Polychlorinated dibenzo-*p*-dioxins, dibenzofurans and biphenyls in food samples from areas with potential sources of contamination in Slovakia

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Summary

The contents of persistent organic pollutants (POPs) involving 2,3,7,8-substituted polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs), dioxin-like (DL-) and non-dioxin-like polychlorinated biphenyls (NDL-PCBs) were determined in 243 food samples of animal origin collected in 2006–2007 from sites in five regions of Slovakia with industrial POP sources (Košice, Krompachy, Nemecká, Šaľa) and one background region (Starina). Milk (cows', goats', ewes'), pork (bacon, lard, liver), hen eggs, game and fish samples were pre-treated, extracted and, after semi-automated clean-up and fractionation, target compounds were determined by isotope dilution analysis using high-resolution gas chromatography coupled to high-resolution mass spectrometry. The total toxic equivalent ranges and medians in picograms per gram of fat were as follows: cows' milk 0.59–16, 3.0 (n = 31); goats' milk 0.88–33, 5.4 (n = 14); ewes' milk 0.61–20, 1.7 (n = 14); samples of pork origin 0.07–9.2, 0.55 (n = 38); hen eggs 0.19–181, 22 (n = 64); game 0.48–109, 4.8 (n = 45) and fish 0.04-6.5 pg·g^{-1} wet weight (median 0.88, n = 37). The levels of summed four NDL-PCB (IUPAC No. 101, 138, 153, 180) ranged between 0.19 ng·g^{-1} and 4 500 ng·g^{-1}, median 13 ng·g^{-1}.

Keywords

dioxins; furans; biphenyls; food; Slovakia

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs) are toxic compounds belonging to the group of persistent organic pollutants (POPs). In generally, these are characterized by high environmental and biological resistance with a high potential for long-range environmental transport. High solubility in lipids makes them suitable for significant bioaccumulation in living organisms. Levels of these widespread environmental contaminants increase along the food chain to humans. Unfortunately, exposure to POPs can result in a biochemical and toxic response, and in subsequent adverse health effects, from birth and developmental disorders to cancer, endocrine disruption, immune system dysfunction, thyroid disorders and dysglycemia, neurodegenerative diseases, dental defects, deficits in cochlear functions, hearing loss etc. [1–4].

It was found that the exposure of the general population to POPs occurs mainly through consumption of contaminated food, in particular high-fat foods of animal origin, such as milk and dairy products, animal meat, liver and fat from fatted cattle and wild game, some fish meat and eggs [5–8]. The exposure of animals depends in particular on the state of environment in the given area.

The most significant POP sources in Slovakia, with the potential for high formation and release of PCDDs and PCDFs to the environment, are high temperature metallurgical processes and municipal, hospital and hazardous waste incineration [9]. Smouldering of copper cables, cement kilns, pulp and paper production based on chlorine

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bleaching, uncontrolled combustion of household/garden waste, chemical production, fossil fuel burning and forest fires also belong to important dioxin sources [6, 10].

PCBs were produced in Chemko Strážske chemical plant in eastern Slovakia and were widely used in the industry in the past. Great production and extensive use of PCBs in everyday life led to their leakage and environmental pollution. Due to the persistence of PCBs, these compounds are still present in the environment.

Monitoring activities of POPs listed in the Stockholm Convention on POPs are recommended to evaluate the effectiveness of this treaty. Despite EU monitoring in the recent years, there was still little information about levels of POPs in the environment and food chain of Slovakia. This study was intended to fill this gap. It was focused on the impact of selected POP sources on the environmental contamination and the transfer of the contaminants to feed and food.

MATERIALS AND METHODS

Food samples and sampling sites

Altogether 243 food samples of animal origin were collected in 2006–2007 in four regions of Slovakia potentially contaminated with POPs (Šaľa, Nemecká, Košice, Krompachy), and in a background region of the Starina water reservoir (Fig. 1). The sampling areas were selected on the basis of existing data on polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDDs/Fs) in emissions in Slovakia, and on the basis of data from previous research projects [11–14]. The Duslo chemical plant producing pesticides, fertilizers, rubber-making and other chemicals with hazardous waste incineration is situated in the Šaľa region. Near Nemecká, Petrochema refinery is situated, with production of mineral oils, including transformer oils and with hazardous waste incineration. Near Košice, U.S. Steel metallurgical plant is situated, with iron ore sintering and municipal waste incineration. Near Krompachy, Kovohuty metallurgical plant is situated, with copper production and scrap copper processing. Details on the food samples are given in Tab. 1.

The food samples from big farms together with adipose tissue and meat from game animals and fish samples were collected in the target localities by Regional Veterinary and Food Administrations, organized by the State Veterinary and Food Administration of the Slovak Republic in Bratislava. Food samples originating from small family farms were collected by our department. A global positioning system (GPS) was used for precise localization of small family farms sampling points.

Standards, analytes and chemicals

We focused on selected pollutants included in the Stockholm Convention on POPs [10]. Com-

Fig. 1. Map of Slovakia with the investigated industrial regions Šaľa, Nemecká, Krompachy, Košice and a background area of the Starina water reservoir.

Tab. 1. Number and type of food samples collected from five selected potentially contaminated areas.

Sample		Area	Košice		Krompachy		Nemecká		Šaľa		Starina	
			SFF	BF	SFF	BF	SFF	BF	SFF	BF	SFF	BF
		Σ	*n*									
Cows' milk		31	4	4	2	2	7	4	0	3	2	3
Goats' milk		14	3	0	1	0	4	0	1	0	5	0
Ewes' milk		14	0	2	0	5	0	5	0	0	0	2
Hen eggs		64	10	2	13	0	18	0	11	2	8	0
Pork	Liver	38	0	0	0	2	3	1	0	4	3	0
	Bacon		2	0	1	3	4	0	0	3	2	0
	Lard		3	0	1	0	2	0	3	0	1	0
Fish	Predatory	37	0		0		0		1		2	
	Non-predatory		1		2		8		7		2	
	Other		7		6		0		0		1	
Game	Deer	45	5		8		6		1		6	
	Hare		0		0		0		7		0	
	Boar		2		0		2		0		0	
	Pheasant		1		0		0		7		0	
Total		243	46		46		64		50		37	

Σ – sum of food samples, *n* – number of food samples, SFF – small family farms, BF – big farms.

pounds of interest were seven 2,3,7,8-chlorosubstituted PCDDs and ten PCDFs, twelve dioxin-like PCBs (DL-PCBs), thereof four non-ortho (IUPAC No. 77, 81, 126 and 169) and eight mono-ortho PCB congeners (IUPAC No. 105, 114, 118, 123, 156, 157, 167 and 189) and six non-dioxin-like (marker) PCBs (NDL-PCBs, IUPAC No. 28, 52, 101, 138, 153 and 180).

In accordance with the isotope-dilution methods US EPA 1613 and US EPA 1668 [15, 16], internal (extraction) standard solution consisted of all ^{13}C-labelled compounds of interest except for 1,2,3,7,8,9-hexachlorodibenzo-*p*-dioxin and octachlorodibenzofuran. To determine recoveries of $^{13}C_{12}$-PCDD/F and non-ortho PCB congeners added to the sample before its extraction or cleanup, a recovery (syringe) standard solution consisting of 1,2,3,4-tetrachlorodibenzo-*p*-dioxin and 1,2,3,7,8,9-hexachlorodibenzo-*p*-dioxin was used. ^{13}C-labelled internal standard recoveries of mono-ortho-PCBs and NDL-PCBs were determined using a recovery standard of $^{13}C_{12}$-PCB-32 and $^{13}C_{12}$-PCB-188.

The following chemicals were used: acetone (SupraSolv; Merck, Darmstadt, Germany), *n*-hexane for the analysis of dioxins, furans and PCBs (Sigma-Aldrich, Steinheim, Germany), diethyl ether (extra pure, stabilized; Merck), ethanol (absolute; Merck), sulphuric acid (p.a., 95–97%; Merck), silica gel 60 (for column chromatography, particle size 0.063–0.200 mm; Merck), anhydrous sodium sulphate (residual analysis grade quality; Merck), nitrogen 3.0 and helium 4.6 (both from Messer Tatragas, Šaľa, Slovakia).

Extraction and clean-up

Food and fauna samples, except for hen eggs, were kept frozen (–20 °C) until processed. Meat samples from game animals, muscle meat of fish and porcine liver were homogenized in a laboratory blender, freeze-dried and the fat, together with fat-soluble organics, was extracted with *n*-hexane : acetone (4 : 1, v/v) using an accelerated solvent extractor (ASE 300; Dionex, Sunnyvale, California, USA).

Adipose tissue samples were cut into approx. 5 mm pieces, mixed with anhydrous sodium sulphate, deeply frozen using liquid nitrogen and then homogenized in a laboratory blender. After homogenization, a glass column was packed with the mixture and fat extracted with *n*-hexane.

Egg yolks were mixed with anhydrous sodium sulphate to get a fine powder that was poured into a glass column. Fat and dissolved organics were extracted with diethyl ether.

Milk fat was extracted by shaking the sample in a separating funnel with a mixture ethanol : diethyl ether : *n*-hexane (1 : 0.4 : 0.6, v/v) and, afterwards, twice with *n*-hexane.

After the gravimetric determination of the

contents of fat, internal standard solutions of ^{13}C-labelled surrogates were added to the fat sample diluted in *n*-hexane. The fat extracted was treated on the prepared high-capacity 44% H_2SO_4/silica column. The pre-cleaned extract, free of most lipids, was then cleaned-up and fractionated using a semiautomatic PowerPrep system (Fluid Management Systems, Waltham, Massachusetts, USA) equipped with three commercial disposable columns: multilayer column containing H_2SO_4/silica and KOH/silica, basic alumina one and active carbon one (Fluid Management Systems). Two fractions were captured: the *n*-hexane – dichloromethane (1 : 1) fraction from the basic alumina column, which contained mono-ortho PCBs and NDL-PCBs, and the toluene fraction from the carbon column, which contained PCDDs/Fs and non-ortho PCBs. Both fractions were carefully concentrated using the TurboVap II concentrator (Zymark, Hopkinton, Massachusetts, USA) and quantitatively transferred into conical 1.1 ml vials.

Gas chromatographic and mass spectrometry

After dilution with the $^{13}C_{12}$-labelled recovery standard solutions, the fractions were analysed by high-resolution gas chromatography – high-resolution mass spectrometry (HRGC-HRMS). For determination of target compounds, an HP 6890 gas chromatograph (Hewlett-Packard, Palo Alto, California, USA) operating in the splitless mode, with the flow rate of helium 1 ml·min^{-1}, coupled to an MAT 95XP mass spectrometer (Thermo Finnigan, Bremen, Germany) was used.

PCDDs, PCDFs and non-ortho PCBs present in the carbon column toluene fraction were injected (5 μl) into GC and separated on a polar (90% biscyanopropyl – 10% cyanopropylphenyl polysiloxane) capillary column Rtx-2330 (60 m × 0.25 mm × 0.1 μm; Restek, Bellefonte, Pennsylvania, USA). Injector and GC-MS transfer line temperature were 280 °C and 260 °C, respectively. The following GC oven temperature programme was used: from initial temperature 120 °C (1.5 min) to 200 °C at a rate of 30 °C·min^{-1} and then 2 °C·min^{-1} to 260 °C.

NDL-PCBs and mono-ortho PCBs present in the alumina column fraction were injected (4 μl) into GC and the compounds were separated on a non-polar (5% phenyl methylpolysiloxane) DB-5MS capillary column (60 m × 0.25 mm × 0.25 μm; J&W Scientific, Folsom, California, USA). Temperatures of both the injector and the transfer line were 280 °C. The following temperature program was used: from initial temperature 120 °C (1.5 min) to 200 °C at a rate of 30 °C·min^{-1}, and then 2 °C·min^{-1} to 287 °C. The analysis was performed by isotope-dilution methods [15–16]. The HRMS MAT 95XP (Thermo Finnigan) was operating at 10 000 resolution (10% valley) and 48 eV electron energy set. In the multiple ion detection (MID) mode, two most abundant ions for each congener, i. e. $[M+2]^+$, and M^+ or $[M+4]^+$, and their ^{13}C-labelled equivalents were monitored.

Quality Assurance/Quality Control (QA/QC)

Each analytical batch included ten food samples and one method blank. Every third batch contained also a sample of reference material to check the accuracy of the analytical process. Certified reference material CIL-EDF-2525 (fish; Cambridge Isotope Laboratories, Andover, Massachusetts, USA) and an in-house reference material (lard spiked with known amounts of PCDDs/Fs and PCBs) were used. Control charts were plotted for QC samples and blanks as a basis for the check of accuracy, precision and reliability of the analytical process. All measurements were carried out in an accredited laboratory (ISO/IEC 17 025), which successfully participated in a proficiency test on the determination of PCDDs/Fs and PCBs in food (2008, 2009) organized by European Union Reference laboratory for Dioxins and PCBs in Feed and Food, State Institute for Chemical and Veterinary Analysis of Food, Freiburg, Germany.

RESULTS AND DISCUSSION

The levels of 2,3,7,8-substituted PCDDs and PCDFs, and DL-PCBs expressed as toxic equivalent (*TEQ*) and the levels of summed four NDL-PCBs (IUPAC No. 101, 138, 153 and 180) are presented in Tab. 2. If some congener was present at a level lower than its limit of detection (*LOD*), *LOD* value was used for *TEQ* calculation.

Polychlorinated dibenzo-*p*-dioxins, dibenzofurans and dl-PCBs

Since EU legislation concerning PCDD/F and DL-PCB presence in food and feed is based on the toxic equivalent factor (TEF) and *TEQ* concept, all PCDD/F and DL-PCB levels measured in this study were expressed as picograms of toxic equivalent per gram. For *TEQ* calculation, the toxic equivalent factors according to World Health Organisation (WHO_{1998} TEFs) [17] were used. WHO_{2005} TEFs [18], which are actually in force, were not used because all samples were collected and analysed before introducing the WHO_{2005} TEFs and before their acceptance in the amended EU legislation related to maximum levels (ML) for PCDDs/Fs and DL-PCBs in food and feed

Tab. 2. PCDD, PCDF, DL-PCB and sum of NDL-PCB contents in food samples from Slovakia.

Sample		WHO_{1998} *TEQ* (WHO_{2005} *TEQ*) [pg·g^{-1}]					Σ NDL-PCBs [ng·g^{-1}]
		PCDDs	PCDFs	Non-ortho DL-PCBs	Mono-ortho DL-PCBs	Total *TEQ*	
Cows' milk (*n* = 31)	Min–Max	0.03–3.0 (0.03–3)	0.21–5.4 (0.14–3.9)	0.26–8.7 (0.27–9.4)	0.05–1.2 (0.01–0.24)	0.59–16 (0.48–15)	1.9–49
	Mean	0.54 (0.54)	1.2 (0.82)	2.8 (2.9)	0.38 (0.07)	5.0 (4.4)	15
	Median	0.25 (0.25)	0.84 (0.58)	1.5 (1.6)	0.30 (0.05)	3.0 (2.6)	9.3
Goats' milk (*n* = 14)	Min–Max	0.06–9.2 (0.06–9.2)	0.18–11 (0.13–7.7)	0.45–20 (0.45–20)	0.08–2.3 (0.02–0.44)	0.88–33 (0.78–30)	2.0–104
	Mean	1.2 (1.2)	2.1 (1.5)	5.7 (5.8)	0.58 (0.11)	9.6 (8.7)	22
	Median	0.42 (0.42)	0.91 (0.63)	3.3 (3.4)	0.27 (0.05)	5.4 (4.8)	9.9
Ewes' milk (*n* = 14)	Min–Max	0.02–5.0 (0.02–5)	0.15–5.7 (0.11–4.0)	0.29–8.6 (0.31–9.3)	0.04–0.31 (0.01–0.04)	0.61–20 (0.54–18)	1.4–12
	Mean	0.84 (0.84)	1.1 (0.77)	1.9 (2.1)	0.16 (0.02)	4.0 (3.7)	4.8
	Median	0.15 (0.15)	0.30 (0.23)	1.1 (1.1)	0.15 (0.02)	1.7 (1.5)	4.4
Pork (*n* = 38)	Min–Max	0.02–4.0 (0.02–4.4)	0.03–7.5 (0.02–5.5)	0.003–1.3 (0.004–1.3)	0.01–0.95 (0.001–0.1)	0.07–9.2 (0.06–7.8)	0.19–55
	Mean	0.23 (0.24)	0.84 (0.63)	0.21 (0.23)	0.11 (0.01)	1.4 (1.1)	8.0
	Median	0.08 (0.08)	0.18 (0.12)	0.09 (0.10)	0.04 (0.01)	0.55 (0.36)	3.0
Hen eggs (*n* = 64)	Min–Max	0.03–23 (0.03–23)	0.07–55 (0.05–38)	0.06–89 (0.06–92)	0.04–40 [a] (0.01–5.7) [a]	0.19–181 [a] (0.14–138) [a]	2.0–4 500 [a]
	Mean	2.7 (2.7)	7.0 (5.1)	16 (16)	6.5 [a] (1.0) [a]	32 [a] (25) [a]	435 [a]
	Median	1.1 (1.1)	4.7 (3.3)	7.8 (8.0)	3.0 [a] (0.40) [a]	22 [a] (17) [a]	158 [a]
Game (*n* = 45)	Min–Max	0.05–9.0 (0.05–9.0)	0.23–62 (0.17–42)	0.13–37 (0.19–38)	0.07–1.2 (0.01–0.22)	0.48–109 (0.42–89)	3.2–111
	Mean	1.1 (1.1)	3.0 (2.0)	3.9 (4.1)	0.44 (0.08)	8.4 (7.3)	18
	Median	0.67 (0.67)	1.3 (0.91)	2.4 (2.5)	0.40 (0.07)	4.8 (4.1)	11
Fish [b] (*n* = 37)	Min–Max	0.003–0.74 (0.003–0.74)	0.01–1.3 (0.01–0.97)	0.02–3.5 (0.02–3.6)	0.005–2.7 (0.001–0.65)	0.04–6.5 (0.04–5.0)	0.22–77
	Mean	0.10 (0.1)	0.22 (0.16)	0.82 (0.84)	0.35 (0.07)	1.5 (1.2)	15
	Median	0.04 (0.04)	0.12 (0.09)	0.46 (0.47)	0.27 (0.04)	0.88 (0.64)	13

Values are expressed per gram of fat.
WHO_{1998} *TEQ*, WHO_{2005} *TEQ* – toxic equivalent calculated according to toxic equivalent factors of WHO_{1998} [17] and WHO_{2005} [18], respectively, PCDDs – polychlorinated dibenzo-*p*-dioxins, PCDFs – polychlorinated dibenzofurans, DL-PCBs – dioxin-like polychlorinated biphenyls, Σ NDL-PCBs – sum of non-dioxin-like polychlorinated biphenyls.
a – mono-ortho PCBs and NDL-PCBs were determined in 63 samples only, b – values are expressed per gram of wet weight.

as late as in 2012. It means that the determined PCDD/F/DL-PCB levels were evaluated according to ML and action levels for PCDDs/Fs and PCDDs/Fs/DL-PCBs in foodstuffs set in the Commission Regulation (EC) No 1881/2006 [19] and Commission Recommendation 2006/88/EC [20] respectively (Tab. 3), i.e. legislation then in force. It was shown [21] that *TEQ* calculated by using WHO_{2005} TEFs are on average 14% lower than those calculated by using WHO_{1998} TEFs. Findings of this study correspond with those of Malish et al. [21] but, in some cases, the differences were slightly higher. Total *TEQ* values calculated for 243 analysed food samples using WHO_{2005} TEFs were on average lower than *TEQ* calculated using WHO_{1998} TEFs as follows: 13% for milk and game samples, 19% for hen eggs and fish samples, and 23% for pork samples. The arithmetic mean, median, minimum and maximum upper bound *TEQ* levels of PCDDs/Fs and DL-PCBs calculated by both WHO_{1998} TEFs and WHO_{2005} TEFs are presented in Tab. 2.

Cows' milk

The highest total *TEQ* was determined in a cows' milk sample from a farm at a village in the Krompachy region. A value of 16 pg·g^{-1} fat is almost 3-times higher than a maximum level of

6 pg·g^{-1} fat set by the European Commission (EC). PCDDs/Fs contribution to this level (6.8 pg·g^{-1} fat) was more than twice higher than ML. The highest PCDD/F-*TEQ* was determined in a cows' milk sample from a farm at another village in the Krompachy region (8.4 pg·g^{-1} fat), which was almost 3-times higher than ML. Similarly, other two cows' milk samples from the Krompachy region (both from a farm nearby Krompachy) showed values well above the maximum level set by EC either in 2006 or 2011, even though they would need recalculation using WHO_{2005} TEFs. The minimum and maximum *TEQ* in four cows' milk samples from Krompachy region were 4.1 pg·g^{-1} and 8.4 pg·g^{-1} fat for PCDDs/Fs, and 5.9 pg·g^{-1} and 9.2 pg·g^{-1} fat for DL-PCBs. This area was the most polluted by PCDDs/Fs out of all studied regions, regardless of whether the sample was from a small family farm or a big farm.

The second highest levels of PCDDs/Fs and DL-PCBs were observed in the Nemecká region, where 6 out of 11 samples exceeded the maximum level for total *TEQ*, thereof three samples almost twice (10 pg·g^{-1}, 11 pg·g^{-1} and 11 pg·g^{-1} fat). PCDDs/Fs were below ML and only three samples from the Nemecká region slightly exceeded the action level (2.1 pg·g^{-1}, 2.7 pg·g^{-1} and 2.9 pg·g^{-1} fat). Dioxin-like PCBs had the largest contribution to total *TEQ* in the Nemecká region, accounting mostly for more than 70%, with a maximum of 85%, whereas only for 50–60% in other regions. In Nemecká village, which is adjacent to the refinery, the levels were higher than in villages more distant from the pollution source.

In the Košice region, only one sample exceeded the action level for PCDDs/Fs, and three samples out of 8 were above the action level for DL-PCBs.

In the Šaľa region and Starina background region, the determined levels were well below the maximum level and action level.

Important finding was that, in Krompachy region, the samples from small family farms and big farms had similar highest levels (Fig. 2). These results clearly indicate the high contamination of the local environment. In other polluted regions, a marked difference between the highest levels in samples from small family farms compared to samples from big farms were observed.

Goats' milk

All the goats' milk samples were collected from small family farms. The highest total *TEQ* in goats' milk was determined in a sample collected from a village in the Krompachy region (33 pg·g^{-1} fat), whereas the contribution of PCDDs/Fs was 20 pg·g^{-1} fat. It represented 5.4- and 6.6-fold excess above ML, respectively. Another sample from Krompachy region had also the levels of PCDD/F-*TEQ* and total *TEQ* above ML (6.3 pg·g^{-1} and 12 pg·g^{-1} fat, respectively). These two samples

Tab. 3. Maximum and action persistent organic pollutants levels in foodstuffs according to EU legislation [19, 20, 23].

Foodstuffs	Maximum level		Action level		Maximum level
	PCDDs/Fs	PCDDs/Fs/DL-PCBs	PCDDs/Fs	DL-PCBs	Σ NDL-PCBs [ng·g^{-1}]
	Toxic equivalent [pg·g^{-1}]				
Meat, meat products and fat of the following animals[a]:					
bovine animals and sheep	3.0	4.5	1.5	1.0	40
poultry	2.0	4.0	1.5	1.5	40
pigs	1.0	1.5	0.6	0.5	40
Liver of terrestrial animals mentioned above, and derived products thereof	6.0	12.0	4.0	4.0	40
Muscle meat of fish and fishery products[b]	4.0	8.0	3.0	3.0	125[c]
Raw milk and dairy products, including butterfat	3.0	6.0	2.0	2.0	40
Hen eggs and egg products	3.0	6.0	2.0	2.0	40

Values are expressed per gram of fat. The maximum level applies to crustaceans, excluding the brown meat of crab and excluding head and thorax meat of lobster and similar large crustaceans. Persistent organic pollutants levels are expressed on a wet weight basis.
PCDDs – polychlorinated dibenzo-*p*-dioxins, PCDFs – polychlorinated dibenzofurans, DL-PCBs – dioxin-like polychlorinated biphenyls, Σ NDL-PCBs – sum of non-dioxin-like polychlorinated biphenyls.
a – excluding edible offal; b – excluding eel; c – muscle meat of wild caught fresh water fish, with the exception of diadromous fish species caught in fresh water, and products thereof

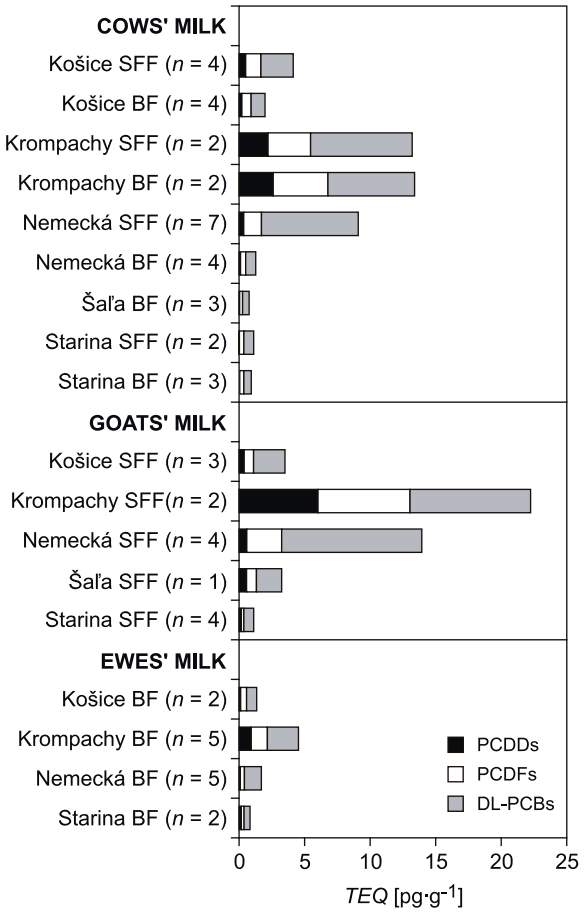

Fig. 2. Comparison of *TEQ* medians in milk samples.

SFF – small family farms, BF – big farms, *n* – number of samples, *TEQ* – toxic equivalent (expressed on fat basis), PCDDs – polychlorinated dibenzo-*p*-dioxins, PCDFs – polychlorinated dibenzofurans, DL-PCBs – dioxin-like polychlorinated biphenyls.

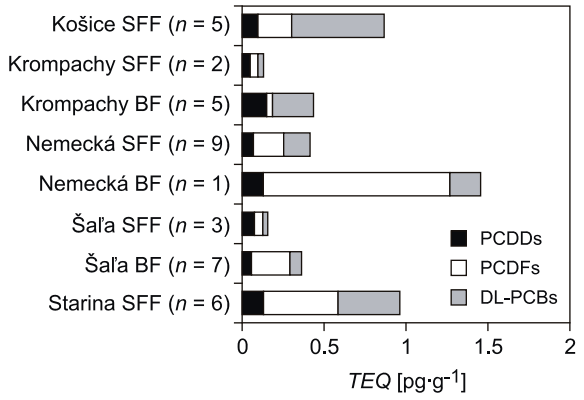

Fig. 3. Comparison of *TEQ* medians in pork samples.

SFF – small family farms, BF – big farms, *n* – number of samples, *TEQ* – toxic equivalent (expressed on fat basis), PCDDs – polychlorinated dibenzo-*p*-dioxins, PCDFs – polychlorinated dibenzofurans, DL-PCBs – dioxin-like polychlorinated biphenyls.

of goats' milk from the Krompachy region had two of the highest contributions of PCDDs/Fs to total *TEQ* (53% and 61%).

The second highest total *TEQ* level (28 pg·g^{-1} fat), exceeding ML almost 5-fold, was observed in a sample from the Nemecká region. Moreover, PCDD/F-*TEQ* content (6.0 pg·g^{-1} fat) in the sample was twice as high as ML for PCDDs/Fs in milk. The other two samples from the Nemecká region only slightly exceeded the maximum level and action level for PCDDs/Fs. Nevertheless, DL-PCBs present in the samples mostly contributed (around 70–80%) to total *TEQ*, whereas in other regions, the contribution was about 40–60% except for a sample from the Košice region, in which it was 89%. Comparison of *TEQ* medians in goats' milk samples is presented in Fig. 2.

Ewes' milk

All 14 ewes' milk samples were collected from big farms. Two of them contained substantially higher total *TEQ* levels than the remaining ones (20 pg·g^{-1} and 14 pg·g^{-1} fat). Not surprisingly, the samples were collected from flocks of sheep grazed in the Krompachy area in the neighbourhood of the metallurgical plant processing copper scrap. This plant seems to be a significant source of these compounds in the region, which was confirmed by the findings of high PCDD/F levels in ambient air samples taken around the plant in 2007 [22]. However, no ewes' milk sample originating from animals grazed farther from the potential source of pollution exceeded the ML value. Comparison of *TEQ* medians in ewes' milk samples is presented in Fig. 2.

Samples of pork origin

Porcine meat, liver, bacon and lard belong to favourite foods in Slovakia. PCDD/F and/or DL-PCB levels especially in porcine liver indicate that the levels in farmed animals are often markedly lower than those from small family farms. The reason might be the fact that, in hog industry, the animals are kept inside buildings without any contact with outside possibly contaminated environment and are fed with less contaminated feed due to its testing on the presence of pollutants. This is in contrast to animals bred at small family farms in fenced pens enabling them to be in contact with soil. Such animals are fed with feeds of local origin without any testing of PCDD/F/PCB content.

Only three pork samples exceeded ML (Tab. 3) – a sample of bacon from a village near the ironworks in the Košice region exceeded ML for total *TEQ* (3.0 pg·g^{-1} fat), and two samples of

pig liver from Nemecká village and a village in the background Starina area contained PCDD/F-*TEQ* above ML (7.7 pg·g^{-1} and 8.2 pg·g^{-1} fat, respectively). Total *TEQ* levels similar to those determined within this study (0.07–9.2 pg·g^{-1} fat; $n = 38$) were found in pig samples tested within monitoring activities in Slovakia in 2008–2011 (0.02–2.4 pg·g^{-1} fat; $n = 12$).

All samples of pork liver irrespective of the origin had very high contribution of PCDDs/Fs to total *TEQ* (66–95%, median 88%) in comparison to bacon and lard samples (12–80%, median 50%). Lower contribution of DL-PCBs may indicate a faster metabolism of PCBs in pig liver compared to dioxins and furans [7]. Comparison of *TEQ* medians in pork samples is presented in Fig. 3.

Hen eggs

In general, samples of hen eggs were collected from small family farms, i.e. from hens containing the highest PCDD/F and DL-PCB levels from all the foodstuffs analysed in the study (Fig. 4). It means that the increased consumption of such hen eggs may significantly contribute to exposure to dioxins, furans, PCBs and other POPs. On the contrary, hen eggs originating from big farms with cage factory farming generally contained POPs at levels well below ML [7].

Only 7 of 64 small family farm hen egg samples did not exceed ML for total *TEQ*. The highest total *TEQ* level was determined in a sample from Kluknava village not far from the copper producing plant in Krompachy (181 pg·g^{-1} fat; thereof PCDD/F-*TEQ* 52 pg·g^{-1} fat). The second highest total *TEQ* was found in a sample directly from Krompachy town (153 pg·g^{-1} fat) with the highest level of PCDDs/Fs out of all 243 food samples (78 pg·g^{-1} fat).

Out of 12 samples from the Košice region, 7 were approx. twice higher than ML for PCDDs/Fs, and one sample even 8-fold higher (25 pg·g^{-1} fat). The most relevant contamination source seems to be the ironworks in Košice since all such samples originated from the adjacent villages.

Most of small family farm egg samples from the Nemecká and Šaľa regions exceeded ML for PCDDs/Fs from 2- to 4-fold (max. value 12 pg·g^{-1} fat) and from 2- to 6-fold (20 pg·g^{-1} fat), respectively. Total *TEQ* of all 18 egg samples from the Nemecká region were from 2- to 12-fold above ML, with the highest value of 74 pg·g^{-1} fat in eggs from a small family farm close to the petrochemical plant. In other 5 samples, it ranged from 60 pg·g^{-1} to 72 pg·g^{-1} fat. Levels in eggs from the background Starina region were mostly below ML. As regards the contribution of PCDDs/Fs and DL-PCBs to the total *TEQ*, predominant contributors in the Krompachy area were PCDDs/Fs (29–77%, median 65%) and, in the Nemecká region, DL-PCBs (50–94%, median 85%).

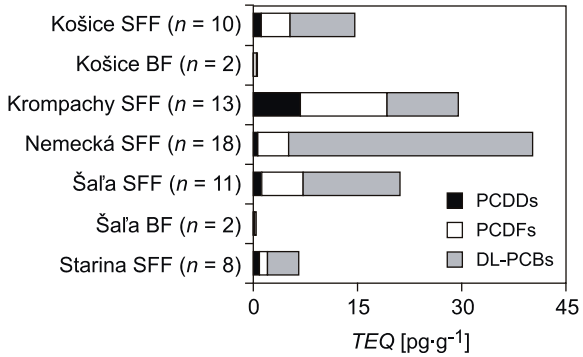

Fig. 4. Comparison of *TEQ* medians in hen egg samples.

SFF – small family farms, BF – big farms, *n* – number of samples, *TEQ* – toxic equivalent (expressed on fat basis), PCDDs – polychlorinated dibenzo-*p*-dioxins, PCDFs – polychlorinated dibenzofurans, DL-PCBs – dioxin-like polychlorinated biphenyls.

Fish samples

PCDD/F and DL-PCB levels in fish muscle meat samples analysed within this study are expressed as picograms per gram wet weight (ww). The levels of these pollutants in the fish samples were predominantly well bellow the maximum level and action level (Fig. 5). Only two samples from the Košice region and four samples from Krompachy town slightly exceeded the action level for DL-PCBs (3.1 pg·g^{-1}, 5.1 pg·g^{-1} and 3.1 pg·g^{-1}, 3.4 pg·g^{-1}, 3.4 pg·g^{-1}, 4.1 pg·g^{-1} ww, respectively).

Predominant contributors to the total *TEQ* in fish samples in the Košice, Krompachy and Nemecká regions were DL-PCBs (median approx.

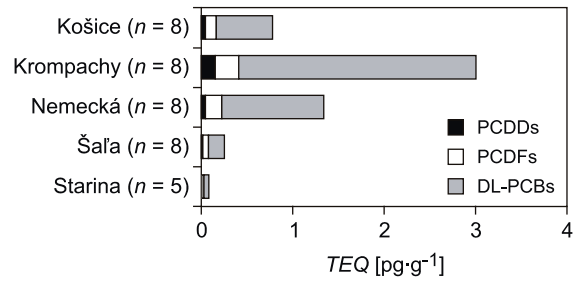

Fig. 5. Comparison of *TEQ* medians in fish samples.

n – number of samples., *TEQ* – toxic equivalent (expressed on fat basis), PCDDs – polychlorinated dibenzo-*p*-dioxins, PCDFs – polychlorinated dibenzofurans, DL-PCBs – dioxin-like polychlorinated biphenyls.

80%). In the Šaľa and Starina regions, DL-PCBs contributed by 70% and 60%, respectively. In general, fish and fishery products had no significant contribution to the human daily intake of PCDDs/Fs/DL-PCBs due to their relatively low consumption in Slovakia, and because they did not originate from contaminated rivers, lakes or water reservoirs.

Wild game

Although no maximum and action levels are set by EC for this type of foodstuff, analysis of samples of wild game animals may produce information on the contamination of the local environment. The levels of PCDDs/Fs/DL-PCBs in Nemecká, Šaľa and Starina regions, expressed as total *TEQ*, were only in some cases (9 from 29 samples) slightly above 5 pg·g^{-1} fat (Fig. 6). In one sample from Nemecká region, total *TEQ* was slightly higher (13 pg·g^{-1} fat). In Krompachy region, in 5 out of 8 samples, this level was approx. 5 pg·g^{-1} fat and in two samples it was slightly above 15 pg·g^{-1} fat. The highest total *TEQ*, from samples originating from Krompachy region, was found in adipose tissue of deer (29 pg·g^{-1} fat). In Košice region, 6 out of 8 samples had the level around 5–10 pg·g^{-1} fat. The lowest total *TEQ* level was found in a sample of boar (0.48 pg·g^{-1} fat) and the highest one, from all wild game samples, was found in deer (109 pg·g^{-1} fat), both from sites farther from the ironworks in Košice. Whereas samples from Krompachy and Nemecká regions had higher contribution of DL-PCBs to total *TEQ* (58–73%), the most polluted sample of deer liver contained more PCDDs/Fs (65%). The higher POP levels in this sample might have been caused by feed composition or quality of the watering place. The sample was obtained in area were a waste dump is located.

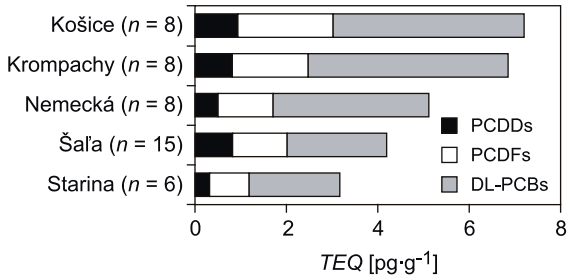

Fig. 6. Comparison of *TEQ* medians in game samples.

n – number of samples, *TEQ* – toxic equivalent (expressed on fat basis), PCDDs – polychlorinated dibenzo-*p*-dioxins, PCDFs – polychlorinated dibenzofurans, DL-PCBs – dioxin-like polychlorinated biphenyls.

Other polychlorinated biphenyls

Maximum levels of the sum of six NDL-PCBs in foodstuffs, according to the Commision Regulation (EC) No 1259/2011 [23], are listed in Tab. 3. In all food samples from the study, only the sum of four NDL-PCBs (IUPAC No. 101, 138, 153 and 180) is presented.

In the Nemecká region, only three samples of cows' milk from small family farms exceeded slightly ML, with the highest sum of NDL-PCBs of 49 ng·g^{-1} fat. Similarly, in other samples from this village adjacent to the refinery, levels close to ML were observed. Other samples from villages more distant from this pollution source, as well as all other cows' milk samples, had very low levels of NDL-PCBs.

Also in goats' milk, summed NDL-PCB levels were above ML only in two cases. Both were from Nemecká region, with the highest sum at 104 ng·g^{-1} fat. Other samples from Nemecká, Košice and Starina had higher NDL-PCB levels than samples from other sites, but these were still below ML.

Ewes' milk samples were collected only from big farms. These were the least polluted from all samples in this study. All sums of 4 NDL-PCBs were below ML, with the highest level in a sample from a village in Nemecká region (12 ng·g^{-1} fat).

From samples of pork origin, only two were above ML. These two highest sums of NDL-PCBs were determined in bacon and lard from a small family farm in Košice region (55 ng·g^{-1} and 41 ng·g^{-1} fat, respectively). Around a half of ML (14–27 ng·g^{-1} fat) was observed in lard samples from Nemecká, Starina and Šaľa, in bacon from Košice and Krompachy (big farm) and in a liver from Starina region. All other samples of pork origin were well below ML.

Hen eggs were the most contaminated from all samples, except for four eggs from big farms. Noticeably, the most polluted egg samples were those from sites in Košice (4 500 ng·g^{-1} fat), Krompachy (3 240 ng·g^{-1} and 1 227 ng·g^{-1} fat), Šaľa (1 827 ng·g^{-1}, 1 726 ng·g^{-1} and 1 351 ng·g^{-1} fat) and Nemecká regions (1 190 ng·g^{-1} fat). These levels represented a 30- to 90-fold excess of ML. High levels of NDL-PCBs were observed also in a half of eggs from sites in Nemecká, Krompachy and Šaľa regions (600–769 ng·g^{-1} fat). All other egg samples had lower sums of NDL-PCBs, but these were still above ML, except for around 20 samples. It is not surprising that the least polluted egg samples were collected from big farms in Šaľa and Košice regions (2.0 ng·g^{-1} and 2.3 ng·g^{-1} fat, respectively).

All fresh water fish samples had the sum of NDL-PCBs below ML. The highest level (77 ng·g^{-1} ww) was observed in fish from a gravel

pit, about 8 km down the valley from the iron-works in Košice, close to the border with Hungary. Five from eight samples from Krompachy, and two samples from Košice region, were below ML (29–36 ng·g^{-1} ww). All other fish samples were well bellow ML.

The highest sums of NDL-PCBs in wild game animals were determined in samples of pheasant from Šaľa region (111 ng·g^{-1} fat), deer from Košice region (76 ng·g^{-1} fat), boar and deer from Nemecká region (66 ng·g^{-1} and 48 ng·g^{-1} fat, respectively). Next, four samples from Krompachy, four samples from Nemecká, two samples from Košice, one from Šaľa and one from Starina had levels from 15 ng·g^{-1} to 30 ng·g^{-1} fat. Sums of NDL-PCBs in all other game samples were in the range 3.2–14 ng·g^{-1} fat, with the lowest values in deer from the background area Starina.

Predominant pollutants

Predominant congeners among chlorodibenzodioxins (CDD) and chlorodibenzofurans (CDF) were 2,3,4,7,8-pentaCDF, octaCDD, 1,2,3,7,8-pentaCDD, 1,2,3,6,7,8-hexaCDF and 1,2,3,4,6,7,8-heptaCDD and, in hen eggs and fish samples, also 2,3,7,8-tetraCDF. In Košice and Krompachy regions, also other PCDD/F congeners showed increased values, mainly in cows' and goats' milk samples, hen eggs, fish and partially in game samples. In general, all samples of ewes' milk from big farms and samples of bacon showed similarly low levels of all PCDD/F congeners. The most significant PCB congeners in all samples were PCB-118, PCB-156, PCB-105, PCB-153, PCB-126 (milk, game) and PCB-77 (pork, eggs, fish).

Comparison of PCDD/F and dl-PCB levels in foodstuffs from Slovakia and from other countries

Arithmetic means of PCDD/F-*TEQ*, DL-PCB-*TEQ* and total *TEQ* levels in foodstuffs from big farms, wild game and fish samples were compared with averaged data from monitoring activities in EU member states in 2000–2008, collected by European Food Safety Authority (EFSA) [24] and with results from Belgium [25] (Tab. 4). The most data in EFSA report were from Germany, United Kingdom, Sweden, Belgium, Ireland and Finland. The most contaminated game sample of deer from Košice area was excluded to avoid excessive overestimation of the average of the results.

Levels in milk from Slovakia were twice as high on than analogical samples from EU member states. Samples of game animals from Slovakia had POP levels from 3- to 5-fold higher than those

Tab. 4. Comparison of PCDDs/Fs, DL-PCBs and total toxic equivalent in foodstuffs from big farms in Slovakia with levels in EU member states and Belgium [24, 25].

Foodstuff	*TEQ* [pg·g^{-1}]											
	Slovakia (arithmetic mean)				EU (arithmetic mean)				Belgium (mixed samples)			
	n	PCDDs/Fs	DL-PCBs	Total *TEQ*	*n*	PCDDs/Fs	DL-PCBs	Total *TEQ*	*n*	PCDDs/Fs	DL-PCBs	Total *TEQ*
Milk and milk products[a]	30	1.6	1.8	3.4	931	0.78	1.00	1.78	19	0.72	1.13	1.99
Liver from terrestrial animals[b]	7	1.27	0.17	1.43	91	3.34	2.38	5.72	10	1.40	1.86	3.26
Pork fat[c]	6	0.10	0.10	0.20	89	0.92	0.95	1.87	–	–	–	–
Hen eggs	4	0.32	0.18	0.50	785	0.94	1.09	2.03	10	0.41	0.48	0.89
Meat of wild game animals	44	2.52	3.62	6.14	–	–	–	–	10	0.87	0.80	1.67
Muscle meat of fish[d]	37	0.32	1.16	1.48	1 976	1.89	2.08	3.98	14	0.23	0.74	0.97

TEQ – toxic equivalent (expressed per gram of fat), PCDDs – polychlorinated dibenzo-*p*-dioxins, PCDFs – polychlorinated dibenzofurans, DL-PCBs – dioxin-like polychlorinated biphenyls. a – Slovakia: cows' and ewes' milk, Belgium: only cows' milk; b – Slovakia: only pork liver, Belgium: mix of bovine, pork, rabbit and goose liver; c – Slovakia: lard and bacon; d – values are expressed per gram of wet weight.

presented by WINDAL et al. [25]. On the other hand, contamination of samples of pork origin and hen eggs was significantly lower than in other EU member states. Levels in fish samples were similar to those from Belgium.

CONCLUSIONS

The previously published data indicated that the most polluted region in Slovakia, from the ones selected for this study, was the Krompachy region [22, 26, 27]. In contrast to other regions, the very high POP levels in cows' milk from small family farms in Krompachy did not differ from big farms, since the animals grazed freely on the same meadows. This clearly illustrates the pollution of the local environment. In Krompachy region, the highest levels of PCDDs/Fs were observed, while food samples originating from Nemecká and Košice regions had significantly higher levels of PCBs. Higher levels of POPs were mainly observed in samples from sites adjacent to metallurgical plants and incinerators. The results from analysis of foodstuffs support the findings from previous examination of environmental samples and confirm the bad environmental status in industrial areas of Slovakia. Moreover, any food exceeding the limits set by European Commission should be excluded from the food chain, no matter if it originates from big farms or from small family farms.

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