Polychlorinated biphenyls in muscle tissue of freshwater fish in East Slovakia

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Summary

Levels of polychlorinated biphenyls (PCBs) in freshwater fish in the area of Chemko Strážske were compared to those of other regions of the Slovak Republic. The highest average PCB levels in samples from lake Zemplínska Šírava were identified in goldfish (108.8 $mg.kg^{-1}$) and the lowest ones in brown trout (13.7 $mg.kg^{-1}$). The average values measured for the country in general ranged between 0.01 and 5.1 $mg.kg^{-1}$ (northern pike and goldfish); the PCB levels measured in the contaminated area were 14 to 6 900 times higher than those in other regions of the country. Samples from the contaminated area showing values exceeding the limits made up as many as 75.8% (9.5% for the other regions of Slovakia).

Keywords

polychlorinated biphenyls; predatory and non-predatory freshwater fish; Slovak Republic; water streams; lake Zemplínska Šírava

Polychlorinated biphenyls (PCB) are compounds classified as persistent organic pollutants (POPs), capable of remaining in the environment and able to be transported, to accumulate in plant and animal tissues over long periods of time. When released into the environment, POPs may be transported by air or water to areas that are often rather distant from the place of origin. In such remote areas, the compounds may concentrate in living bodies, including humans, reaching concentrations, which may be harmful to health. An increased attention has been paid to polychlorinated biphenyls in recent decades because they significantly contribute to the pollution of the different components of environment.

In the former Czechoslovakia, PCBs had been produced at the Chemko Strážske chemical plant in the period of 1959–1984 as commercial PCB mixtures under the brand names Delor, Hydelor, and Delotherm [1]. During the PCB production at Chemko, extensive environmental leaks occurred, in particular via wastewater discharge. As a result, the sediments of the wastewater channel of Chemko, of the River Laborec, as well as of the Zemplínska šírava artificial lake (Fig. 1) have become contaminated. The region of Zemplín is considered among the most heavily PCB-burdened regions in Europe. At present, Chemko shows a relatively responsible attitude to the solution of problems of toxic wastes from PCB production stored within the company premises, and shows interest in participation in the Alternative PCB Destruction Project to be conducted under the sponsorship of UNIDO, the UN Agency for Economic Development, and under the co-funding by the World Bank Environmental Fund. One extensive problem however remains unaddressed [2], namely the collateral contamination of adjacent areas.

Exposure to high PCB doses has been associated with serious skin disorders (chloracne), itching and burning, eye irritation, pigmented skin and nail alterations, functional disorders of the liver and the immune system, irritation of the airways, headaches, dizziness, depression, loss of memory, irritability, fatigue, and impotence [3]. The various PCB forms are responsible for numerous toxic effects, including disorders of the immune and nervous system, reproductive abnormities, behavioural abnormities, and likely carcinogenicity (they induce cancer in animals, no similar effect however could be unambiguously demon-

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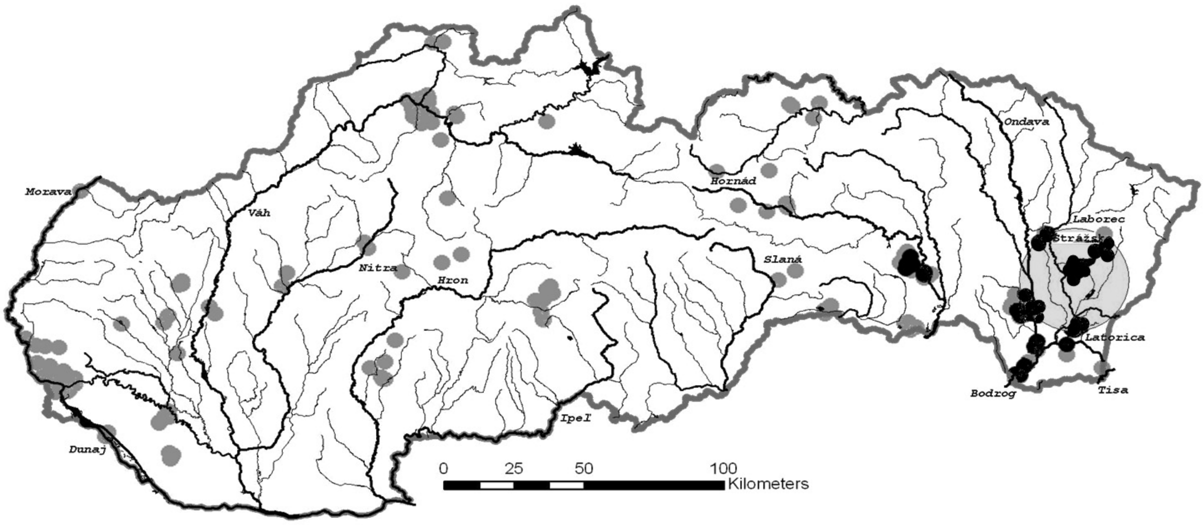

Fig. 1. Overview of sampling places and of fish samples from the Slovak Republic showing above-limit concentrations, and localisation of the polychlorinated biphenyls-contaminated region.

strated in humans) [4]. Strong toxicity may even result in death [5]. Children exposed to higher PCB levels during intrauterine development were reported to suffer from impaired visual cognition, impaired verbal memory and learning ability, lower IQ, and their ability to understand read text was delayed [6].

The toxic nature of even very low concentrations of PCBs was conclusively demonstrated in the seventies; in addition, the hazards of the presence of PCBs in the general environment and the food chain was shown to be enhanced by the ability of PCBs to accumulate in predominantly fat tissues of living organisms. These findings resulted in the use of PCBs in the OECD countries being restricted to closed systems. A quite different situation arised the former Czechoslovakia. Regardless of the alarming and generally available information about the hazards of PCBs, the production of PCBs kept increasing after 1972, peaking at around 1980, without any control of the associated health or environmental hazards. The PCB production was stopped in Czechoslovakia after identification of massive contamination of beef, milk and fish in 1984. In 1989 the distribution and sale of PCB-based products was finally discontinued [7].

The chemical and environmental stability of PCBs and their extensive use resulted in the contamination of the whole ecosystem, including the air, water and marine systems, waste deposits, sediments and all forms of life [5, 8]. PCBs get dissolved in water in minimum concentrations, and they may therefore spread in also aqueous environment. There, they get adsorbed to mud and sediments. In marine ecosystems PCBs get incorporated into the plankton and small arthropods, which are at the bottom of the food chain of fish, birds, and mammals. The incorporation is rather effective for the organisms at the end of the chain to take in significant amounts of PCBs with their food [9, 10]. Fish living over prolonged times in water contaminated by trace PCB concentrations are able to concentrate these substances up to a thousand fold. The distribution of PCBs in the bodies of the fish is not uniform. In carps, e.g., PCBs accumulate mainly in fat tissues, the head, the central nervous system, the gallbladder, and other internal organs, with concentrations in the blood and the smooth muscle being significantly smaller [10]. Biologists point to the fact that chemicals such as polychlorinated biphenyls and heavy metals influence the behaviour of animals: e.g., fish are hyperactive due to the action of chemicals upon the system of endocrine glands, fish get dazed, and galls lose orientation [11].

The most frequent way how PCBs get into the human body (apart from professional exposure) is food which accounts for as many as 95% of the intake [12]. As PCBs accumulate in fat tissue, they reach the highest concentrations in foods with high fat contents originating from PCB-contaminated regions, most of all from domestic breeds [13]. An additional important source is fish and bodies of game from PCB-contaminated regions [14]. Also, cases of direct contamination by PCB-containing products of edible oils were reported. For example the Belgium dioxin affair of 1999 caused by contamination by PCBs of fats used in feed mixtures [10, 15], or mass intoxications of humans due to PCB-contaminated rice oil from heat media in Japan in 1996–1998 and in Taiwan in 1978–1979 [16-20], as well as milk and butter contaminations by PCBs originating from paint in silage pits and agricultural machinery [10, 21-23].

Experts point not only to the positive aspects of fish consumption on human health (source of n-3 polyunsaturated fatty acids, source of minerals - Ca, P, marine fish iodine, and vitamins - A, B, D, E) [24, 25], they also warn of hazards to consumers' health caused by some toxic substances (heavy metals, organochlorine compounds and PCBs), which may accumulate in also freshwater fish. Numerous papers have been published suggesting the possibility of significant exposure to such toxic substances from fish. Thus the study by DARNERUD et al. [26] described problems connected with PCB- and dioxins- contaminated fish in Sweden, and provided advice on how to reduce the exposure in a programmatic manner. MENDOLA et al. [27] analysed the issue of the consumption of PCB-contaminated sport fish caught by anglers from the US. VELICER et al. [28] addressed issues of fishermen from the area of the Ontario Lake in the USA, including those concerning the extent of the consumption of contaminated fish and the associated health hazards. SVENSSON [29] compared human exposure to essential fatty acids and toxic substances from fish, including PCB, DDT, methyl mercury, dioxins and dibenzofurans. LOMMEL et al. [30] measured the concentrations of organochlorine pesticides, octachlorostyrene and mercury in the blood of people living in the area close to the River Elbe in Germany. Also, there have been reports in the Czech literature on contamination of freshwater fish by PCBs and other POPs in the ecosystem biota of the River Skalice [31, 32]. In Slovakia, contents of PCBs and other POPs have also been measured in the biota as part of the monitoring of persistent organic compounds, and the relations to the locality where the game or fish were caught and the degree of their contamination were addressed [33].

Consequently, situations may arise where increased fish consumption can be recommended, whereas consumption of excessively contaminated fish is warned against. This conflicting situation where benefits of the consumption of freshwater fish is opposed to the health hazards stemming from exposure to undesired substances may be reasonably resolved by assessing the assumed exposure levels to contaminants of potential consumers. As the basis, we must obtain knowledge of the values of the contamination of the fish and of the envisaged consumption volumes [34].

MATERIALS AND METHODS

The degree of the contamination by polychlorinated biphenyls of fish caught in waters of the Eastern Slovakian region (District Michalovce) was studied and compared with the values measured for samples of fish caught in other regions of the Slovak Republic. Altogether, 318 samples of fish muscle tissue were analysed, which had been taken in Slovakia between 2002 and 2004 as part of the project Monitoring of Huntable Game and Fish (hereinafter HGF), one of the three sub-systems of the project Partial Monitoring System of Foreign Substances in Food and Feed, and part of the Environmental Monitoring of the Slovak Republic. The aim of HGF is to observe penetration of contaminants into the bodies of wildlife and fish, which represents an objective indicator of the condition of the general environment within the

Tab. 1. Terms of sampling of particular predatory and non-predatory fish species in the Slovak Republic.

Predatory fish		Protective periods
Northern pike	*Esox lucius*	between 1 Jan. and 15 May
Zander	*Lucioperca lucioperca*	between 15 March and 15 June
European chub	*Leuciscus cephalus*	between 15 March and 31 May
Brown trout	*Salmo trutta m. fario*	between 1 Sept. and 15 April
Non-predatory fish		**Protective periods**
Common carp	*Cyprinus carpio*	between 15 March and 31 May
Goldfish	*Carassius auratus*	not set
Bream	*Abramis brama*	between 15 March and 31 May

Tab. 2. Overview of ΣPCB values measured for the different fish species from the area of lake Zemplínska šírava, District Michalovce (in $mg.kg^{-1}$ fresh weight).

Commodity	SNo	VEL	% SEL	Average	Median	95th percentile	Standard deviation
Common carp	67	40	59.7	33.406	9.767	111.189	79.506
Goldfish	17	16	94.1	108.752	41.702	442.594	156.880
Bream	18	18	100.0	76.551	44.356	228.489	86.564
European chub	20	15	75.0	36.645	4.332	99.713	64.474
Brown trout	16	11	68.8	13.671	13.435	34.331	12.800
Northern pike	7	6	85.7	55.339	41.535	165.746	71.777
Zander	16	16	100.0	65.485	67.763	103.677	28.824
Total	161	122	75.8	–	–	–	–

SNo - numbers of samples, VEL - samples with values exceeding the limit, % SEL - percentages of samples with values exceeding the limits.

Tab. 3. Overview of ΣPCB values measured for the various fish species from the territory of the Slovak Republic (in $mg.kg^{-1}$ fresh weight).

Commodity	SNo	VEL	% SEL	Average	Median	95th percentile	Standard deviation
Common carp	60	7	11.7	2.352	0.004	10.569	10.259
Goldfish	7	3	42.9	5.129	1.081	15.256	6.657
Bream	3	1	33.3	4.415	1.006	10.384	6.073
European chub	4	2	50.0	1.539	1.434	2.784	1.204
Brown trout	76	1	1.3	0.498	0.012	0.558	3.649
Northern pike	4	0	0.0	0.008	0.008	0.012	0.005
Zander	3	1	33.3	1.903	0.885	4.337	2.472
Total	157	15	9.5	-	-	-	-

SNo - numbers of samples, VEL - samples with values exceeding the limit, % SEL - percentages of samples with values exceeding the limits.

given region as wildlife and fish are the primary consumers in their respective ecosystems.

The monitoring (development of methodology, sampling plan, analyses and drafting of annual reports) is coordinated by the State Veterinary and Food Authority of the Slovak Republic. Regional Veterinary and Food Administrations and organisations of the Slovak Fishermen's Union participated in the sampling procedure. Three accredited institutions of the State Veterinary and Food Authority (Bratislava, Dolný Kubín and Košice) performed the analyses. The VÚP Food Research Institute, Bratislava approved the overall HGF strategy, it is also taking care of the coordination of the monitoring, processing and evaluation of information obtained, and of the review of annual reports.

Altogether, four species of predatory and three species of non-predatory fish were analysed; the dates of the sampling were chosen so as to be outside the closed game period of the different species (Tab. 1).

Each of the 318 samples of fish muscle tissue was analysed for the presence of six PCB congeners (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, PCB 180) using the GC–MS method (1 g fresh weight of fish tissue was extracted with a mixture of petroleum ether (90%) and acetone (10%) using the Accelerated Solvent Extraction (ACE) method. The extract was purified using a Florisil chromatographic column, STN EN 12393-2 [35]. For the purposes of this paper, PCB contents were evaluated as the sum of all the six PCB congeners. The results of the analyses obtained were statistically processed and evaluated pursuant to the Ordinance of Ministry of Agriculture of the Slovak Republic and of Ministry of Health of the Slovak Republic No. 981/1996-100 [36].

RESULTS AND DISCUSSION

Fish samples were taken from the territory of the Slovak Republic and in a targeted manner from the high-risk locality of the Eastern Slovakian region where significant environmental leaks, in particular via waste waters, had occurred in the past during PCB production at Chemko Strážske. As the result, sediments of the wastewater channel of Chemko, of the River Laborec, as well as the artificial lake Zemplínska Šírava were contaminated (Fig. 1).

Out of the total of 318 fish samples taken (Fig. 1), 137 (43.1%) samples exceeded the permissible concentration limits set for the different PCB congeners (where at least one of them exceeded the limit set by the Food Code of the Slovak Republic). The analyses included 146 samples from predatory and 172 samples from non-predatory fish. Every sample was analysed for the presence of 6 PCB congeners. Out of the total number of 161 samples of fish taken in the District Michalovce and analysed, applicable limit concentrations (Food Code of the Slovak Republic) were exceeded in 122 (75.8%) samples. There was a substantially smaller proportion of samples taken in other regions of the Slovak Republic showing values exceeding the limits set for the different congeners (9.5%). Higher contaminated samples originated mostly from areas of the Eastern Slovakian region, Districts Trebišov and Košice, i.e. the watershed of streams previously contaminated by polychlorinated biphenyls (Bodrog, Tisa, Latorica, Laborec and Hornád) (Tab. 2 and 3).

The average values of sum of 6 PCBs measured for samples of freshwater fish from the area of the Zemplínska šírava lake ranged from 13.671 to 108.752 mg.kg^{-1}, the corresponding figures measured for samples from other regions of the Slovak Republic were much smaller (between 0.008 and 5.129 mg.kg^{-1}); thus, the concentrations of PCBs in the samples from the contaminated area were 14 (common carp) to as many as 6 900 (Northern pike) times higher than those measured for the various fish species from other regions of Slovakia. Among the species monitored, the highest values were measured for samples from goldfish (from both regions compared), a fish living close to the bottom where insoluble PCB congeners could have adsorbed to sediments. The species living in the territory of Slovakia and being the least contaminated was northern pike; the least PCB concentrations in the area of lake Zemplínska Šírava were measured for brown trout. Similar values were also obtained for the 95th percentile: its values for District Michalovce ranged between 34.331 and 442.594 mg.kg^{-1}, and the average values for the Slovak Republic ranged between 0.012 and 15.256 mg.kg^{-1} (Tab. 2 and 3).

Comparing the different sampling periods (2002–2004), the highest PCB concentrations were measured for goldfish (133.716 and 139.554 mg.kg^{-1}) in Zemplínska Šírava (Fig. 2) during the initial years of the monitoring; high PCB concentrations were also measured for other fish species (northern pike, bream, and zander) in 2003. The concentrations measured for all the species monitored (save brown trout) were smaller

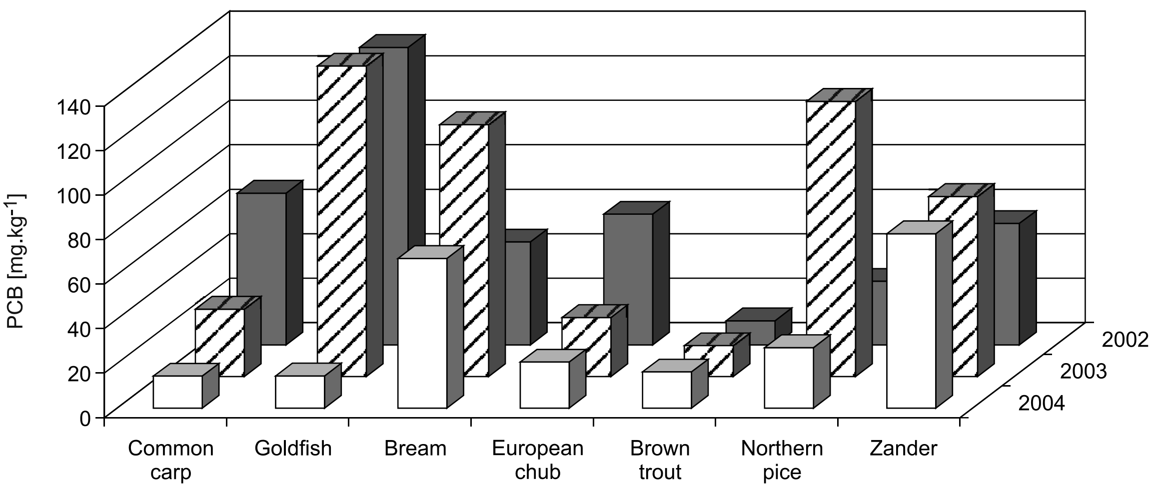

Fig. 2. Average ∑PCB concentration values measured for different fish species from the lake Zemplínska šírava, District Michalovce, in 2002–2004 (in mg.kg^{-1}).

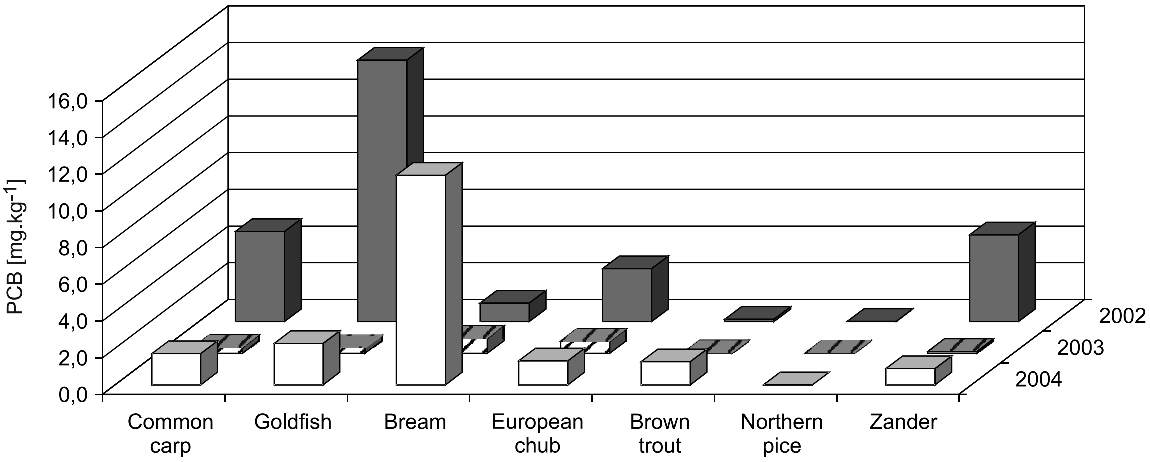

Fig. 3. Average ∑PCB concentration values measured for the various fish species from the Slovak Republic in 2002–2004 (in mg.kg^{-1}).

in the subsequent year, the reduction being most pronounced for goldfish (to reach the value of 14.485 mg.kg^{-1}). Among the freshwater fish species monitored, the smallest PCB concentrations were measured for brown trout (10.829 mg.kg^{-1} in 2002). PCB concentrations measured for non-predatory fish species showed a decreasing tendency since the start of the sampling, whereas the highest values for predatory species were measured in 2003 (except for brown trout, with PCB values showing gradual increases since 2002).

The average PCB concentrations measured for fish from other parts of Slovakia were markedly lower than those measured for the fish from the lake Zemplínska šírava (Fig. 3). The highest PCB values measured in 2002 were those obtained for samples of all the freshwater fish except for bream and brown trout, with the peak value as high as 14.247 mg.kg^{-1}measured for goldfish. The smallest PCB values were measured in 2003 for all predatory and non-predatory species (except for northern pike). There was a slight increase in the values for all fish species (except for bream) in the subsequent year (to reach 11.426 mg.kg^{-1}). The smallest PCB value was measured for northern pike in 2004 (0.003 mg.kg^{-1}).

Because of the gravity of findings of PCB concentrations in the fish, the State Veterinary and Food Authority (SVFA SR) in Michalovce introduced emergency veterinary measures in 2002, banning fishing and fish consumption (with the exception of „catch and release“ fishing for sports) throughout the region of lake Zemplínska šírava; corresponding warning boards were placed at the entrances to all recreation facilities. As any further dealing with these serious issues was within the purview of the Ministry of Environment and it was recognized as a serious environmental problem (the area concerned is one of major recreational areas in Slovakia), SVFA SR asked the Ministry of Environment of the Slovak Republic to urgently address the issue, to develop an overall strategy, and to consider making the affected region part of an international recovery project [37].

CONCLUSIONS

Regular monitoring of huntable game and fish, as part of the Monitoring of the General Environment of the Slovak Republic has confirmed that streams close to the Chemko Strážske chemiucal plant, where PCB production was concentrated in the past, have been contaminated by polychlorinated biphenyls as suggested by analyses of muscle tissue of predatory and non-predatory freshwater fish. The PCB contamination is excessive, with the values being as many as 14- to 6 900-fold those measured for other regions of Slovakia. The most contaminated fish species in both regions compared was goldfish (*Carassius auratus*). The smallest PCB values were measured in muscle tissue of northern pike (*Exox lucius*) from Slovakia, and for brown trout (*Salmo trutta m. fabio*) from the high-risk locality Michalovce. Being good indicators of natural pollution, fish tend to accumulate toxic substances in their bodies, thus jeopardising the health of the future consumers of the dietary, easy to digest meat. This hazard mainly concerns the river basins of the streams Bodrog, Tisa, Latorica, Laborec, Hornád as well as the areas adjacent to the water reservoir Zemplínska šírava, which is the second largest water reservoir in Slovakia (33 km^2). Evidently, the contamination of the area emanating from the Chemko Strážske chemical plant is that extensive that it will hardly be possible to eliminate it fully. All measures should nevertheless be taken to prevent further spreading of the contamination, and to decontaminate the most contaminated areas. Measures should at the same time be taken to mitigate the impacts of the contamination upon the population living in the region.

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