

**ANNALS OF THE FACULTY OF ENGINEERING HUNEDOARA** 

2005, Tome III, Fascicole 1

# RESIDUAL LEVELS OF POLYCHLORINATED BIPHENYLS IN HUMAN ADIPOSE TISSUE AND LIVER IN VOYVODINA (SERBIA)

Jasna ADAMOV<sup>1</sup>, Jan SUDJI<sup>2</sup>, Mirjana VOJINOVIC-MILORADOV<sup>1</sup>, Zivka ERI<sup>3</sup>, Ferenc GAÁL<sup>1</sup>

> <sup>1</sup>UNIVERSITY OF NOVI SAD, FACULTY OF SCIENCES, SERBIA & MONTENEGRO, <sup>2</sup>INSTITUTE OF OCCUPATIONAL HEALTH, NOVI SAD, SERBIA & MONTENEGRO <sup>3</sup>INSTITUTE OF ONCOLOGY SREMSKA KAMENICA, NOVI SAD, SERBIA & MONTENEGRO

## ABSTRACT

In this paper the content of polychlorinated biphenyls (PCBs) was determined in the samples of human adipose tissue and liver, collected during obductions from random donors of both gender and different ages. Residues of PCBs were determined by GC/ECD. Concentration of total PCBs was in the range 21.9 to 205.8 ng/g of adipose tissue and 3.6 to 47.8 ng/g of liver. The most abundant were congeners PCB 138, PCB 153 and PCB 180, comprising >80% of the total content of PCBs in the adipose tissue and liver. There is no significant correlation between age and gender of the donors and the levels of PCBs in their tissue.

# 1. INTRODUCTION

Polychlorinated biphenyls (PCBs) are ubiquitous pollutants that bioaccumulate in the food chain. Because of the bioaccumulation and toxicity, PCBs are considered by the Stockholm Convention [1] to be members of the 'dirty dozen' chemicals (persistent organic pollutants – POPs) to be phased out worldwide. Despite restrictions on their manufacture and use in most industrialized countries since the 1970s, they are still of public concern. Exposure to POPs, either acute or chronic, can be associated with a wide range of adverse health effects, including illness and death. PCBs are known to cause chronic reproductive effects, gastric disorders, and skin lesions in laboratory animals. Laboratory investigations and environmental impact studies in the wild have implicated that POPs cause endocrine disruption, reproductive and immune dysfunction, neurobehavioural and developmental disorders and cancer. The U.S. Environmental Protection Agency (1996) [2] suspects PCBs and some OCPs to be probable human carcinogens. While there is evidence that PCBs concentrations in the environment have generally been declining in the last decade, data available on humans suggest that the declining was much slower to respond to the restriction in use or incident. Presumably, this is due to the high persistence of PCBs in the fatty tissues in which they are deposited. Data on human contamination with PCB and pesticides are focused on blood or milk [3], the only biological materials conveniently available from healthy subjects. Adipose tissue is another choice for the investigation of human body burden [4]. The detection of organochlorine contaminants in blood, breast milk, and adipose tissue samples from the general population indicates widespread exposure to these hazardous toxicants from environmental sources.

The aim of this study was to determine level of PCBs in human adipose tissue and liver of the population of Vojvodina. Being a highly developed industrial and agricultural region of Serbia, where PCBs have been used for decades, it can be expected a high residual level of these contaminants in humans. Moreover, numerous incidental spills of transformer oils during 1999 Balkan conflict in Serbia occurred in this region, when large quantities of PCBs leaked into the ground and watercourses. It is reasonable to expect that 5 years later these contaminants have entered the food chain, raising the levels of PCBs in the general population of Vojvodina.

### 2. MATERIALS AND METHODS

**Sampling**: Samples of human adipose tissue and liver were collected in 2004 from 15 test subjects during obductions at the Institute of Oncology in Sremska Kamenica, from randomly chosen donors who had been living in Novi Sad area for at least 10 years, allegedly not professionally exposed to PCBs. A group of 15 patients was studied: 11 males and 4 females, with ages 43-82 (average age: 60 9 for males and 70 8 for females). Tissue samples were kept frozen at -25 C in glass containers until analysis.

**Experimental:** Determination of PCBs in human adipose and liver samples was performed according to Vojinovic-Miloradov et al. [5]. Samples of human adipose tissue (approximately 2 g) were extracted with 50 ml of the mixture chloroform:methanol:water (2:2:1) for 24 hours on the mechanical shaker. After filtration, organic layer was evaporated, dissolved in 5 ml *n*-hexane and transferred into the separation funnel. Cleanup was performed with concentrated sulfuric acid until discoloration. Concentration of PCBs in the samples was determined by gas chromatography (GC/ECD).

**GC parameters**: *Instrument:* HP 6890; Initial temperature: 120 C; *Injector:* splitless; Initial temperature: 290 C; Pressure: 22.0 psi; Carrier gas: Helium; *Column:* Capillary DB-5MS 123-5562 HP-5 5% Phenyl Methyl Siloxane; 60.0 m x 320.00 m x 0.25 m; Carrier gas: helium, constant pressure (22.0 psi); Initial flow: 2.2 ml/min; *Detector:* ECD, working temperature: 310 C; carrier gas: nitrogen. *PCB standard*:

EPA standard mixture of 7 congeners (PCB 28, PCB 52, PCB 101, PCB 118, PCB 153, PCB 138, PCB 180).

### 3. RESULTS AND DISCUSSION

Previous studies by Adamov (2004) have shown that POPs are present in foodstuffs used in the average diet of the population of Vojvodina, and the assessed daily human dietary exposure to PCBs is 11 ng/kg day [6]. In the present study residues of PCBs were detected in all investigated samples of human adipose tissue and liver. However, their concentrations are not uniformly distributed and the total PCBs ranged from 21.9 to 205.8 ng/g of adipose tissue and 3.6 to 47.8 ng/g of liver. Individual results are given in Tables 1 and 2.

TABLE 1. RESIDUES OF PCBS IN HUMAN ADIPOSE TISSUE [ng/g]

	bject er, age)	PCB28	PCB52	PCB101	PCB118	PCB153	PCB138	PCB180
1	M,43	0.83	0.44	0.30	3.5	20.1	16.7	15.2
2	M,49	1.5	0.78	0.46	4.1	38.0	31.1	30.8
3	M,50	1.0	0.94	0.33	1.3	15.9	10.9	14.6
4	M,50	2.0	1.3	0.38	6.5	30.6	26.1	25.2
5	M,51	1.4	0.91	0.27	4.6	21.7	18.6	17.9
6	F,64	3.4	0.57	0.72	10.8	71.2	55.8	63.3
7	F,67	0.40	0.53	0.23	1.2	6.8	4.5	8.3
8	F,67	3.3	0.64	0.57	7.9	23.4	20.4	14.8
9	M,69	2.2	0.70	0.26	2.8	17.3	14.0	12.3
10	M,72	1.5	0.24	0.27	5.4	22.4	16.7	24.5
11	M,76	0.66	0.45	0.64	3.1	13.3	10.4	18.1
12	M,76	2.8	2.2	2.7	14.6	16.6	16.4	11.3
13	M,76	0.82	0.25	0.20	7.11	27.4	26.3	17.2
14	F,82	0.47	0.09	0.11	3.6	22.0	19.2	17.5

M – males

F – females

TABLE 2. RESIDUES OF PCBS IN HUMAN LIVER [ng/g]

	bject er, age)	PCB28	PCB52	PCB101	PCB118	PCB153	PCB138	PCB180
1	M,43	0.52	0.14	0.13	0.37	2.3	2.0	1.6
2	M,49	0.73	0.21	0.08	0.59	4.5	3.6	3.1
3	M,50	0.27	0.28	0.12	0.22	2.1	1.6	2.1
4	M,50	0.75	0.49	0.09	0.43	5.0	3.6	6.2
5	M,51	0.62	0.35	0.11	0.57	2.9	2.5	2.4
6	M,51	0.51	0.43	0.11	1.1	7.4	5.4	5.5
7	F,64	1.5	0.76	0.19	1.6	6.8	5.2	4.6
8	F,67	0.59	0.45	0.11	0.19	1.1	0.83	1.1
9	F,67	1.4	0.63	0.26	1.6	5.6	4.4	3.6
10	M,69	1.2	0.31	0.12	0.59	4.0	3.4	2.6
11	M,72	0.55	0.09	0.46	0.97	3.8	3.8	3.8
12	M,76	0.37	0.20	-	0.20	1.0	0.82	0.97
13	M,76	0.52	0.13	0.09	1.2	4.0	3.2	2.9
14	M,76	1.1	0.71	0.44	1.6	3.1	2.7	2.4
15	F,82	1.5	0.28	0.16	4.0	16.8	14.2	11.0

Average content of PCBs in adipose tissue and liver of the investigated samples was 74.4 ng/g (676.6 ng/g fat), and 14.4 ng/g, respectively. PCB concentration in adipose tissue is similar to the data for the population of other European countries in the period 1994-2003 and generally follows the same pattern of congener ratios, which are most similar to Aroclor 1254 (Table 2).

Country	Content of PCB congener [ng/g fat]							Ref.	
Country	PCB28	PCB 52	PCB 101	PCB 118	PCB 138	PCB 153	PCB 180	Total	Rel.
Poland 1994 (n = 20)	13.0	1.7	4.2	71.0	230.0	290.0	175.0	784.9	[0]
Belgium 2000 (n = 46)	2.8	2.7	3.0	57.1	68.3	145.3	93.7	372.9	[8]
Italy 2000 (n = 10)	*	*	3.0	20.7	58.0	112.0	85.5	279.2	[9]
Spain 2000 (n = 35)	4.9	0.9	2.0	47.0	220.0	300.0	280.0	854.8	
Sweden 2000 (n = 28)	4.1	1.4	2.3	40.0	230.0	300.0	200.0	777.8	[8]
Turkey 2001 (n = 29)	5.7	10.3	6.6	18.9	54.3	110.0	59.8	265.6	
Serbia 2003 (n=15)	14.5	6.4	4.8	49.7	225.7	186.4	188.9	676.4	Present study

TABLE 2. COMPARISON OF THE LEVELS OF PCBS IN ADIPOSE TISSUE FROM NOVI SAD<br/>(SERBIA) WITH SIMILAR STUDIES FROM SOME OTHER EUROPEAN COUNTRIES

\*Not analyzed.

The most abundant congeners detected in human adipose and liver tissue were PCB138, PCB153 and PCB180, comprising >80% of the total content of PCBs in the tissues (Table 3). This is in accordance with the fact that bioaccumulation of higher PCBs from the environment into the living organisms (including foodstuffs) is larger than for lower congeners, due to the increasing partition coefficients and lipophilicity.

Individual contents of total PCBs in test subjects are given in Figures 1 and 2.

TABLE 3. DISTRIBUTION OF PCB CONGENERS IN THE HUMAN TISSUE SAMPLES IN VOJVODINA

PCB congener	Human tissue samples [%]						
	Adipose tissue	Liver					
PCB 28	2	6					
PCB 52	1	2					
PCB 101	1	1					
PCB 118	7	7					
PCB 138	28	26					
PCB 153	33	33					
PCB 180	28	25					

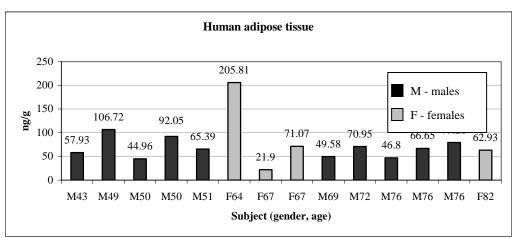


FIG. 1. CONTENT OF TOTAL PCBS IN THE HUMAN ADIPOSE TISSUE

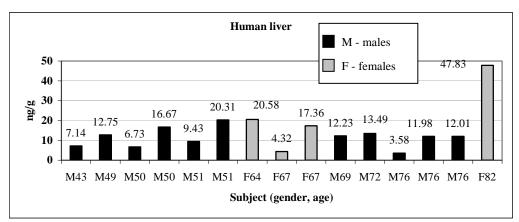


Fig. 2. Content of total PCBs in the human liver

Levels of PCBs in the investigated samples differ in adipose tissue and liver. The higher content was determined in adipose tissue for all investigated PCB congeners (Table 4).

AND LIVER FOR 7 INDIVIDUAL CONGENERS							
	Average conter	Ratio of PCB contents					
	Adipose tissue	Liver	(adipose tissue/liver)				
PCB 28	1.6	0.81	2.0				
PCB 52	0.71	0.36	2.0				
PCB 101	0.53	0.16	3.3				
PCB 118	5.5	1.0	5.5				
PCB 153	24.8	4.7	5.3				
PCB 138	20.5	3.8	5.4				
PCB 180	20.8	3.6	5.8				

 TABLE 4. RATIO OF PCB CONTENTS IN HUMAN ADIPOSE TISSUE

 AND LIVER FOR 7 INDIVIDUAL CONGENERS

Levels of PCBs in the investigated samples vary for the factor 10 (adipose tissue) to 13 (liver) between the highest and the lowest concentration. The difference between individual donors is probably due to their different backgrounds, occupation and dietary habits. According to the results, there is no significant correlation between age and gender of the donors and the levels of PCBs in their adipose tissue and liver. In both tissues, the highest content of PCBs was determined in a woman, aged 82. The total PCB body burden of investigated test subjects is relatively high and represents an important risk factor for possible adverse health effect in general population.

Although PCBs have been banned in Serbia for two decades, this study shows that the exposure to these contaminants seems to continue in Serbia in spite of the fact that it was carried out on the limited number of subjects and because of the selected population results are not representative for the whole Serbian population. Further research needs to be conducted in highly industrialized and polluted areas in Serbia and several environmental sources need to be studied (air, drinking water, sediments, foods, human milk, adipose tissue). In addition, studies should focus on the toxicological implications (for instance, effects on fertility) on humans and wildlife, which could be made possible by measuring exposure levels in these different sources.

#### ACKNOWLEDGEMENTS

This research was financially supported by the Ministry of Science and Environmental Protection of the Republic of Serbia (Development of new and improvement of the existing analytical methods and techniques in monitoring of the quality of the environment, Project No. 1622).

#### REFERENCES

- [1.] Stockholm Convention on Persistent Organic Pollutants, online at: http://www.pops.int/documents/background/proceedings/default.htm
- [2.] U.S. Environmental Protection Agency. PCB: cancer dose- response assessment and application to environmental mixtures. US Environmental Protection Agency, National Center for Environmental Assessment, Office of Research and Development, EPA/600/P-96/001; 1996.
- [3.] BORDET F, MALLET J, MAURICE L, BORRE S, VENANT A. Organochlorine pesticides and PCB congeners content of French human milk. *Bull Environ Contam Toxicol* 1993;50:425–32.
- [4.] PHILLIPS LJ, BIRCHARD GF. Regional variations in human toxic exposure in the USA: an analysis based on the National Human Adipose Tissue survey. *Arch Environ Contam Toxicol* 1991; 21: 159–168.
- [5.] VOJINOVIC-MILORADOV M, BUZAROV D, ADAMOV J, SIMIC S, POPOVIC E. Determination of polychlorinated biphenyls and polyaromatic hydrocarbons in frog liver. *Wat Sci Tech* 1996; 34: 153-156.
- [6.] ADAMOV J. Mathematical model of human dietary intake to persistent organic pollutants, *Ph.D. Thesis*, 2004: University of Novi Sad, Serbia and Montenegro.
- [7.] ADAMOV J, VOJINOVIC-MILORADOV M, JOVETIC S, BUZAROV D, Mathematical modeling of human dietary exposure to polychlorinated biphenyls, 24<sup>th</sup> International Symposium on halogenated Environmental Organic Pollutants and POPs, Berlin, Germany, Proceedings, 2004; 66, 2615.
- [8.] MARIOTTINI M, AURIGI S, FOCARDI S. Congener profile and toxicity assessment of polychlorinated biphenyls in human adipose tissue of Italians and Chileans. *Microchem J* 2000; 67:63–71.
- [9.] COK I, SATIROGLU MH. Polychlorinated biphenyl levels in adipose tissue of primiparous women in Turkey, *Env Int* 2004; 30:7–10.