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Selected organochlorine and organobromine pollutants in breast milk from Slovakia and infant daily intake

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Summary

Toxic organic pollutants including organochlorine pesticides (OCPs) are anthropogenic compounds accumulated in human tissues through the food chain. In this study, 63 samples of breast milk were collected from urban and rural areas of three regions (west, central and east), including eight districts of Slovakia, and analysed for specific pollutants, including seven polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyl (PCB-153), polybrominated biphenyl (PBB-153), hexachlorobenzene (HCB), pentachlorobenzene (PeCBz) and α-, β-, γ- isomers of hexachlorocyclohexane (HCH). Observed levels of sum of seven PBDEs were to be one of the lowest from those reported in worldwide studies with a median of 0.491 $ng\cdot g^{-1}$ of lipid weight (lw). This study also contains the first information on the presence of PBB-153 in breast milk in Slovak mothers with a median of 0.014 $ng\cdot g^{-1}$ lw. The highest levels of infant daily intake were determined for PCB-153 and HCB (median of 413 $ng\cdot kg^{-1}$ and 235 $ng\cdot kg^{-1}$ body weight per day, respectively). Slovakia still belongs to the countries with the highest HCB contamination. However, it is noteworthy that the concentration of HCHs, HCB and PeCBz in human milk decreased by more than a half in the period of 1993–2005.

Keywords

flame retardants; pesticides; biphenyls; breast milk

Polychlorinated biphenyls (PCBs), polybrominated biphenyls (PBBs) and polybrominated diphenyl ethers (PBDEs) are synthetic compounds also known as persistent organic pollutants (POPs). Because of their beneficial industrial properties, they have found a place in many daily used products. PBDEs and PBB-153 were used as flame retardants in plastics, textiles, insulating foams, electronics and other building materials, while PCBs (also PBB-153, but less) could be found as a dielectric fluid or oil in transformers, capacitors, hydraulics systems, etc. As most of the toxic organic pollutants, PCBs, PBDEs and PBBs can be also characterized by similar properties: chemical and biological persistence, toxicity, hydrophobicity and high bioaccumulation capacity [1, 2]. They are distributed into the lipid compartment of the human body mainly through the food from polluted environment. The industrial use of the technical mixtures of PCBs [3], penta- and octa-BDEs [4] and deca-BDE mixture [5] is forbidden, but high concentration of those pollutants had been positively identified in different biological matrices [6–9]. According to several studies, higher contents of PBDEs are related to many health issues such as attention [10, 11], endocrine and reproduction problems [12, 13]. Several studies indicated an increase of PBDEs content in biological matrices [14]. On the other hand, some authors reported an opposite or static trends [15, 16].

γ-Hexachlorocyclohexane (γ-HCH), also known as lindane, belongs to a group of organochlorine compounds. It was used as an insecticide in agriculture and pharmacy [17]. γ-HCH is classified as moderately hazardous and its trade is

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regulated and restricted under the Rotterdam Convention [18]. γ-HCH can persist in environment for a relatively long time and it occurs in countries where it had never been used due to occurrence in atmosphere and global transportation. As a consequence, it bioaccumulates in the food chain even though it can rapidly degrade after its use is discontinued [19]. Besides γ-HCH, of concern are also α- and β-HCH isomers, which are formed at its production and are even more toxic than γ-HCH [19]. In 2009, the production and agricultural use of HCH isomers was banned under Stockholm Convention on persistent organic pollutants [20]. Several studies indicated human health issues associated with HCH isomers [21].

Pentachlorobenzene (PeCBz) and hexachlorobenzene (HCB) are chlorinated aromatic hydrocarbons considered as POPs. PeCBz was used as a fire retardant, fungicide and dielectric fluid in mixtures with other chlorobenzenes and PCBs [22]. In 2006, United States Environmental Protection Agency (USEPA) and several authors reported deleterious effects of PeCBz on human health [23].

HCB is a fully chlorinated aromatic compound which, was also used in agriculture for seed treatment. Despite the production and usage of HCB is under Stockholm convention regulation, it is an unintended by-product from several processes, such as production of chlorinated solvents and municipal waste incineration. Several studies showed an association with many health issues [24, 25].

Since the milk contains appreciable amount of lipids, it is recommended and commonly used as a representative biological material for analysis and detection of organic pollutants with subsequent human health risk assessment. The aims of this study were to measure the content of PBDEs, PBB-153, PCB-153, PeCBz, HCB and α-, β-, γ- HCH in breast milk samples collected from mothers living in three different regions in Slovakia (west, central and east region) and to estimate daily infant intake of each group of pollutants. The intention was to obtain data that could provide a basis for follow-up cross-sectional epidemiological studies focused especially on the impact of brominated flame retardants on the health of the human population of Slovakia.

MATERIALS AND METHODS

Samples of human breast milk from 63 women, in the age from 16 to 36, were collected between 3rd–8th week after delivery from three regions of Slovakia in 2005–2006 to cover all area of the country. Almost all of the women involved in this study (98%) were having their first child. Information about their lifestyle and eating habits were collected from the questionnaires filled out by mothers after the milk sampling. First part of the questionnaire included basic information about participant, such as pre-, post- and present pregnancy weight, age, date of sampling, region. Second part of survey was focused on food and eating habits, such as consumption of certain food groups, which significantly contribute to dietary exposure to toxic organic pollutants, intake of cheese, eggs, poultry, fish and mammals and performed by five pre-defined answers ranging from "never" to "more than twice a week or twice a day". Additional information about origin and kind of meat or fish, and daily intake of milk, was collected. Due to the lack of samples in some districts, the attention was given to content comparison of target pollutants in three regions of Slovakia (west, central and east region). West region consisted of the following districts: Bratislava (n = 13), Trnava (n = 7), Trenčín (n = 6) and Nitra (n = 5) with together 31 samples. Central region of Slovakia was made up of two districts, namely, Banská Bystrica (n = 7) and Žilina (n = 7), 14 samples being collected. A number of 18 samples came from east region, specifically from districts Košice (n = 15) and Prešov (n = 3).

Extraction and clean-up

The known volumes of thawed and homogenized milk samples (50–100 ml) were taken for lipid extraction [26]. Isolated lipid fraction ranged between 1.2% and 6.6% with a mean of 3.2% and a median of 3.1%. An aliquot of lipids (0.2–0.3 g) were dissolved in n-hexane and spiked with $^{13}C_{12}$-labelled compounds: PBDE congeners (BDEs 28, 47, 99, 100, 153, 154, 183 and 209), organochlorine pesticides (OCPs: α-HCH, β-HCH, γ-HCH, HCB, PeCBz), PCB-153 (Cambridge Isotope Laboratories, Andover, Massachusetts, USA), and PBB-153 (Wellington Laboratories, Ontario, Canada). The lipids were transferred onto florisil-silica gel with H_2SO_4 column [26]. Analytes were eluted with 15 ml of 10% (v/v) dichloromethane in n-hexane. The residues were dissolved in 20 μl of the solution of $^{13}C_{12}$-labelled standards, consisting of BDE 79, 138 and 206 in n-nonane. The $^{13}C_{12}$-labelled mixture of PCB-32, 188 and $^{13}C_{12}$-labelled PCB-80 were used as recovery standards for analysis of OCPs and PCB-153.

Analysis of PBDEs and PBB-153

Seven PBDE congeners: 2,4,4'-triBDE (BDE-28, BDE-33), 2,2',4,4'-tetraBDE (BDE-47), 2,2',4,4',5-pentaBDE (BDE-99), 2,2',4,4',6-pentaBDE (BDE-100), 2,2',4,4',5,5'-hexaBDE (BDE-153), 2,2',4,4',5',6-hexaBDE (BDE-154), 2,2',3,4,4',5',6-heptaBDE (BDE-183), and one congener from polybrominated biphenyls, 2,2',4,4',5,5'-hexabromobiphenyl (PBB-153), were analysed by high resolution gas chromatography coupled to high resolution mass spectrometry in Trace GC Ultra DFS (Thermo Scientific, Bremen, Germany) operating at a mass resolution $R = 10000$ (10% valley definition). Chromatographic separation of the target compounds was performed using Rtx-1614 capillary column (30 m × 0.25 mm internal diameter, 0.1 μm film thickness; Restek, Bellefonte, Pennsylvania, USA). Helium was used as a carrier gas at a constant flow rate of 1 $ml \cdot min^{-1}$. Injected volume of sample extract was 2 μl using splitless surge mode (surge pressure 150 kPa for 2 min) with an injector temperature of 260 °C. The oven temperature was programmed from initial 120 °C held for 2 min, increased at 20 $°C \cdot min^{-1}$ to 230 °C and finally increased at 6 $°C \cdot min^{-1}$ to 325 °C and held for 13 min. Temperature of the ion source and transfer line were set at 260 °C and 280 °C, respectively. The electron impact ionization was used with electron energy of 45 eV. Perfluorokerosene (PFK; Cambridge Isotope Laboratories, Andover, Massachusetts, USA) or perfluorotributylamine (PFTBA; MasCom Technologies, Bremen, Germany) were used as reference compounds.

Analysis of PCB-153, HCB, PeCBz, α-HCH, β-HCH and γ-HCH

An HP 6890 Plus gas chromatograph (Hewlett-Packard, Palo Alto, California, USA) coupled to a high resolution mass spectrometer MAT 95XP (Thermo Finnigan, Bremen, Germany) operating at a mass resolution $R = 10000$ (10% valley definition) in the selected ion monitoring mode was used for PCB and OCP analysis. PCB congeners as well as all organochlorine pesticides were separated on a DB-5MS column (60 m × 0.25 mm internal diameter, 0.25 μm film thickness; J&W Scientific, Folsom, California, USA) according to Čonka et al. [27].

Tab. 1. Characteristics of the 60 participating mothers from three regions of Slovakia.

Characteristic of mother	Median	Range
Age [years]	25	16–36
Body mass index [$kg \cdot m^{-2}$]	20.8	16.6–31.9
First time pregnant	98 %	
Participant received breast feeding	91 %	
Residential area		
West region	49 %	
Central region	22 %	
East region	29 %	
Rural area	41 %	
Urban area	59 %	

Quality assurance and quality control

Qualitative and quantitative analyses were performed using isotope dilution method based on relative response factors (RRF) of individual compounds. To ensure high quality, blank samples were measured continuously with each batch of 10 samples. The limit of detection (*LOD*) was calculated as a triple value of noise.

The mean value of *LOD* for individual PBDE congeners 28, 47, 99, 100, 153, 154, 183 in nanograms per gram of lipid weight was: 0.001, 0.0008, 0.009, 0.01, 0.005, 0.005 and 0.01, respectively; for PBB-153 0.001 $ng \cdot g^{-1}$ lw, for PCB-153 0.002 $ng \cdot g^{-1}$ lw, for PeCBz and HCB 0.0003 $ng \cdot g^{-1}$ lw and 0.0004 $ng \cdot g^{-1}$ lw, for α-, β-, γ-HCH 0.01 $ng \cdot g^{-1}$ lw. Extraction efficiency of selected PBDEs varied from 28% to 109%, while recovery of PBB-153 and OCPs ranged between 40% and 62%. Expanded uncertainty of measurement (21%) was determined from measurements of internal reference material (pork fat from the market in Bratislava, Slovakia). Satisfactory repeatability and intermediate precision were achieved as, at analysing standard solutions, relative standard deviation (*RSD*) was below 14%. All measurements were carried out in an accredited laboratory (ISO/IEC 17025:2005), which previously successfully participated in European proficiency tests.

Data analysis

For statistical evaluation, IBM SPSS version 22 (IBM, Armonk, New York, USA) was used. The data were log-transformed prior to conducting statistical data analysis. Shapiro-Wilk test was used for testing normality. In the case of lower content than *LOD* value, *LOD*/2 was used for evaluation. Spearman rank and Mann-Whitney non-parametric test were used to calculate the correlations between the PCB-153, PBDEs, PBB-153, PeCBz, HCB, α-, β-, γ-HCH levels and age, urban and rural living mothers and body mass index (*BMI*). All *p*-values were two-tailed, and the level of $p < 0.05$ was considered statistically significant. To evaluate the significance of differences between west, central and east of Slovakia, ANOVA test was used.

Infant intake estimations

Infant daily intake (*DI*) of halogenated aromatic compounds expressed as nanograms per kilogram of body weight (bw) per day was calculated as follows:

$$DI = C \times V \times C_1 \qquad (1)$$

where C represents the content of certain compound (expressed as nanograms per gram lw), V represents the rate of breast milk consumption per body weight (in millilitres per kilogram bw per day) estimated on assumption of 600 ml per day and 5 kg of infant body weight, C_1 stands for lipid concentration in milk (in grams per millilitre) being specific for each sample.

RESULTS AND DISCUSSION

The general information about the breast milk donors is listed in Tab. 1. It is necessary to point out that even though this study included 63 milk samples, only 53 of them were used for *BMI* evaluation since seven volunteers did not provide information about their height. All donors of breast milk participating in this study were chosen under the request of living in current location at least for five years. Median, mean, minimum and maximum values of determined POPs are shown in Tab. 2.

PBDEs, PBBs and PCB-153 in breast milk

The most abundant PBDE congeners were BDE-47 and BDE-153 making up 41 % and 25 % (medians) of the sum of PBDEs, respectively. Dominancy of these congeners was also observed in a previous study from Slovakia [7], however, their contribution to the total PBDE content was different (31 % and 30 %, respetively). Using Spearman's non-parametric function, positive association between BDE-153 and PBB-153 ($r = 0.477$; $p < 0.001$) was identified, which is the first observation of this kind in Slovakia. The phenomenon might be explained by using products with addition of flame retardants consisting of higher BDEs in combination with PBB-153. Negative association was found between BDE-153 and *BMI* ($r = –0.6$, $p < 0.001$). Positive relations were also indentified among tri-, tetra-, penta-BDEs ($p < 0.001$) and among hexa- and hepta-BDEs ($p < 0.001$), which was most likely caused by usage of more (octa-BDE) or less (tetra-BDE, penta-BDE) brominated technical mixtures in different products. Negative relations between BDE-99, BDE-153 and age ($r = –0.25$ adn $r = –0.26$, respectively; $p < 0.05$) were revealed. This observation is consistent with the study of KANG, who also

Tab. 2. Estimated contents of persistent organic pollutants in three major regions of Slovakia.

Selected POPs [ng·g^{-1}]	West region ($n = 31$)			Central region ($n = 14$)			East region ($n = 18$)			Total ($n = 63$)	
	Median	Mean	Range	Median	Mean	Range	Median	Mean	Range	Median	Mean
ΣPBDEs	0.491	0.685	0.134–2.619	0.511	0.594	0.316–1.954	0.379	0.466	0.217–0.822	0.491	0.608
PBB-153	0.008	0.016	0.001–0.162	0.016	0.038	0.005–0.302	0.015	0.016	0.001–0.061	0.014	0.021
PCB-153	92	104	22–285	90	92	34–194	155	267	66–2096	109	148
HCB	52	69	23–423	67	109	38–527	69	87	29-259	62	83
PeCBz	0.210	0.245	0.116–0.743	0.186	0.376	0.125–2.675	0.214	0.218	0.110–0.318	0.208	0.266
α-HCH	0.180	0.205	0.079–0.835	0.199	0.214	0.055–0.478	0.084	0.102	0.007–0.295	0.172	0.177
β-HCH	16.7	18.0	10.9–34.1	14.8	14.4	7.4–24.4	15.3	16.1	4.0–40.5	15.6	16.7
γ-HCH	0.48	0.76	0.13–6.96	0.57	3.19	0.21–35.56	0.37	0.36	0.17–0.76	0.44	1.18

The values are expressed in nanograms per gram of lipid weight.
POPs – persistent organic pollutants, ΣPBDEs – sum of polybrominated diphenyl ethers, PBB-153 – 2,2',4,4',5,5'-hexabromobiphenyl, PCB-153 – 2,2',4,4',5,5'-hexachlorobiphenyl, HCB – hexachlorobenzene, PeCBz – pentachlorobenzene, HCH –hexachlorocyclohexane.

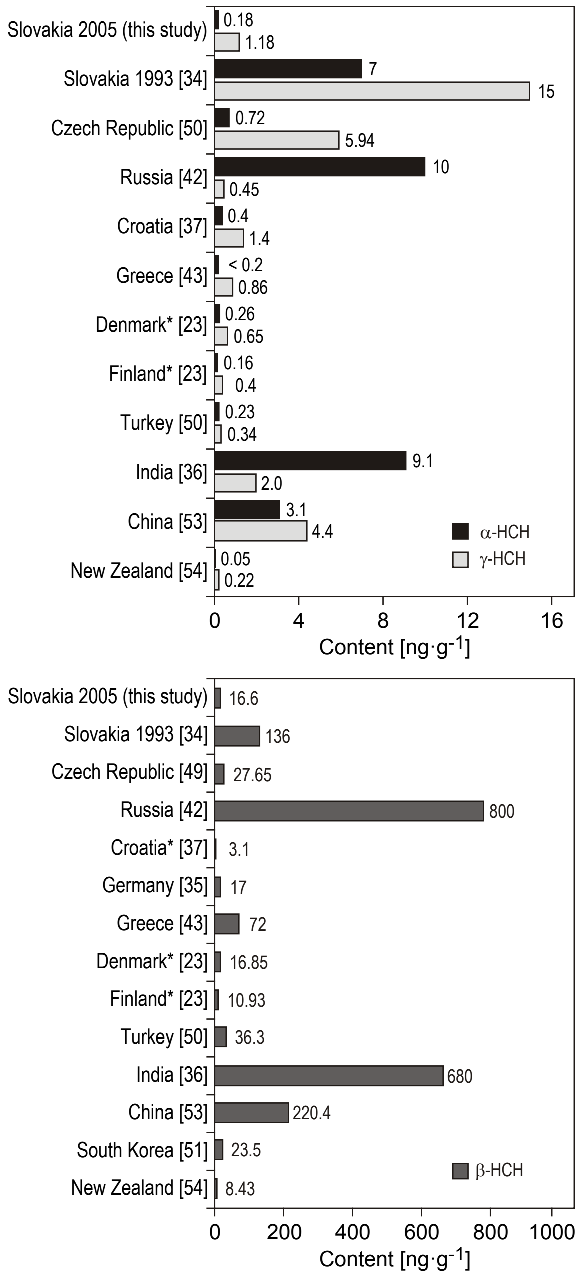

Fig. 1. Comparison of α-HCH, γ-HCH and β-HCH means with other countries.

Content is expressed per gram of lipid weight. * – median.

reported increased values of PBDEs in younger mothers [28].

As it is shown in many world studies [6, 28, 29] congener profile of most abundant PBDEs is not same for every country, what is most likely caused by usage of different technical mixtures [30].

Our study provides the first information on PBB content in the Slovak population in general. PBB-153 above *LOD* was found in 70% samples. Although this study investigated also other PBB congeners such as PBB-154, PBB-155, PBB-169, they were not statistically processed due to the fact that only 15 % of results were above *LOD*. The highest content of PBB-153 was expected, since it was the major congener of PBBs used in flame retardant mixtures [31]. Significant correlation was identified between age and content of PBB-153 ($p < 0.001$). The negative relation between age and flame retardants, such as PBB-153 and BDE-153, was probably due to the different lifestyle and activities of younger generations, which lead to higher exposure, although the pharmacokinetics of these chemicals needs to be considered [31].

Our study also includes results on PCB-153, which can be considered as bio-indicator since it is one of the most abundant congeners from indicator PCBs. The median content of PCB-153 in all samples was 109 $ng \cdot g^{-1}$ lw. The non-parametric Spearman's rank revealed the positive correlation between PCB-153 and age ($r = 0.425$, $p < 0.0001$).

Comparison of PBDE, PBB and PCB-153 levels in different regions of Slovakia

The data analysed by ANOVA showed significant regional differences between east and west regions ($p < 0.05$) regarding the most abundant congener BDE-47. As it can be seen in Tab. 2, the total PBDE contents did not differ so much among regions, while the levels of congener BDE-47 were significantly different ($p < 0.05$). The summed mean values were higher than medians in all regions, which was caused by several samples that had higher levels. The pattern of the most abundant congeners BDE-47, BDE-99, BDE-100 and BDE-153 was observed in each region with the contribution of 91 % to total content of seven measured congeners. These dominant congeners, as well as BDE-28, were detected in all 63 samples, while BDE-154 was found just in 65 % of breast milk samples. BDE-183 occurred in 92 % analysed samples. One sample of the breast milk from eastern Slovakia (Košice district) was not considered in statistical evaluation, because hexa- and hepta- PBDEs were 100–1 000 times higher than in the other samples. No relation between these high contents and information from questionnaire was found, so we can only assume that this mother could be exposed to PBDE mixture with dominance of higher congeners. The comparison of total PBDE median content in urban areas (0.491 $ng \cdot g^{-1}$ lw, $n = 37$) and in rural areas (0.490 $ng \cdot g^{-1}$ lw, $n = 26$) did not reveal a significant difference ($p > 0.05$).

PBB-153 was found to be on very low levels in all Slovak regions, which were 2 times higher in central and east region than in west region. Its content was below *LOD* in all samples taken in the Trenčín district (west). It was found slightly higher in urban areas (median content 0.0157 ng·g-1 lw) than in the nearby areas (median content 0.0107 ng·g1 lw).

Significant regional differences (east and west, east and central) in PCB-153 content were identified (ANOVA test, $p < 0.05$). The increased value of PCB-153 in the east region was most likely caused by the former production of PCBs (named DELORs) in Chemko Strážske chemical plant close to Michalovce, one of the sampling areas. In this environmentally contaminated part of Slovakia, increased levels of PCBs in blood, human milk and also sediment samples were previously determined [32, 33]. The other source of contamination could be the U.S. Steel factory in Košice or near the waste incinerator. The comparison of PCB-153 median contents in urban areas (111 ng·g-1 lw) and rural areas (89.1 ng·g-1 lw) revealed no significant difference.

Levels of HCH, HCB and PeCBz

Median content (all samples) of α-, β-, γ-HCH was 0.172 ng·g-1 lw, 15.6 ng·g-1 lw and 0.44 ng·g-1 lw, respectively. Isomers β- and γ-HCH were positively detected in all 63 samples, while α-HCH was below *LOD* in 13 % of samples. The ratio of total γ-HCH and β-HCH content was 0.03, while the earlier Slovak study reported ratio 0.25 [34]. The drop of γ-HCH/β-HCH ratio was expected, as the usage of lindane has been forbidden in Slovakia since 2004, and γ-HCH is easily transformed by human metabolism to β-HCH. Although the trend of the most abundant isomer β-HCH was consistent with many studies [35, 36, 37], the percentage profile of individual isomers was different.

We found significant association between α-HCH and γ-HCH ($r = 0.538$, $p < 0.0001$), and between β-HCH and both age and PCB-153 concentration in breast milk ($r = 0.423$, $p < 0.001$, and $r = 0.428$, $p < 0.001$). Similar results of positive relation between age and OCP levels were also reported in other studies [33, 37, 38].

Despite of the ban and restriction of HCB usage since 1970s in many countries, it is still formed as a by-product at a number of chemicals synthesis [39]. Even though the usage of this fungicide in Slovakia is forbidden since 1985, it is still present in the environment and in Slovak population. When we compare our results on α-, β-, γ-HCH and HCB with those in a previous Slovak study [34], we can observe a significant decline of these compounds in human milk in Slovakia (Fig. 1, Fig. 2). In all samples, HCB as well as PeCBz levels were above *LOD*. PeCBz was found to be present at a mean content of 0.266 ng.g-1 lw, which was 10 times lower than in 1992–1994 [34].

Comparison of HCH, HCB and PeCBz levels in different regions of Slovakia

As can be seen in Tab. 2, median content of HCB was found to be very similar in east and central regions, and slightly lower in west region. The highest HCB levels of 527 ng·g-1 lw and 423 ng.g-1 lw were found in two samples from Banská Bystrica (central region) and Trenčín (west region), which could have been caused by some local source of this pollutant. However, the data from questionnaires did not help to trace the origin of this pollution. There was a significant difference between HCB levels in urban and rural areas ($p < 0.032$).

In the case of PeCBz, the median content was highly similar in all three Slovak regions. Also, the median content of PeCBz in human milk in rural (0.205 ng·g-1 lw) and urban area (0.210 ng·g-1 lw) was comparable.

The data were again evaluated using ANOVA

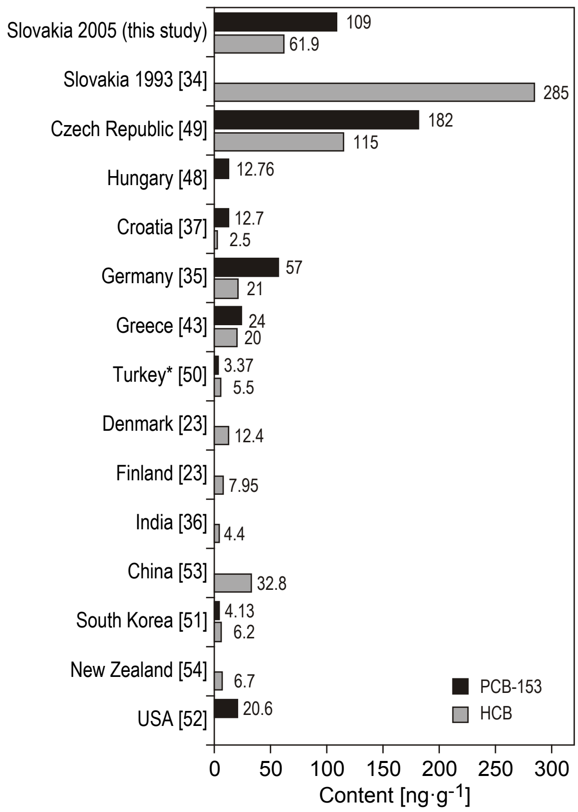

Fig. 2. Comparison of PCB-153 and HCB medians with other countries.

Content is expressed per gram of lipid weight. * – mean.

test to see, if there were any regional differences, mainly in γ-HCH content. The differences of γ-HCH and β-HCH contents were significant among all three regions ($p < 0.001$). Slightly higher contents of γ-HCH were found in west and central regions. The reason might be that west and central regions are still important agricultural areas of Slovakia, where this compound was used as pesticide until 1994. β-HCH was the most abundant isomer in all breast milk samples, except for one sample from Žilina with the highest content of γ-HCH (35.56 $ng\cdot g^{-1}$ lw). Since γ-HCH is, after entering metabolism, rapidly transformed to β-HCH, it could be assumed that this mother was exposed to a direct source of γ-HCH at a couple of weeks before sampling. Given information in the questionnaire did not reveal any connection between high content of γ-HCH and dietary habits. The ratio of γ-HCH and β-HCH in this milk sample was 1.46. We found a significant difference between β-HCH levels in urban and rural areas ($p < 0.038$).

Estimation of infant daily intake

Although human milk contains all necessary nutrititive compounds for infants, numerous studies all over the world demonstrated also increased contents of anthropogenic compounds including PCBs, PBDEs, PBB-153 and OCPs in it. Since all these compounds are highly bioaccumulative and breast milk is the only source of food in first weeks, infants may be exposed to various kinds of organic pollutants. Based on this fact, the primary source of infant exposure to halogenated pollutants is breast milk [40]. In a previous Slovak study, no significant difference between levels of POPs and breast milk sampling time after delivery was found. Colostrum and mature milk lipid-based concentrations of PCBs and OCPs were highly correlated ($p < 0.001$) despite the lower lipid content in colostrum. Lipid-based concentrations did not correlate with the percentage of lipids. The average decline in PCB and OCP levels was 2–11 % [41].

We estimated that infant exposure in Slovakia in 2005 through breast milk was 1.8 $ng\cdot kg^{-1}$ bw per day (median value) with mean 2.4 $ng\cdot kg^{-1}$ bw per day for the sum of PBDEs, assuming individual percentage of lipids in milk, 600 ml per day consumption and 5 kg of body weight according to Stasinska et al. [8]. Our results shown in Tab. 3 are very similar to the most recent Slovak study [7] that reported the mean value of daily infant intake of total PBDEs in Košice ($n = 8$) to be 3.0 $ng\cdot kg^{-1}$ bw per day, while in this study the estimated mean value for the same district ($n = 15$) was 2.0 $ng\cdot kg^{-1}$ bw per day. In current study, the median daily exposure to BDE-47, BDE-99 and BDE-153 was 0.73 $ng\cdot kg^{-1}$ bw per day, 0.15 $ng\cdot kg^{-1}$ bw per day and 0.51 $ng\cdot kg^{-1}$ bw per day, respectively. Furthermore, this study presents first results concerning the daily intake of PBB-153 with median value of 0.05 $ng\cdot kg^{-1}$ bw per day. Reported estimated *DI* median values of β-HCH, γ-HCH, HCB, PeCBz are listed in Tab. 3.

Comparison of obtained results with Europe and worldwide

It was hard to compare total content of PBDEs with other studies, because of the wide range of measured congeners. Nevertheless, when the look

Tab. 3. Estimated daily infant intakes and its medians and ranges of POPs in all samples and in three major regions of Slovakia.

Selected POPs [$ng\cdot kg^{-1}$]	West region ($n = 31$)		Central region ($n = 14$)		East region ($n = 18$)		Total ($n = 63$)
	Median	Range	Median	Range	Median	Range	Median
ΣPBDEs	1.78	0.18–10.17	1.83	0.89–15.50	1.72	0.45–3.46	1.77
PBB-153	0.031	0.001–0.788	0.057	0.023–1.227	0.060	0.002–0.211	0.051
PCB-153	321	58–1 467	321	115–870	648	285–9 938	413
HCB	185	62–1 315	254	117–4 182	306	90–1 064	235
PeCBz	0.763	0.261–2.961	0.630	0.357–9.181	0.902	0.441–1.780	0.788
α-HCH	0.648	0.297–2.485	0.678	0.214–2.077	0.355	0.042–0.936	0.633
β-HCH	60.2	15.9–135.2	52.2	34.6–83.7	65.2	14.2–208.5	58.7
γ-HCH	1.8	0.8–25.4	1.9	0.9–122.1	1.6	0.7–2.4	1.7

The values are expressed in nanograms per kilogram body weight per day.
POPs – persistent organic pollutants, HCB – hexachlorobenzene, PeCBz – pentachlorobenzene, HCH – hexachlorocyclohexane, ΣPBDEs – sum of polybrominated diphenyl ethers, PBB-153 – 2,2’,4,4’,5,5’-hexabromobiphenyl, PCB-153 – 2,2’,4,4’,5,5’-hexachlorobiphenyl.

is taken on data of the sum of seven major PBDEs (28, 47, 99, 100, 153, 154 and 183) in breast milk described in last ten years all over the world, Slovak PBDE breast milk levels are significantly lower (Fig. 3). The contribution of BDE-47, 99, 100 and 153 is 90 % to the sum of 7 congeners. These congeners were analysed in all studies used for comparison. In general, as we can see from the available published median content in other countries, human exposure to PBDEs is significantly lower in Europe than in China or Australia, while it seems to be the highest in Canada and USA [6, 9, 29, 30, 35, 42–46].

When data on individual PBDEs are evaluated, BDE-47 is usually the highest-abundant congener in human tissues. This finding is comparable with our study. This is probably caused by exposure to commercial mixtures where BDE-47 is one of the major components [42]. Several studies published predominance of BDE-153, which can be caused by exposure to higher brominated commercial mixtures [29, 35].

At present, only a few studies are available on the content of PBB-153 in breast milk. The median value of PBB-153 in our study (0.014 $ng\cdot g^{-1}$ lw) was approx. 2–6 times lower than the data reported by Yang et al. [47] in Chinese breast milk and Bramwell et al. [44] in United Kingdom (0.024 $ng\cdot g^{-1}$ lw and 0.08 $ng\cdot g^{-1}$ lw, respectively). Our mean value (0.021 $ng\cdot g^{-1}$ lw) was 6–10 times lower than that for Finnish and Danish breast milk (0.134 $ng\cdot g^{-1}$ lw and 0.200 $ng\cdot g^{-1}$ lw) [23].

As shown in Fig. 2, the median content of PCB-153 in all samples from our study was much higher than that in some other countries, except for Czech Republic [35, 37, 43, 48–52,]. Mikeš et al. [49] reported nearly twice as high median as our study, determined during 15 years of Czech monitoring campaigns, analysing 4 753 breast milk samples since 1994. Levels of toxic pollutants in Slovakia and Czech Republic can be comparable because of a very similar exposure history and former production of PCBs in Czechoslovakia. Levels of α-, β-, γ-HCH determined in this study are slightly lower than in Czech monitoring [49]. In general, α-, β-, γ-HCH levels in Slovakia (this study) are comparable with other European countries and much lower than in Russia, India or China, while higher than in New Zeland [23, 35–37, 42, 43, 50, 51, 53, 54] (Fig. 1).

The HCB median level determined in this study (61.9 $ng\cdot g^{-1}$ lw) is much higher than those reported in last 10 years in studies from some other countries [23, 36, 37, 50, 51, 54] and 2–3 times higher than in Germany, Greece or China [35, 43, 53] (Fig. 2). This median level is about a half

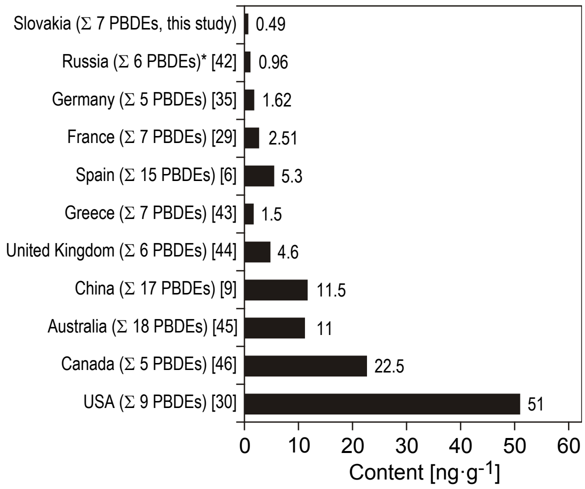

Fig. 3. Comparison of PBDE medians with other countries.

Content is expressed per gram of lipid weight. * – mean.

of those reported by Mikeš et al. [49] for Czech Republic in 1994–2009, but it decreased over the years in Slovak population approx. 4.6 times since 1993 [34].

The present median content of PeCBz (0.21 $ng.g^{-1}$ lw) determined in this study is comparable to those found in Finland, Denmark or Turkey, but lower than in New Zealand (0.25 $ng\cdot g^{-1}$ lw, 0.32 $ng\cdot g^{-1}$ lw, 0.31 $ng\cdot g^{-1}$ lw and 0.56 $ng\cdot g^{-1}$ lw, respectively) [23, 50, 54].

For breast-fed infants with average human milk consumption, the mean daily exposure to BDE-47, BDE-99 and BDE-153 in 24 European countries was 0.64–13.8 $ng\cdot kg^{-1}$ bw, < 0.14–5.05 $ng\cdot kg^{-1}$ bw and 0.46–11.03 $ng\cdot kg^{-1}$ bw per day, respectively [55]. Higher values of *DI* for BDE-47, BDE-99, BDE-153 were reported in United Kingdom [44], assuming average daily consumption of 800 ml and a different body weight than in our study. Even considering the same conditions, daily exposure levels of three most abundant PBDE congeners would be still lower in the Slovak population.

The total *DI* (median) of HCB determined in this study was similar to China (210 $ng\cdot kg^{-1}$ bw per day [53]), but 2–3 times higher than in Croatia or South Korea (100 $ng\cdot kg^{-1}$ bw and 81.5 $ng\cdot kg^{-1}$ bw per day [37, 51]) and much higher than in Turkey (30 $ng\cdot kg^{-1}$ bw per day [50]).

CONCLUSIONS

The presented results regard concentrations of seven PBDE congeners, PBB-153, PCB-153, HCB, PeCBz and three isomers of HCH in human milk as well as estimation of daily infant intake of these compounds. Although this study was limited by the number of participants, it describes the level of contamination in Slovakia as well as its specific regions by individual organohalogen pollutants and also compares their levels with other countries. The data from this study show several significant associations mainly between PBB-153, BDE-153 and *BMI* and also between β-HCH, PCB-153, BDE-153 and age. The major added value of this study is determination of PBB-153 levels and the estimation of its daily infant intake, because there has been no study investigating this group of pollutants in Slovak breast milk. These results also contribute to epidemiological studies, which are still trying to examine relation between PBDEs, PBBs and negative health issues.

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