beef, game, cow milk and milk products consumption did not exceed 0.5 % of ADI value for the OCPs studied and 1.0 % of ADI value for total PCBs. Than, the EDI of total DDT and PCBs in Baltic sea fish and fish products consumption did not exceed 2.21% and 0.07 % of ADI_{DDT} and ADI_{PCBs}, respectively.

Human breast milk and adipose tissue

Human breast milk, due to the relatively high fat content, is on the one hand, a major source of bioaccumulated contaminants for nursing infants and, on the other hand, a suitable matrix for studies of long term exposure to PCBs (**Schutz et al. 1998; Brouwer et al. 1998**).

Until 2001, the only available data concerning the levels of specified OCPs (HCB, α HCH, β HCH, γ HCH and p,p'-DDT, p,p'-DDE and p,p'-DDD) in Polish human milk samples collected in various region of Poland were presented by Ludwicki et al. (**Ludwicki et al. 1996**) and Czaja et al. (**Czaja et al. 1997; Czaja et al. 2001**). In Fig. 5, these data are compared to those reported by Szyrwińska and Lulek (**Szyrwińska and Lulek, 2002**) for human milk samples (n=28), collected in 2000-01 in Poznań and Wielkopolska region.

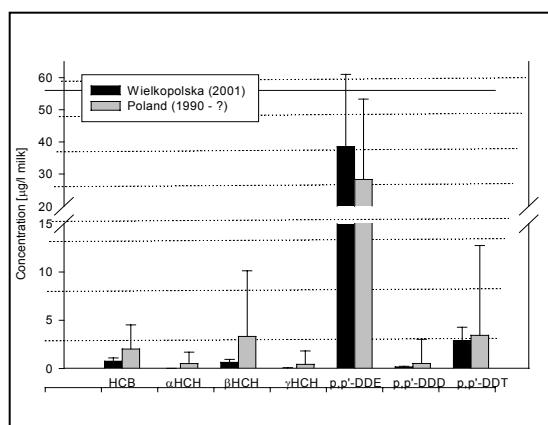


Figure 5. Mean concentration of selected chlorinated pesticides and their metabolites in human breast milk in Poland. Data from references (**Czaja et al. 2001; Szyrwińska and Lulek, 2002**)

There is no significant difference between the levels of OCPs detected in Wielkopolska region and in the area of whole Poland. Lower concentrations of all OCPs, except p,p'-DDE, determined in the samples from Wielkopolska region can be explained by the fact, that the samples from the other regions of Poland were probably collected some time earlier. The

concentrations of OCPs detected in Polish samples (except for p,p'-DDE) are slightly higher than those detected in Swedish human milk collected in 1997 (Figure 6) and lower than those determined in human milk from Germany, collected in 1995-97 (**Schade, and Heinzow 1998**).

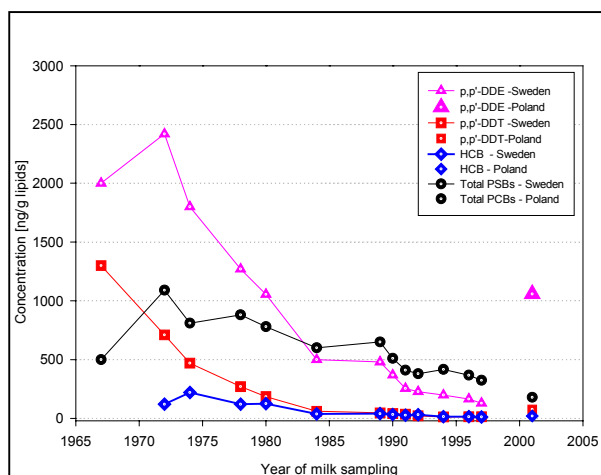


Figure 6. Residues of some OCPs and PCBs in Swedish (n= 40-401) and Polish human milk (n=28).Data from references (**Norén and Meironyté, 2000, Szyrwińska and Lulek 2002**)

As results from Figs 3c and 6, the concentration of p,p'- DDE in Polish milk samples is more than 8 times higher than in Swedish milk samples collected in 1997. It should be particularly striking in view of the relatively high levels of this compound determined in raw and processed Polish food products as illustrated by the data in Table 3 and Figs 3a and 3b.

The recommended cessation of use and the authority measures taken to reduce the environmental pollution have brought about a significant decrease in the levels of OCPs and PCBs in human milk over the last 30 years, as shown by the data in Fig. 6.

In Sweden, the use of several OCPs and PCBs was restricted or prohibited in 1970. Since that time a most consistent decline has been observed in the levels of p,p'-DDT and p,p'-DDE. The level of p,p'- DDE in 1997 (14ng/g lipids) was only 1% of that recorded in 1967 (1300 ng/g lipids). In 1997 the concentration of p,p'-DDE (129 ng/g lipids) was 5% of that recorded in 1972 (2420 ng/g lipids). The concentration of HCB in human milk varied in the 1970s (120-220 ng/g lipids) but a manifest decline in the mean level of HCB in Swedish human milk has been apparent since 1980. In 1997 the mean level of HCB in Swedish human milk did not exceed 12 ng/ g lipids.

The level of total PCBs in 1997 (324 ng/g lipids) was about 30% of that determined in 1972 (1090 ng/g lipids).

Similar declining trends in the POPs levels in human milk from mothers living in northern Germany over the period of the last 12 years have been presented by Schade and Heinzow (**Schade and Heinzow, 1998**). The median DDT (p,p'-DDE+p,p' DDT) level has decreased from 920 ng/g lipids to 230 ng/g lipids, the median HCB level from 510 ng/g lipids to 60 ng/g lipids and β HCH level from 190 ng/g lipids to 30 ng/g lipids. Relative to the median, the concentration level of DDT in 1996 and 1997 was approximately 81% lower than 10 years earlier. The median HCB level and the median β -HCH were reduced by 88% and 84% respectively. The median PCBs [total PCBs = (PCB138+PCB153+PCB180)*1,64] level decreased from 1280 ng/g lipids in 1986 to 470 ng/g in 1997, witch corresponds to a reduction in the level of these xenobiotics by 61% over ten years.

The PCB levels in human milk from different parts of world have been extensively studied both in terms of total PCBs (packed column gas chromatography) (**Frkovic et al. 1996; Czaja et al. 1997; Konishi et al. 2001; Dekoning and Karmaus 2000**) and, more recently, for individual congeners (capillary column gas chromatography) (**Craan et al. 1998; Schade and Heinzow 1998; Kivaranta et al. 2000; Lulek et al. 2002**).

Fig. 7 presents the mean concentration of total PCBs in human milk from different countries, between 1987 and 2001.

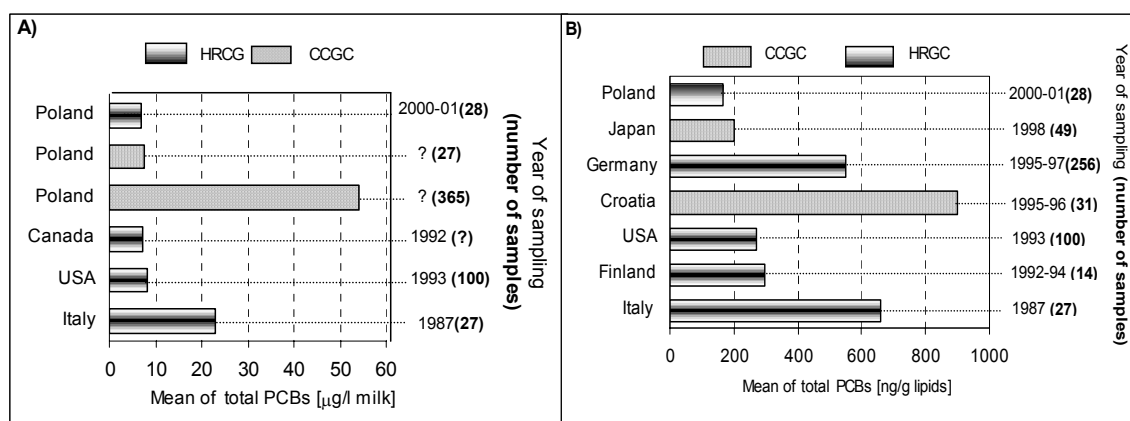


Figure 7. Mean concentration of total PCBs in human milk form different countries, detected by capillary chromatography (HRGC) and packed column chromatography (CCGC): a) in µg/g milk, b) in ng/g lipids. Data from references: (**Larsen et al. 1994; Schade and Heinzow, 1998; Czaja et al. 2001; Kostyniak et al, 1999; Konishi at al, 2001; Frkovic et al. 1996; Kivaranta et al. 2000; Szyrwińska and Lulek, 2002**)

The highest mean concentration of total PCBs was detected in human milk samples (31) from Croatia, collected in 1995-6 (898 ng/g lipids) (**Frkovic et al. 1996**) and in 365 milk samples from Poland (5440 ng/l) (**Czaja et al., 2001**). Unfortunately, the Polish authors did not specify the year of the samples collection and the manner of quantification of total PCBs content. The results presented in Fig. 7 of the levels of PCBs determined in human milk from different countries do not show significant differences according to the site of sample collection. The differences in the PCB levels are rather a consequence of different times of human milk sampling.

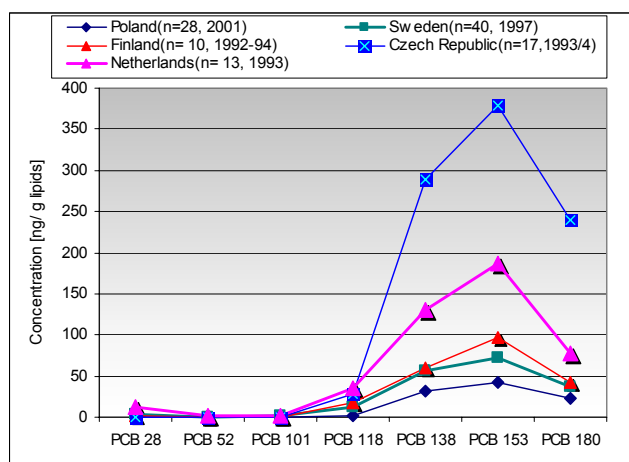


Figure 8. Profiles of specified PCB congeners in human milk from different countries in Europe. Data from references (**Schoula et al. 1996; Kiviranta et al. 1999; Norén and Meironyté 2000; Szyrwińska and Lulek 2002**)

As follows from Fig. 8, the concentration profiles of the seven indicator congeners (28, 52, 101, 118, 138, 153, 180) in Polish human milk from Wielkopolska region are very similar to those reported by other authors in the other countries of Europe. The presented data confirm the finding reported by Norén et al. (**Norén and Meironyté 2000**), that PCB 153 (2,2',4,4',5,5' – hexa-chlorobiphenyl) and PCB 138 (2,2',3,4,4',5 – hexa-chlorobiphenyl) are the most stable congeners in the environment.

The approximate daily intake of pesticides and PCB congeners examined ($\mu\text{g/kg}$ body weight/ day) by the breastfed infants from Wielkopolska region was calculated by multiplying the concentration of a particular substance in whole breast milk ($\mu\text{g/kg}$) (**Szyrwińska and Lulek 2002**), by a factor 0.12 according to Schutz et al. suggestions (**Schutz et al. 1998**). The obtained EDI values were compared to the reference intakes proposed by Codex Alimentarius and WHO as well as to the revised tolerable daily intakes TDI recently proposed by National Institute of Public Health and Environment, Netherlands (RIVM), (**Baars et al. 2001**).

Table 5. Estimated daily intakes of selected OCPs and PCBs by infants breast-feed in Wielkopolska region in Poland in 2000-01

Congener	EDI ($\mu\text{g/kg}$ body weight/day)	Pesticide	EDI ($\mu\text{g/kg}$ body weight/day)
PCB 28	0,004	HCB	0,090
PCB 52	0,005	α HCH	0,012
PCB 101	0,004	β HCH	0,076
PCB 118	0,033	γ HCH	0,002
PCB 138	0,157	$\Sigma\text{HCH}^{\text{c)}$	0,09
PCB 153	0,210	p,p'-DDE	4,610
PCB 180	0,114	p,p'-DDD	0,017
Σ PCB ^{a)}	0,527	p,p'-DDT	0,343
Total PCBs ^{b)}	0,818	Σ DDT ^{d)}	4,970

^{a)} Σ PCB = PCB28+PCB52+PCB101+PCB118+PCB138+PCB153+PCB180;
^{b)} Total PCBs=(PCB138+PCB153+PCB180)*1,7;
 Σ HCH^{c)}= α HCH+ β HCH+ γ HCH;
 Σ DDT^{d)}=p,p' DDT+p,p' DDE+p,p' DDD;

Data from reference (**Szyrwińska and Lulek, 2002**)

The daily intake of the total PCBs (0,818 $\mu\text{g/kg}$ body weight/day) have been estimated below the ADI value established by USA/FDA (1 μg total PCBs/kg body weight/day) (**Ludwicki et al. 1996**). It was found to be about four times lower than average daily intake (2,56 $\mu\text{g/kg}$ body weight/day) reported recently for Slovakia infants (**Holoubek et al. 2000a**) and five times lower than those reported by Czaja et al (2,8 $\mu\text{g/kg}$ body weight/day) (**Holoubek et al. 2000a**).

It is estimated that the daily intakes of PCBs of the breastfed babies is by 1 to 2 orders of magnitude higher when compared to adults (**van Leeuwen and Younes, 1998**). The EDI based on the sum of indicator congeners (table 5) was found to be 0,527 $\mu\text{g/kg}$ body weight/day. Thus, it is not surprising that TDI of sum of PCB indicators (28,52,101,118,138,153 and 180) is about 50 times higher than proposed in the RIVM report (0,01 $\mu\text{g/}$ kg body weight /day) (**Baars et al. 2001**).

Nevertheless, as results from figure 9, the intake of selected PCBs by the breastfeed infants is lower in Wielkopolska region then in other European countries. It is worth noting

that the PCB levels in breast milk from Wielkopolska region are significantly lower (166,3 ng/ g lipids in 2001) than those determined recently in breast milk from Czech samples (mean total PCBs- 959 ng/g lipids in 1996) (**Holoubek et al. 2000a**), German samples (median total PCBs – 470 ng/g lipids in 1997) (**Schade and Heinzow, 1998**) and somewhat lower than in Swedish samples of human milk (324 ng/g lipids in 1997) (**Norén and Maironyté, 2000**).

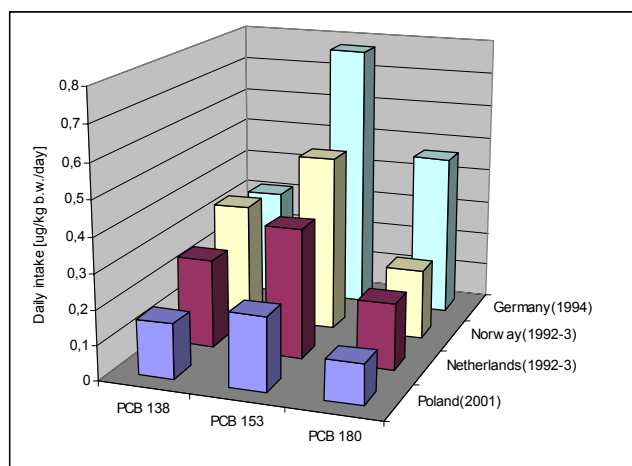


Figure 9. Daily intake of selected PCB congeners by infants from human milk in different countries of Europe. Data from references (**Schutz et al. 1998; Szyrwińska and Lulek 2002**)

Concerning Σ DDT the daily intakes of this xenobiotics (table 5) have been estimated below the ADI value established by WHO ($20\mu\text{g } \Sigma \text{ DDT / kg body weight/day}$) (**Ludwicki et al. 1996**). Taking into account the p,p'- DDE levels detected in Polish food products and the fact that many authors have proved a relationship between the mother's diet and organochlorine concentrations in breast milk, it is not surprising that the estimated dietary intake of DDT and its metabolites in human milk from Poland exceeds the recent TDI value ($0.5 \mu\text{g/kg body weight/day}$) (**Baars et al. 2001**).

The $\text{EDI}_{\gamma\text{HCH}}$ in Wielkopolska region is below not only the ADI value from the Codex Alimentarius ($1\mu\text{g/ kg body weight/day}$) (**Report, 2001**) but also below the very restricted TDI γHCH value ($0,04\mu\text{g/ kg body weight/day}$), proposed by Dutch authorities (**Baars et al. 2001**).

On the basis of the data presented and the report of the current WHO working group (**Brouwer et al. 1998**) there is insufficient scientific evidence which would imply a change of the current WHO recommendations supporting breast feeding.

In order to find out whether the modest PCB exposure experienced with declining global load involves a real health risk, it is necessary to examine the exposure more accurately and in more details.

Recently, much attention has been paid congener-specific analysis of human breast milk and human tissues, these matrices are particularly suitable for monitoring long term exposure to PCBs.

The most recent data on the fourteen specified congeners (IUPAC Nos: 28, 52, 74, 101, 105, 114+118, 128, 138, 149, 153, 156, 170, 180 and 187) determination in 7 samples of human adipose tissues coming from Wielkopolska region in Poland have been reported by Lulek et al. (**Szafran et al. 2002a**). The determined total PCBs levels ranged between 76,6

and 472 ng/g wet weight (mean – 276,07 ng/g wet weight) and were generally lower than those determined in Polish adipose tissues by other authors in the years between 1975 and 1990 (190 ng/g wet weight. – 1500 ng/g wet weight) (Falandysz et al. 1993; Ludwicki and Góralczyk 1994a). As shown in figure 9, the concentrations of congeners 156, 170 and 180 in human adipose tissues from Poland are lower than those from Italy but significantly higher than in those from Chile.

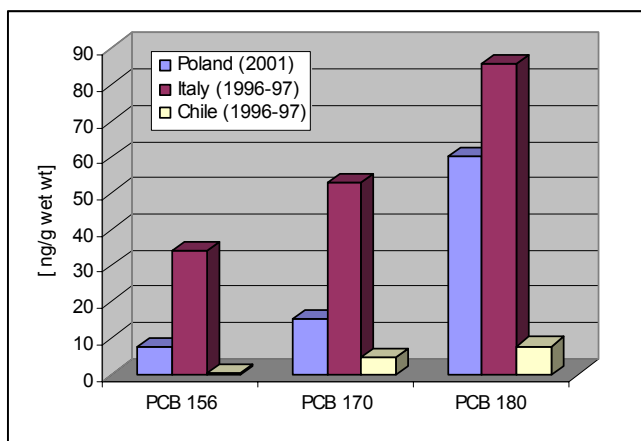


Figure 11. Concentration of specific PCB congeners in human adipose from different countries. Data from references (Szafran et al. 2002; Mariottini et al. 2000)

Similarly to Falandysz results (Falandysz et al. 1994) and the results reported for Italian and Chilean adipose tissues (Mariottini et al. 2000), PCB-153 accounted for more than 20% of the total PCBs residue in all samples examined. The recent results are in good agreement with those presented by Ludwicki et Góralczyk (Ludwicki and Góralczyk 1994a) showing a good correlation between the age of donors and the concentration of PCBs.

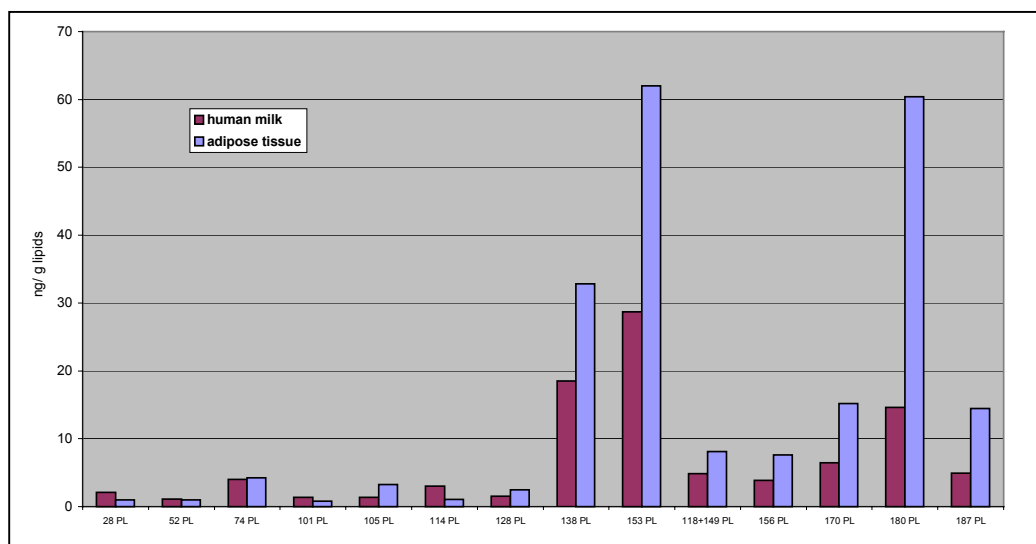


Figure 12. Comparison of the specified “state study” and “episodic” PCB congeners in human milk (n=27) and adipose tissue (n=7) of the people living in Wielkopolska region in Poland. Data from reference (Szafran et al, 2002; Szafran et al 2002a)

Fig.12 presents a comparison of the results of the congener-specific analysis of 27 Polish breast milk samples (Szafran et al, 2002) and 7 human adipose tissue samples (Szafran et al, 2002a), collected in 2000-01 in Poznań and Wielkopolska region. They revealed higher

concentration of all the “steady state” congeners examined (74, 105, 118+149, 138, 153, 156, 170, 180 and 187, except of 28) in adipose tissue. Overwhelming contribution of “episodic” congeners (CB 52 and 101) in breast milk when compared to the adipose tissue was shown.

These results confirmed the report of by Hansen (**Hansen, 2001**) (Welsh breast milk and adipose tissue).

These presented results from the pilot studies (Grant 754/PO5/99/16 of Polish State Committee of Scientific Research) involved a relatively small number of specimen (27 human milk and 7 adipose tissue donors) and it was questionable to evaluate any general critical parameters. Nevertheless its represent the first human data characterizing the background exposure of Wielkopolska region population to the specific PCB congeners.

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