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Original Research

Soil Contamination by PCBs on a Regional Scale: the Case of Strážske, Slovakia

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Abstract

Trace amounts of polychlorinated biphenyls (PCBs) are ubiquitous in the environment. Because of industrial activity, other human activities, and accidents, higher concentrations of these chemicals may be present in soil in residential and recreational areas. Human uptake of these chemicals from such soil has been assumed by regulators, and people contacting such soil may be concerned about potential adverse health effects. For these reasons, the area around a former chemical manufacturing site (Stražske district – east Slovakia) is still important for monitoring PCBs in the environment.

The present study aimed to characterize the PCB contents in two soil (gl FL, st ct LV) types near sources of contamination (precint of former chemical factory Chemko Strážske, waste dump of former chemical factory, sewage channel of former production). PCB content was measured at 22 parcels of land (spring and summer term of sampling) in 2011.

The average value of sum of monitored PCBs (#52, 101, 118, 138, 153, 180) ranged from 4.7 $ng \cdot g^{-1}$ to 91 $ng \cdot g^{-1}$ and at five sites exceeded the limit of 50 $ng \cdot g^{-1}$ valid in Slovak Republic. PCB content in soil was not affected by sampling date (insignificant effect). Significant difference between PCB contents was found depending on soil type. Average content in Luvisols was by 17±2.2 $ng \cdot g^{-1}$ and in fluvisols by 33±2.4 $ng \cdot g^{-1}$ of PCBs. The correlation between the sum of PCBs and humus was significant only at gl FL ($r = 0.64$, $p < 0.05$).

The results demonstrate that PCB substances persist in the soil after nearly 30 years of cessation of production and therefore it is necessary to solve this problem in Slovakia.

Keywords: PCBs, soil type, humus, Slovakia

Introduction

One of the areas of PCB production was also in East Slovakia, in Strážske district. The location is considered as one of the most contaminated territories in Central Europe, content of PCBs was detected in all the environment [1-5].

It is assumed that a source of pollution is about 2,750 tons of products based on PCBs, without any other relevant information. The producer was the chemical company Chemko Strážske [5]. Probably part of this amount was released into the environment, mainly in the surroundings of the above-mentioned former PCB manufacturer (improper disposal, improper storage, in the form of production waste ...). Sediment contamination in the sewer of a chemical factory, the Laborec River, and Zemplínska Šírava water tank contamination in many cases exceeds 1,000 times the limit allowed in Slovak Republic for this matrix [5].

As for PCB content in the soil, it was found that PCBs may be partially degraded [6, 7]. Some of them may be

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transfered to plants, depending on the plant species [6, 8, 9]. Contaminated soils are not only a source of contamination for the environs, but in the form of vapour or adsorbed in solid particles PCBs are transposed over long distances [1].

The behavior of PCBs in the soil is strongly influenced by basic soil properties [10]. Sorption strength of the soil decreases with the length of the binding of contaminants in soil and greatly depends on the character, quality, and stock of organic matter. While leaching of PCBs is the most anticipated in soils with low humus content [11, 12].

PCB content is also influenced by agricultural activities, such as soil tillage [13], application of sludge, or compost [14].

Great attention has been devoted to the problem of soil contamination by PCBs in the United Kingdom, USA, Canada, Japan, and Sweden. Monitoring of PCBs was carried out mostly at the end of the last century, especially in contaminated areas. As a result of monitoring, it was found that soil contamination is high. Total PCB content found in England was 400 $ng \cdot g^{-1}$ [15], 567 $ng \cdot g^{-1}$ in the USA [16], 200 $ng \cdot g^{-1}$ in Canada [17], 1000 $ng \cdot g^{-1}$ in Japan, and 332 $ng \cdot g^{-1}$ in Sweden [18].

Significantly lower levels of PCBs in soil were detected in the Moscow area (up to 42 $ng \cdot g^{-1}$) [19]. Only at the level of microgram per kilogram units or less did soils in Turkey vary [20]. In Italy, in the region of Piedmont, Fabietti et al. 2010 [21] set out a summary content of PCBs in soil used for agriculture in the range of 0.1 $ng \cdot g^{-1}$ to 16 $ng \cdot g^{-1}$, with an average value of 2.2 $ng \cdot g^{-1}$.

The content of PCBs in soil has been previously examined in Slovakia as well. Pollution levels varied, with a maximum value of 300 $ng \cdot g^{-1}$. Contamination of soil showed only local character [1, 2, 22].

The process of liquidation of persistent organic pollutants, such as PCBs, is very difficult and time-consuming [23]. The Slovak Republic ratified the Stockholm Convention on Persistent Organic Pollutants in 2002, and has committed to gradually eliminating them.

Preparatory work to begin disposal of PCBs from the most polluted areas of the Strážske district, Slovakia, is currently being carried out.

The aim of this work is to evaluate the content of PCBs in soil used for agriculture, which is adjacent to the main sources of contamination (precint of former chemical factory Chemko Strážske, sewage channel of former production, dump). This study is focused on the evaluation of current results in the territory to which insufficient attention is adressed despite extreme pollution [24].

Materials and Methods

Study Area and Sampling

The study was conducted at plots situated near Strážske. In the past, there has been a producer of PCBs, the chemical company Chemko Strážske. Negative impacts on the environment in this area persist even 30 years after the cessation of PCB manufacturing. The sampling sites are located in the eastern part of Slovakia (Fig. 1).

The experiment was conducted on two soil types (Stagnic Cutanic Luvisol, sign.: st ct LV, Gleyic Fluvisol, sign.: gl FL), which are the most common in the area.

The state of pollution of land resources has been studied on 22 plots used for agriculture (in total 800 hectares of agricultural land). Basic characteristics of sampling plots

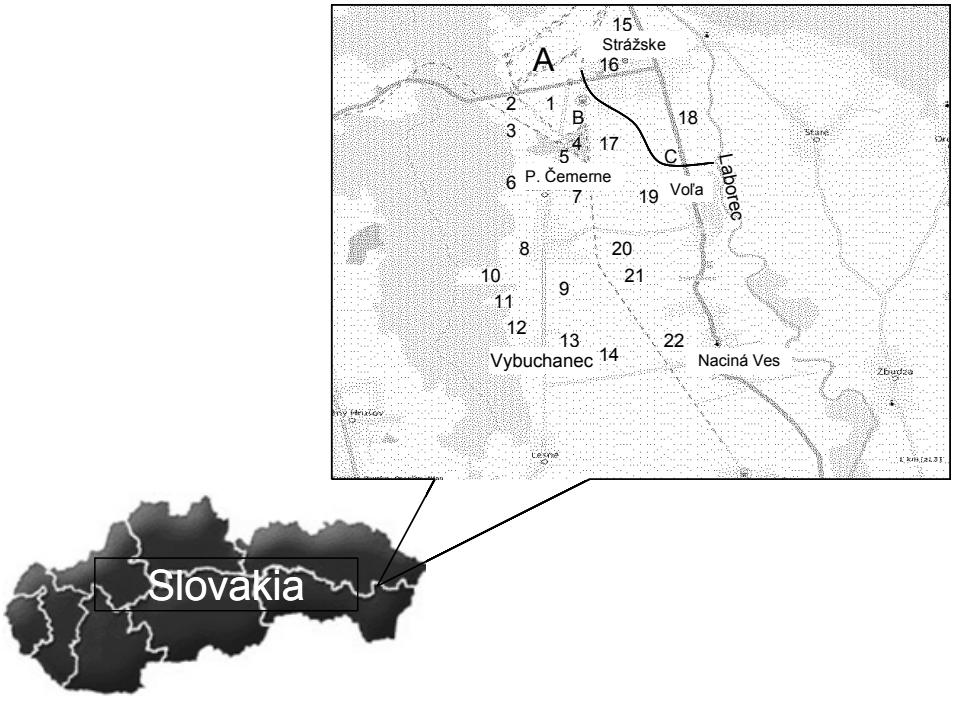

Fig. 1. Detailed map of monitored area with marked sampling plots (A – Chemko Strážske, site of former chemical factory; B – waste dump, C – Strážske channel, sewage channel of former production).

Table 1. Basic characteristics of sampling plots with GPS location and main physical-chemical properties of topsoil.

Sampling sites	Area [ha]	Soil type	GPS location		pH/KCl	Content [%]			
			Latitude	Longitude		Humus	Clay	Silt	Sand
1. Strážske 1	33.9	st ct LV	48º52.170’	021º48.989’	5.1	1.8	11	64	25
2. Strážske 2	25.1	st ct LV	48º52.091’	021º48.485’	7.0	2.0	11	72	17
3. Strážske 3	7.4	st ct LV	48º51.867’	021º48.383’	5.4	2.2	12	71	17
4. Pláne 1	8.5	gl FL	48º51.535’	021º49.345’	6.4	4.8	16	65	19
5. Pláne 2	17.1	gl FL	48º51.458’	021º49.258’	6.8	2.8	18	62	20
6. Pusté Čemerné 1	19.2	st ct LV	48º50.980’	021º48.425’	5.6	1.8	11	55	34
7. Pusté Čemerné 2	6.2	gl FL	48º50.896’	021º49.102’	3.8	1.9	13	73	14
8. Pusté Čemerné 3	20.4	st ct LV	48º50.069’	021º48.646’	6.7	1.3	13	48	39
9. Pusté Čemerné 4	34.8	st ct LV	48º49.761’	021º48.988’	5.7	1.8	12	64	24
10. Vybuchanec 1	10.0	st ct LV	48º49.752’	021º48.531’	4.0	2.3	16	68	16
11. Vybuchanec 2	10.7	st ct LV	48º49.637’	021º48.428’	4.3	1.9	12	72	16
12. Vybuchanec 3	32.8	st ct LV	48º49.364’	021º48.551’	4.4	1.8	14	67	19
13. Vybuchanec 4	84.9	st ct LV	48º49.138’	021º49.091’	3.7	2.5	20	69	11
14. Naciná Ves 1	69.5	gl FL	48º49.007’	021º50.032’	4.7	2.3	26	57	17
15. Strážske 4	49.1	glL FL	48º53.079’	021º50.069’	6.2	2.9	22	61	17
16. Strážske 5	9.1	glL FL	48º52.506’	021º49.668’	6.5	3.2	25	60	15
17. Voľa 1	138.1	gl FL	48º51.127’	021º50.722’	5.3	2.4	21	61	18
18. Voľa 2	47.8	gl FL	48º51.766’	021º50.719’	6.0	2.0	19	62	19
19. Voľa 3	111.8	gl FL	48º51.001’	021º50.868’	5.3	2.6	20	52	28
20. Voľa 4	27.3	gl FL	48º50.542’	021º50.030’	4.3	2.8	21	66	13
21. Voľa 5	28.6	gl FL	48º50.348’	021º49.975’	4.4	2.3	20	56	24
22. Naciná Ves 2	7.6	gl FL	48º49.142’	021º50.891’	5.1	1.8	19	52	29

st ct LV – Stagnic Cutanic Luvisol
gl FL – Gleyic Fluvisol

(acreage, soil type and GPS location are shown in Table 1. Sampling sites are located close to the hypothetic sources of pollution (Chemko Strážske, waste dump of former chemical factory, sewage channel of former production) and south of mentioned sources, which relates to the frequent wind flow in north-south direction.

Soil samples were collected twice a year from a depth of 0 to 0.3 meters (first collection in March, second collection in July). Collection of soil material was carried out in accordance with standards applicable to the Slovak Republic [25].

Chemical Analysis

The extraction of analyzed congeners (#52, 101, 118, 138, 153, 180) was performed in a Soxhlet apparatus using a mixture of n-hexane and acetone (v/v 1:1). Cleanig procedure consists of two steps. First, extract was cleansed with concentrated sulfuric acid and second with florisil. To obtain a sample for gas chromatography the eluate was evaporated under a stream of nitrogen and dissolved in n-hexane.

Analytical separation, identification, and quantification of PCB extracts was performed on a gas chromatograph HP 5890 Series II (Hewlett-Packard, USA) with ECD detector and automatic injector HP 7673. An HP-5 column (5% phenyl-methyl-polysiloxane, 50 m×0.2 mm×0.33 μm) was used for separation.

Nitrogen was used as the carrier gas at a constant flow rate of 17.3 $cm \cdot s^{-1}$ and also as the dilution gas at a flow rate of 60 $ml \cdot l^{-1}$. The injector temperature was kept at 290ºC and detector temperature at 320ºC. Temperature program: 80ºC for 1 min, increase rate 30ºC $min.^{-1}$ to 180ºC, 180ºC for 0 min, increase rate 3ºC $min.^{-1}$ to 205ºC, 205ºC for 4 min, increase rate 20ºC $min.^{-1}$ to 290ºC, 290ºC for 20 min.

The qualitative analysis is based on comparison of retention times of individual congeners of PCBs. During the measurement, the retention time of the peaks are com-

Table 2. Concentration of PCBs [ng·g^{-1}] in soil.

Sampling sites	Mean	S.D.	Range		Median
			min.	max.	
1. Strážske 1	57	26.6	16	90	51
2. Strážske 2	46	37.0	15	100	26
3. Strážske 3	6.9	2.8	3.2	11	7.0
4. Pláne 1	91	31.6	45	127	90
5. Pláne 2	58	10.3	37	70	59
6. Pusté Čemerné 1	17	13.4	9.6	44	12
7. Pusté Čemerné 2	52	11.4	37	70	50
8. Pusté Čemerné 3	22	7.9	13	34	19
9. Pusté Čemerné 4	19	7.3	12	32	16
10. Vybuchanec 1	11	3.7	6.0	17	10
11. Vybuchanec 2	7.5	1.0	6.6	9.3	7.2
12. Vybuchanec 3	7.2	2.6	3.7	10.8	6.7
13. Vybuchanec 4	7.2	2.3	2.7	10.5	7.6
14. Naciná Ves 1	16	6.4	8.7	32	15
15. Strážske 4	15	5.7	7.2	24	13
16. Strážske 5	32	20.8	8.9	57	30
17. Voľa 1	20	6.0	8.4	28	20
18. Voľa 2	26	13.5	15	57	21
19. Voľa 3	37	9.9	19	52	37
20. Voľa 4	16	5.8	5.0	23	17
21. Voľa 5	4.7	2.4	1.6	8.2	3.9
22. Naciná Ves 2	33	7.5	24	41	34

pared with the retention time of standards in the GC-ECD record. Quantification was performed using the external standard technique. The calibration curve was obtained by measuring five concentration levels of calibration solutions, while detector response was measured three times at one level. Calibration solutions were prepared from a stock solution, which was prepared after the dilution of the standard solution (certified reference standard of PCBs and mixture of PCBs congeners) in n-hexane. Concentration of the standard solution was 10 ng·µl^{-1} (standard deviation ± 0.5%). Measurement specifications were identical for the calibration solutions and samples. The instrumental limit of detection and limit of quantification were expressed as a concentration of specifield PCBs (ng·ml^{-1}) at a signal-to-noise ratio of 3:1 or 10:1 [26]. Method detection limit and the method quantification limit were estimated from limit of detection or limit of quantification, respectively. This means that the limit of detection or limit of quantification value was multiplied by the final volume of cleaned-up extract and divided by the soil sample weight and the volume of the extract injected on the GC column [27]. The method detection limit for the analyzed compounds ranged from 0.03 ng·g^{-1} (for congener #153) to 0.06 ng·g^{-1} (for congener #101), and the method quantification limit ranged from 0.19 ng·g^{-1} (for congener #138 and 180) to 0.29 ng·g^{-1} (for congener #101).

The soil samples were also analyzed to determine soil reaction and humus content. The chemical soil parameters were analyzed using well-known methodologies, exchangeable soil reaction in KCl solution, soil microstructure, and carbon content by Tyurin [25].

Statistical Analysis

Multifactor analysis of variance (Anova) with the least significant difference procedure (LSD) and correlation analysis were performed for evalution of the data (Statgraphics software package).

Results and Discussion

The content of polychlorinated biphenyls has been studied in a territory ideal for agricultural use, because of the territory altitude (117-160 m), climatic conditions (rainfall conditions, air temperature), and soil conditions (st ct LV, gl FL). General properties of topsoil by sampling sites are reported in Table 1. Agricultural soils have a mean pH of 5.3, ranging 3.7-7.0 and mean humus content of 2.3%, ranging 1.3-4.8%. It was also found that the mean content of clay was 16.9%, content of silt 62.6%, and mean content of sand 20.5%.

The land is used for above-mentioned agricultural purposes, but the measured contents of PCBs in soil (Table 2) indicate that the soil is contaminated with PCBs (Fig. 2 shows concentration of PCBs by both sampling dates and soil types, with displayed line connecting the extreme values). The average value of the sum of monitored PCBs (#52, 101, 118, 138, 153, 180) actually exceeded the limit of 50 $ng \cdot g^{-1}$ at five sites. Concentration limits and range of congeners is described by the Decree of the Slovak Republic [28]. The highest concentration has been found in sample 4 (Pláne 1) 91±31.6 $ng \cdot g^{-1}$. The limit value also was exceeded in sample 1 (Strážske 1) 57±26.6 $ng \cdot g^{-1}$, sample 2 (Strážske 2) 46±37.0 $ng \cdot g^{-1}$, sample 5 (Pláne 2) 58±10.3 $ng \cdot g^{-1}$, and sample 7 (Pusté Čemerné 2) 52±11.4 $ng \cdot g^{-1}$. The above-mentioned contaminated soil is located in close proximity to the former chemical factory (Chemko Strážske) and waste dump. Despite the significant pollution of soil, in our opionion not enough attention to the location was paid and is not paid in the present [24]. Domotorova et al. [24] showed five exceeding values found in five locations and 32 sampling sites. As noted above, we found five exceeding values in one location which confirms our view that the site is in therm of PCBs content in the soil much more significant than the rest of Slovakia.

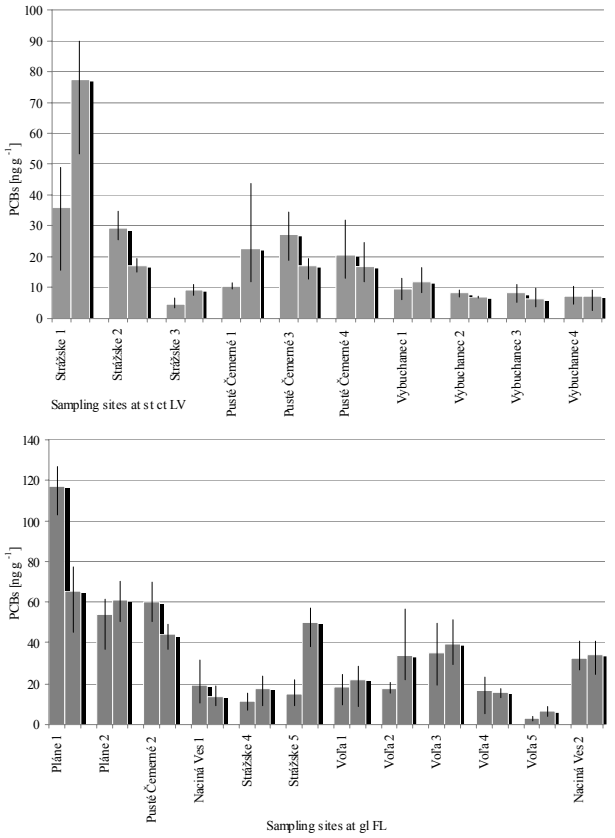

Fig. 2. Average PCB content by both sampling dates and sites at Stagnic Cultict Luvisols and Gleyic Fluvisols.

The level of contamination is below the maximum levels established in the UK, Canada, USA, Japan, and Sweden [15-18]. PCB content measured in the samples ranges from 1.6 $ng \cdot g^{-1}$ to 127 $ng \cdot g^{-1}$. Measured concentrations were on the same level as published by Zhang [29] for the region of Yangtze in China (0.5 to 74 $ng \cdot g^{-1}$).

PCB content in soil significantly depends on the sampling site (ANOVA $F = 41.44$, $P < 0.0001$). There was a significantly higher content of PCBs in the topsoil layer at sampling point Pláne 1 than in the other 21 plots (LSD: $p < 0,05$). The soil from sites with exceeded limit content had the same level of contamination (5.Pláne 2, 1.Strážske 1, 7.Pusté Čemerné 2). It was also found that the sites with the lowest pollution belong to one homogeneous group (LSD: $p < 0.05$). The sites with the least pollution were: 21. Voľa 5 (4.7±2.4 $ng \cdot g^{-1}$), 3. Strážske 3 (6.9±2.8 $ng \cdot g^{-1}$), 13. Vybuchanec 4 (7.2±2.3 $ng \cdot g^{-1}$), 12. Vybuchanec 3 (7.2±2.6 $ng \cdot g^{-1}$), 11. Vybuchanec 2 (7.5±1.0 $ng \cdot g^{-1}$), and 10. Vybuchanec 1 (11±3.7 $ng \cdot g^{-1}$). Listed sampling points, except location Stražske 3, belong to the locations situated far from the pollution sources. The gradual decline of PCBs in soil at Strážske 3 was probably caused by long-term cultivation of clover-grass mixture and alfalfa. Alfalfa is characterized by the phytoextraction effect for PCBs [30, 31].

It was further found that the impact of sampling date (spring, summer) on the content of PCBs in the soil was not significant. The average PCB levels in soil were slightly higher in summer sampling (27 $ng \cdot g^{-1}$) than in the spring sampling (26 $ng \cdot g^{-1}$), which may be related to soil temperature. The positive correlation between the seasonal increase in soil temperature and PCB concentrations was found by Tasdemir et al. [32]. Tasdemir also found that the correlation between the air and soil PCB concentrations is significant and soil PCB levels that are not correlated with seasonal temporal changes, suggested by inputs from local sources agreeing with the findings of the air levels measured at the site. The dominant contribution of local sources may affect the correlation between the air and soil concentrations of PCBs.

PCB content in soil is influenced by the content of humic acids [33-35]. The humus content and its quality are the important characteristics of monitored soil types (st ct LV and gl FL). Luvisols contain less humus, and its quality is worse in comparison with humus in fluvisols [36]. This fact was also confirmed by our measurements. The average content of humus in the st ct LV was 1.9% with an average carbon content of humic acids 0.2%, while the gl FL humus content was 2.7% with an average 0.2% carbon content of humic acids. Probably in connection with the above-mentioned facts, the effect of soil type on PCB content was significant (ANOVA $F=22.98$, $P<0.0001$). It was found that the average sum of PCBs on soil type st ct LV was 17±2.2 $ng \cdot g^{-1}$ and on soil type gl FL was 33±2.4 $ng \cdot g^{-1}$.

The effect of select soil parameters on PCB content has been observed by several authors [13, 18, 37, 38]. They found that higher organic carbon content correlates with enhanced levels of PCBs. Statistically significant correlation between sum of PCBs and soil organic content ($r = 0.64$, $p < 0.05$) also was found at our study for fluvisols. Correlation may be in some cases statistically insignificant [39, 40]. It was partially explained by the extremely low levels of organic carbon in the soil samples, varying between 0.1 and 0.3% [40]. In our study, we did not find a significant correlation between the organic carbon content and PCB contamination in the soils of ($r = - 0.33$, $p < 0.05$). It may also be explained by lower levels of organic carbon varing between 1.3 and 2.5% in comparison with organic carbon in fluvisols 1.8-4.8%.

Presented and evaluated PCB levels in soil are summary values of six PCB congeners (Table 3). The lowest average levels (ranged from not detected levels to 8.8 $ng \cdot g^{-1}$) at all monitoring sites were quantified for tetrachlorobiphenyls No. 52 (1.5±1.5 $ng \cdot g^{-1}$), for low-chlorinated biphenyl. Conversely, the highest average concentration (6.9±7.1 $ng \cdot g^{-1}$) and the highest content range (undetected until 46 $ng \cdot g^{-1}$) was found for hexachlorbiphenyl No. 153, high-chlorinated biphenyl. The dominant congeners in soil were also congener Nos. 118, 138, and 180. Generally, four dominant congeners (Nos. 118, 138, 153, and 180) represent up to 85.1% of the total content. The same conclusions for the evaluation of the content of each congener also reflect the results obtained for different soil types.

The physiochemical characteristics of PCB congeners [41] show that low-chlorinated PCB congeners are characterized by higher volatility. Our sampling sites were located on intensive agricultural territory. We can, therefore, assume that tillage (as another factor) may accelerate loss of law-chlorinated congeners [13]. The effect of time period is also important. Long-term monitoring showed changes in the ratio of low-chlorinated to high-chlorinated biphenyl in favor of high-chlorinated, due to differences in volatility [22, 42]. Probably the most significant factor is the distance from the pollution source [43]. More highly chlorinated congeners would remain closer to their source, since they are less volatile, strongly bound to soil particles, and therefore less readily susceptible to long-range atmospheric transport compared to their less highly chlorinated counterparts [44]. Therefore, the impact of local chemical activity on soil contamination by PCBs was mainly displayed in a relatively high proportion of highly chlorinated PCB congeners.

Conclusion

PCBs have a high organic carbon-water partition (KOC) coefficient, so they associate with soil organic matter. Low mobility together with low water solubility and high persistence results in accumulation in soil. The soil belongs to the greatest reservoirs of these contaminants.

Table 3. Concentrations of indicator PCB congeners [$ng \cdot g^{-1}$] in soil.

PCBs	Mean	S.D.	Range		Median
			Min.	Max.	
All					
52	1.5	1.5	nd	8.8	1.0
101	2.2	2.5	nd	16	1.4
118	4.2	4.7	nk	27	2.6
138	5.4	4.9	nk	26	4.2
153	6.9	7.1	nd	46	4.9
180	4.9	5.3	nk	30	3.1
st ct LV					
52	1.3	1.3	nd	7.2	0.7
101	1.7	1.8	nd	10	1.0
118	2.8	3.5	nk	21	1.7
138	3.8	4.1	0.2	26	2.2
153	6.0	7.4	nk	32	3.0
180	3.3	3.6	0.2	19	2.0
gl FL					
52	1.8	1.7	nd	8.8	1.2
101	2.7	2.8	nd	16	1.8
118	5.3	5.1	nk	27	3.3
138	6.7	4.9	nk	25	5.4
153	7.8	6.9	nd	46	6.0
180	6.2	6.0	nd	30	4.1

nd – non detected
nq- non quantified
st ct LV – Stagnic Cutanic Luvisol
gl FL – Gleyic Fluvisol

Most polluting soils can be expected in areas where the PCBs were manufactured or applied (utilized). To evaluate the impact of pollution source (former producer) and ability to fix PCBs in the soil, in respect to different humus content, we monitored their content in eastern Slovakia. Detected levels of PCBs were compared with current standards in Slovakia, as well as contamination in the other parts of Slovakia. The variability of PCBs content in soil was significantly influenced by soil type and sampling site. Statistically significant change of PCBs in soil is manifested over the longer term. Spring and summer collection in the same year for such a change is too short. Major pollution, as expected, is located next to a chemical factory in the direction of frequent wind flow. The largest share of pollution creates the high-chlorinated congener No. 153.

We point out that, despite the cessation of production of PCBs in the 1980s, soil contamination by former produces are among the critical areas.

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