Health risk assessment of polychlorinated biphenyls (PCBs) in urban soils of Sofia

Anna D. Dimitrova, Yana P. Stoyanova, and Anton K. Tachev

Abstract**—** There is a lack of information regarding levels of polychlorinated biphenyls (PCBs) in soil samples from Bulgaria. This paper reports the concentrations of six indicator PCBs and six dioxin-like PCBs determined in 35 soil samples from urban areas of Sofia City, the capital of Bulgaria. TEQ values of dioxin-like PCBs were calculated also. The results show that the sums of concentration of indicator-PCBs were in the range 7.2–17.2 μg/kg. Small amounts of all dioxin-like PCBs also were found (1.1–5.1 μg/kg). High chlorinated indicator-PCBs: 138, 153 and 180 were the most abundant in the soil samples. Dioxin-like PCB 77 was in minor concentration (0.25 μg/kg) and dl-PCB 118 was predominant dioxin-like congener (0.68 μg/kg). TEQs of soil samples are in the range 0.006 - 0.08 μg/kg. The highest TEQ is found for PCB 126. All concentrations of PCBs determined in this study are below the maximum admissible concentration in soils according to the Bulgarian Legislation and TEQ concentrations met the Canadian soil quality standard. In cases of contamination with low levels of PCBs, as the determined concentration, there is no health risk to humans.

Keywords**—** Dioxin-like polychlorinated biphenyls (dl-PCBs), indicator polychlorinated biphenyls (i-PCBs), risk assessment (RA), toxic endorsed equivalent (TEQ), urban soil samples.

I. Introduction

POLICHLORINATED biphenyls (PCBs) are a class of synthetic organic chemicals made up 209 congeners. They were commercially produced in a large quantity from the 1930s to 1980s because they have extraordinary chemical stability and heat resistance. They were extensively employed as components in electrical and hydraulic equipment such as transformers, capacitors, and hydraulic systems. Also they were used in the electronics industry and as components of adhesives and plastic materials [1-3]. After the establishment that PCBs have adverse health effects their production and using were banned.

Nowadays they still are found as pollutants in the environment compartments in all over the world. They are transported in the atmosphere at over short and long distances [4]. Because of their persistence and hydrophobicity, these ubiquitous compounds accumulate in soils where they are likely to be retained for many years. Consequently, soil is an important reservoir for these compounds [5,6]. The total PCBs burden in global background soils was estimated at about 21 000 tons [7]. Urban areas are an important depot and source of PCBs. Long-range transmission is an important source of PCBs in urban soils [8,9]. In addition, local sources have elevated the PCBs levels in urban soils.

High concentration of PCBs in soils, especially dioxin-like congeners, could cause potential health risks to residents by food and skin intake. Some researchers studied the toxic equivalency/ toxic endorsed equivalent (TEQ) of dioxin-like PCBs in urban soils, and found that PCBs were a potential threat to human health in big cities [10,11]. Therefore the PCB levels and distribution in urban soils are needed to be paid more attention.

Bulgaria has never produced PCBs but used equipment containing biphenyls. Consequently there is a potential risk of pollution. In spite of their negative effect on the environment and human health the levels and distribution of PCBs in the environment has not been well studied in Bulgaria [12].

The objectives of this study were to determine the levels, distribution, and congener profiles of indicator (i-PCBs) and some dioxin- like PCBs (dl-PCBs) in urban soils of Sofia City as a big administrative and industrial centre. Toxic equivalency contributions of dioxin-like PCBs to the total toxic endorsed equivalent (TEQ) and risk assessment were defined.

II. Procedure Materials and Methods

A. Sampling

Sofia City (23°19'39" E, 42°41' 30"N) is the capital of Bulgaria and the biggest city in the country. It has about 1 300 000 residents. It is situated in the west part of Bulgaria and spans an area of over 1349 km^2. There are a lot of unintentional sources of pollution and in particular with PCBs. Therefore it is necessary to investigate the current status of PCBs contamination in the city.

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Five urban sampling areas were chosen (Fig.1). One is in the central part of the City Centre (23°18'16.56"E, 42°40'58.58"N) and another four urban park areas is situated in all directions - North Park (23°18'0.29"E, 42°44'29.28"N), South Park (23°18'31.97"E, 42°40'27.84"N), West Park (23°15'28.89"E, 42°42'9.99"N) and Borisov Garden in the east (23°20'30.58"E, 42°40'43.24"N).

A total of 35 soil samples were collected using the "envelope" method (five individual subsamples - four along the boundaries and one in the center). They were taken from the topsoil over a 50 m^2 area typical for the site. Subsamples were combined and carefully homogenized and then an average sample weight of approximately 1 kg was taken. Total of seven average soil samples were prepared for each sampling area. Samples were collected in all directions. The soil samples were air dried ground and sieved through a < 2 mm sieve before extraction.

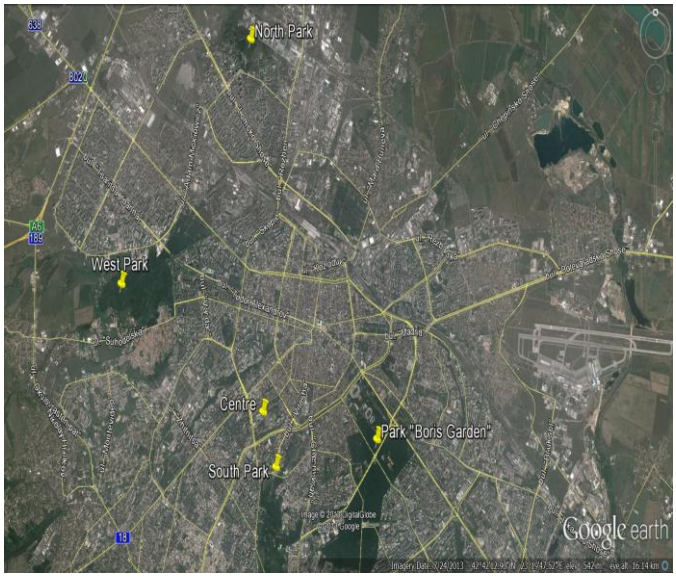


Fig.1 Sampling map of urban soils of Sofia City

B. Chemicals and reagents

Acetone and n-hexane were of analytical grade quality (Merck, Darmstadt, Germany). Anhydrous sodium sulfate and silica gel (70-230 mesh ASTM) for analysis were also from Merck. Cellulose extraction thimbles were from Whatman Ltd (Maidstone, England). Cellulose thimbles were cleaned by Soxhlet extraction with dichloromethane before analysis. Standard solutions of PCB 30, PCB 204 and MIX 20 (mixture of fifteen PCB congeners) were from Dr. Ehrenstorfer (Augsburg, Germany).

C. Extraction and clean up

Soil samples were weighed into Whatman Soxhlet cellulose thimbles, spiked with internal standards, covered with anhydrous sodium sulfate and extracted by Soxhlet technique for 18 h with dichloromethane:hexane (1:1). All extracts were concentrated by rotary vacuum evaporator to 1 mL. The concentrated organic extracts were cleaned with an acid-base-silica column. The analytes were eluted with 50 mL hexane. Elution solvents were concentrated to 1 mL in hexane after rotary vacuum and nitrogen stream evaporation.

D. Instrumental analysis

The analysis was performed with Hewlett Packard Model 5890 Series II PLUS gas chromatograph equipped with Hewlett Packard 5972 Mass Selective Detector (GC/MS). HP-5 fused silica capillary column (30 m length, 0.25 mm i.d., 0.25 μm film thickness) coated with 5% phenyl–95% methylpolysiloxane was used for the analysis.

The column oven temperature program started at 120°C (holding time 1 min), increased to 190°C at 20°C min-1, increased to 230°C at 5°C min-1 and finally to 300°C at 25°C min-1 (holding time 10 min). Injector and detector temperatures were 280°C and 300°C, respectively. Helium was used as carrier gas at a constant flow rate of 0.8 mL min-1. Injection was performed in splitless mode.

The mass spectrometer was operated in electron impact ionization (EI) positive-mode using automatic gain control. The storage window was set between m/z 200 and m/z 500 and selected ion monitoring (SIM). The scan time during data acquisition was set at 1.0 s with four microscans per second. Quantification of the target compounds was performed by monitoring of the characteristic ions m/z 258, 292, 326, 362, 396 and 430. These ions were selected considering the parent ions of the group of PCBs present in MIX 20 (PCB 28, 31, 52, 77, 101, 105, 118, 126, 128, 138, 153, 156, 169, 170 and 180). Calibration was performed by injections of standard solution of MIX 20 at 7 calibration levels (0.01, 0.02, 0.05, 0.1, 0.2, 0.5 and 1 μg mL^{-1}). For all the investigated PCBs no peaks areas in the blanks were found and the limit of detection was defined as the concentration giving a signal-to-noise ratio of 3 (LOD = 0.1 μg/kg). Linearity was investigated within the range between: 0.01–1 μg mL^{-1} and the R^2 values were found to be between 0.992 and 0.999.

E. Statistical analysis

Statistical analyses were conducted with Microsoft Excel (Microsoft Inc., USA) and SPSS 16.0 (SPSS Inc., USA). The distribution of PCBs levels was tested with Kolmogorov–Smirnov test.

III. Results and Discussion

A. PCBs content and distribution in soils

This paper reports initial study results of PCBs levels in urban soil of Sofia City. The results of the analytical chemical analysis of all soil samples were presented on a Fig.2 for each category PCBs – indicator-PCBs (i-PCBs) and dioxin-like PCBs (dl-PCBs). The individual PCBs level of each congener was summarized in Table I.

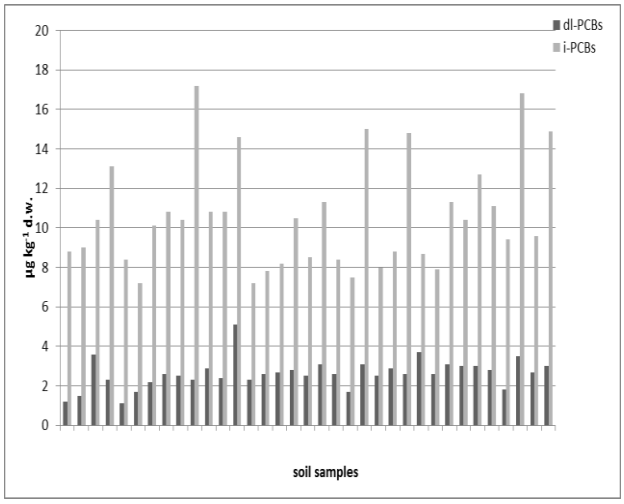


Fig.2 PCBs levels in soils of Sofia (µg/kg d. w.)

Table I. Individual PCB level in soil samples of Sofia City (µg/kg d. w.)

Congener	IUPAC number	Min	Max	Median	Mean	RSD (%)
Tri – CBs*	28	0.2	4.9	1.4	1.56	0.796
Tetra- CBs*	52	0.3	1.9	1	1.06	0.397
Tetra- CBs	77	0.1	0.7	0.3	0.25	0.134
Penta–CBs*	101	0.1	2.6	1.4	1.47	0.596
Penta - CBs	105	0.2	0.9	0.3	0.40	0.184
Penta - CBs	118	0.3	2.1	0.6	0.68	0.341
Penta - CBs	126	0.1	0.8	0.5	0.40	0.202
Hexa–CBs*	138	1.2	5	2.5	2.78	0.983
Hexa–CBs*	153	0.8	3.6	1.9	1.96	0.641
Hexa - CBs	156	0.2	1.1	0.5	0.45	0.174
Hexa - CBs	169	0.1	1.1	0.4	0.44	0.197
Hepta-CBs*	180	0.8	3.7	1.7	1.75	0.542
Σ i- PCBs*		7.2	17.2	10.4	10.58	2.741
Σ dl- PCBs		1.1	5.1	2.6	2.63	0.746

Σ i-PCBs* - indicated the total concentration of six PCB congeners including PCB - 28, 52, 101, 138, 153 and 180.
Σ dl-PCBs - indicated the total concentration of some dioxin-like PCB congeners including PCB – 77,105, 118, 126, 156 and 169.

Analyzed PCBs were detected in all soil samples. It was found that the concentration of total PCBs (sum of indicator-PCBs and dioxin-like PCBs) in all samples were similar and did not vary significantly among sampling locations and direction. The sum of all PCBs ranged in the interval 8.3 - 22.3 µg/kg.

Indicator biphenyls dominated in soil samples. The sum of concentration of six indicator congeners ranged from 7.2 to 17.2 µg/kg, with a mean value of 10.58 µg/kg. These PCBs soil levels were below the maximum admissible concentrations (MAC) of 200 µg/kg in soils according to the Bulgarian Legislation [13]. Also they are below the precautionary target values (20 µg/kg), but exceed the referent background values (5 µg/kg) as set by the ordinance. From this fact it is possible to assume that these biphenyl levels in soils are the result not only of their presence as a global transboundary pollutant in the environment. There are potential sources of biphenyls in the city. But in all studied urban areas of Sofia City are determined similar levels of PCBs ($P>0.05$). It is established a linear correlation between the concentrations of i-PCBs and dl-PCBs in soil samples ($r = 0.41$; $P < 0.05$; $N= 35$). This correlation confirms that the presence of biphenyls in soils due to their emission from the number of unintentional sources such as transport, fuel combustion in households, trade and industry and not from a specific local point source.

The individual distribution of each of indicator PCBs was established and presented on a Fig.3. From indicator PCBs, more chlorinated biphenyls - hexachlorinated PCB 138 and PCB 153 and heptachlorinated PCB 180 were most abundant in soil samples. Probably this abundance is due to their higher hydrophobicty and affinity for strongly adsorption with soil organic matter. The results from Wilcke and Zech [14] have demonstrated that hexachlorinated PCBs are also most abundant in urban and industrial soils.

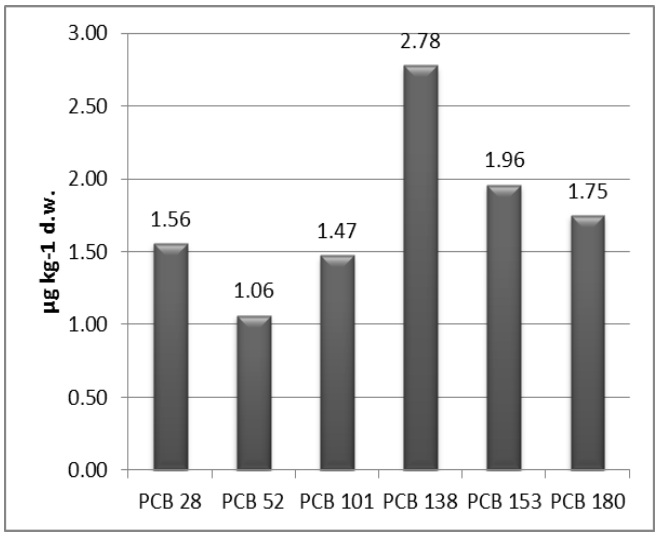


Fig.3 Congener profile of i-PCBs in soil samples

The range of PCB concentration in urban soil samples found in our study was lower in comparison with other countries (Table II).

In Kathmandu (Nepal), Aichner et al. [8] found PCBs content in urban soils from 0.356 to 44.71 µg/kg. Cachada et

al. [15] reported range value between 0.62 – 73 μg/kg, 4.5 – 78 μg/kg, 2.8 – 48 μg/kg, 1.8 – 172 μg/kg and 2.3 – 77 μg/kg for Aveiro (Portugal), Glasgow (Scotland), Ljubljana (Slovenia), Torino (Italy) and Uppsala (Sweden), respectively. The reported data for urban soils from Beijing (China) [16] and Moscow (Russia) [17] are higher than these determined in our study too.

The concentrations of urban soils from Sofia City are higher compared with PCBs levels from woodland regions of Germany (0.2–4.8 μg/kg) [18] and Austria (0.2–7.5 μg/kg) [19] and the Tatra Mountains (Slovakia) (0.87–1.5 μg/kg) [20].

Table II. PCBs concentration in urban soils from other countries (μg/kg d. w.).

Area	Category	Mean	Median	Range	Reference
Kathmandu (Nepal)	Urban	4.965	-	0.356–44.71	Aichner et al., 2007
Aveiro (Portugal)	Urban	-	7.9	0.62–73	Cachada et al., 2009
Glasgow (Scotland)	Urban	-	22	4.5–78	Cachada et al., 2009
Ljubljana (Slovenia)	Urban	-	6.8	2.8–48	Cachada et al., 2009
Torino (Italy)	Urban	-	14	1.8–172	Cachada et al., 2009
Upsala (Sweden)	Urban	-	5.7	2.3–77	Cachada et al., 2009
Moscow (Russia)	Urban and peri-urban	-	-	3.1–42	Wilcke et al., 2006
Beijing (China)	Urban	11.70	13.28	N.D.–37.11	Shan Wu et al., 2011

B. Toxic equivalency of dioxin-like PCBs

Concentration of dioxin-like PCBs in soil samples ranged from 1.1 to 5.1 μg/kg, with a mean value of 2.63 μg/kg (Table 1). The individual distribution of dioxin-like PCBs was established and presented on a Fig.4. The dl-PCB 118 was in major concentration and dl-PCB 77 was in minor concentrations. The levels of highly chlorinated dl-congers (PCBs 105, 126, 156, 169) were relatively low and in the same concentration range.

At a present, in the EU and Bulgarian legislation there are no maximum limits for dl-PCBs in soils in spite of their higher toxicity compared with i-PCBs. The most stringent guideline value is set in Canada (4 pg PCDD/Fs-TEQs g^{-1}) for all land use types of soil [21].

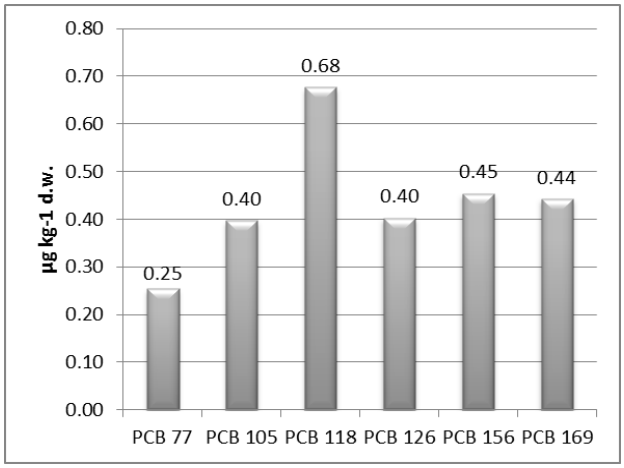


Fig.4 Congener profile of dl-PCBs in soil samples

For the toxicity assessment of dioxin-like PCBs is used toxic equivalency methodology (TEQ) - the levels of dioxin-like PCBs were converted to the TEQ of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin, the most toxic dioxin, with a toxic equivalent factor equal to one [22]. TEQ represents the product of the concentration of individual dl-PCB in a sample and toxicity equivalence factor (TEF) for this dl-compound (1), as defined by the World Health Organization (WHO) for humans and mammals.

$$TEQ = TEF_{congener} \times concentration_{congener} \quad (1)$$

The results of the calculated TEQ concentration of dl-PCBs in soil samples were presented in the Table III.

Table III. TEQ concentration of dl-PCBs in urban soils of Sofia City (x 10^{-3} μg/kg d. w.)

Congener	Min TEQ	Max TEQ	Mean TEQ
77	0.010	0.070	0.025
105	0.006	0.027	0.019
118	0.009	0.063	0.020
126	0.010	0.080	0.040
156	0.006	0.033	0.014
169	0.003	0.033	0.013

Determined TEQ values in soils are in the interval between 0.006 and 0.08 μg/kg. The highest TEQ value is calculated for PCB 126 (0.040 x 10^{-3} μg/kg). It is the most toxic dioxin-like biphenyl with a TEF equal to 0.1. Another dioxin –like congeners have shown similarity in toxic endorsed equivalent.

According to the Canadian soil quality guidelines of dioxins, the TEQ concentrations of all soil samples of Sofia City were below guideline value.

Compared with other urban soils these TEQ levels are lower. In the Chechen Republic the medians of WHO-PCDD/F-TEQ in agricultural, residential, and industrial/post-

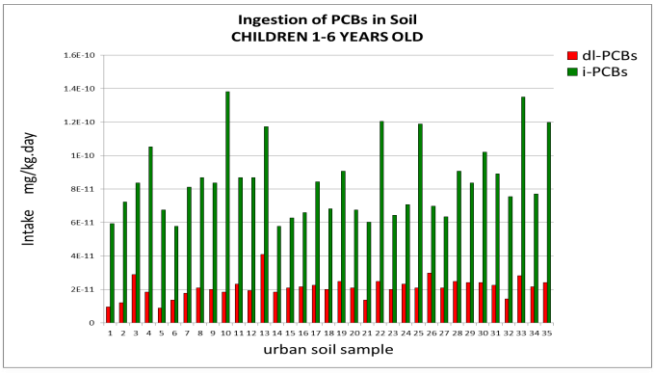
war areas were 0.55, 0.28 and 7.1 x 10^{-3} μg/kg respectively. The medians of WHO-PCBs-TEQs in the same areas were 0.15, 0.13 and 2.9 x 10^{-3} μg/kg [23]. PCDD/F and PCB median content of the humus layer of Norway spruce stands was 6.76 x 10^{-3} μg WHO-TEQ g^{-1} d.w. [24]. Wu et al. found dl-PCBs TEQs values in the range of 0.006 – 1.776, with a mean value of 0.350 x 10^{-3} μg/kg. Their results have demonstrated that TEQ of highly chlorinated dl- PCBs was relatively low [16].

C. Intake of PCBs from soil by ingestion

The ingestion of PCBs from the polluted soil is calculated by the equation:

$$\text{Intake (mg /kg}\cdot\text{day)} = \frac{\text{CS}\times\text{IR}\times\text{CF}\times\text{FI}\times\text{EF}\times\text{ED}}{\text{BW}\times\text{AT}} \quad (2)$$

where CS is the PCBs concentration in the soil (mg/kg), IR is the ingestion rate (200 mg/day for children, 100 mg/day for adults], CF is a conversion factor (10^{-6} kg/mg), FI is the fraction of soil ingested from the polluted site (0.005 for children and 0.001 for adults), EF is the exposure frequency (365 days/year), ED is the exposure duration (70 years for adults, 9 years for children), BW is the body weight (70 kg for an average adult, 16 kg for children), and AT is the averaging time for carcinogenic effects, (70 years x 365 days/year) [25, 26]. Calculations by urban soil samples are given on the fig. 5 for children (1-6 years old) and adults, respectively.



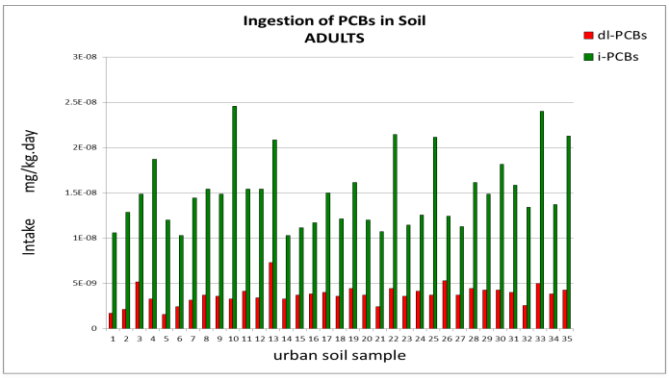
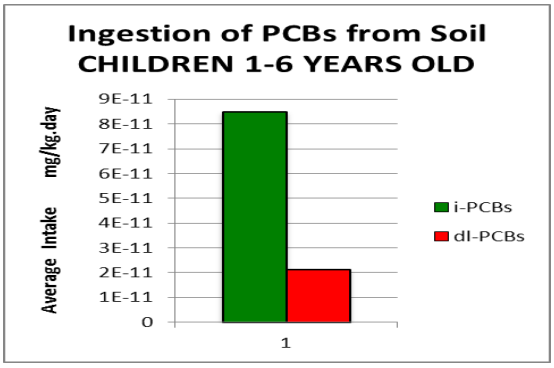


Fig. 5 Ingestion of indicator-PCBs and dioxin-like PCBs from soils for each sample

The cancer risk is calculated by the equation:

$$\text{Risk} = \text{CDI} \times \text{slope factor} \quad (3)$$

where chronic daily intake (CDI) is given by (2) and slope factor for PCBs is 2 mg/kg day [25].



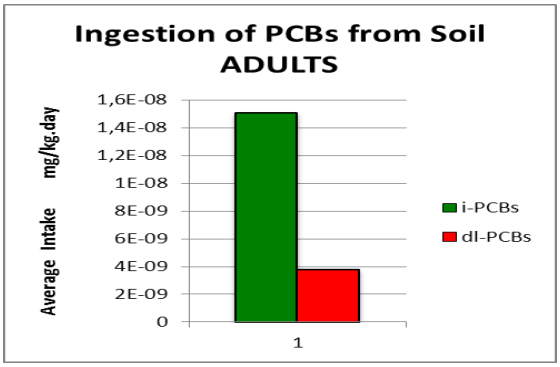


Fig. 6 Average ingestion of indicator-PCBs and dioxin-like PCBs from soil

The calculations of the health risk are made using the average value of CDI from fig. 6 and the results are shown in the Table IV.

Table IV. PCBs Cancer risk from soil ingestion.

Ingestion of PCBs in soil		Cancer risk cases per million people
children	dl-PCBs	0.0000422449
	i-PCBs	0.000169439
adults	dl-PCBs	0.007510204
	i-PCBs	0.030122449

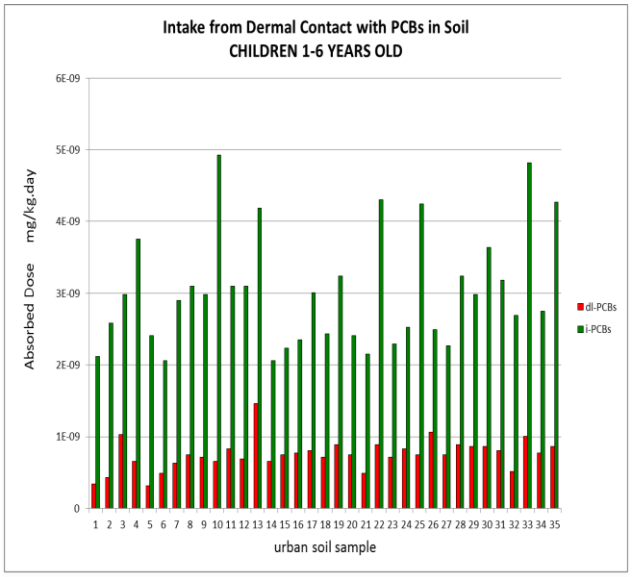
D. Intake of PCBs by dermal contact with contaminated soil

The intake of PCBs from the contaminated soil is calculated by the equation:

$$\text{Absorbed dose (mg /kg}\cdot\text{day)} = \frac{CS \times CF \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \quad (4)$$

where SA is the skin surface area available for contact (410 cm^2/event for children's hands and 820 cm^2/event for adult's hands), AF is the soil to skin adherence factor (1.45 mg/cm2 for commercial potting soil for hands), ABS is the absorption factor (unitless) which accounts for desorption of the pollutant from the soil matrix and absorption of the pollutant across the skin (6 %) [25, 26].

Calculations by urban soil samples are given on the fig. 7 for children (1-6 years old) and adults, respectively.



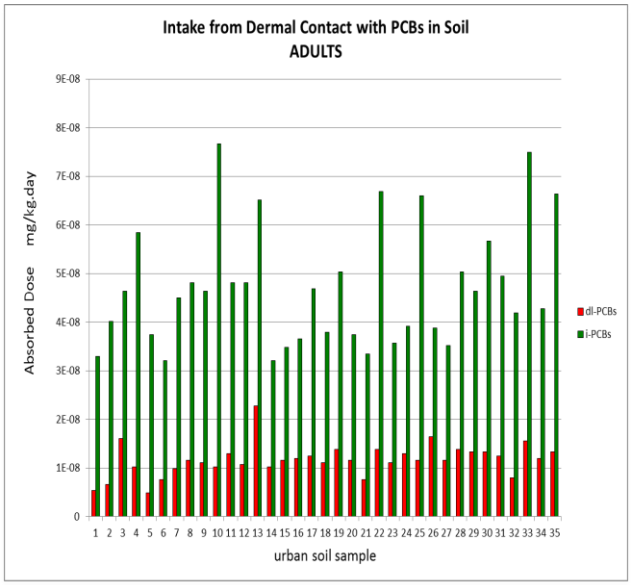
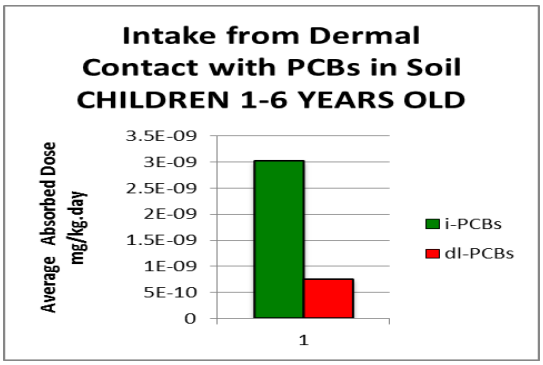


Fig. 7 Intake from dermal contact with indicator-PCBs and dioxin-like PCBs for each soil samples



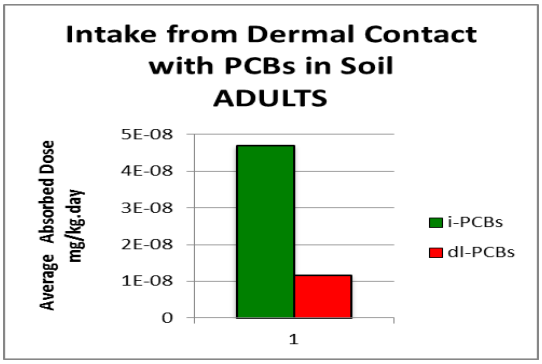


Fig. 8 Average intake by dermal contact with indicator-PCBs and dioxin-like PCBs in soil

The risk of cancer is given by (3) and the calculations of the health risk are made using average value of CDI from fig. 8 and the results are presented in the Table V.

Table V. PCBs Cancer risk from intake of dermal contact with contaminated soil.

Dermal contact with contaminated soil		Cancer risk cases per million people
children	dl-PCBs	0.001506876
	i-PCBs	0.006043881
adults	dl-PCBs	0.023440286
	i-PCBs	0.094015929

IV. Conclusion

The present paper summarized the initial study results of the current levels of indicator and some dioxin-like PCBs in urban soils of Sofia City, the biggest city in Bulgaria.

The results indicated that PCBs (total of indicator and dioxin like congeners) present in all analyzed soil samples. Determined i-PCBs levels in all samples were similar and were below the maximum admissible concentrations of the Bulgarian Legislation.

In general, the PCBs congener specific distribution was similar across all the samples. The high chlorinated indicator biphenyls (hexa- and hepta-CBs) were predominated in the samples. The probable reason is that low chlorinated biphenyls are more volatile while the high chlorinated congeners are more adsorbable to soil organic particles.

This study revealed that the concentrations of dioxin-like PCBs were much lower than those of indicator PCBs. However, the dioxin-like PCBs should be closely monitored in the environmental compartments because of their higher toxicity.

In order to quantify the toxicity of dl-PCBs, toxic endorsed equivalent was adopted to estimate the risk assessment. Calculated TEQs were between 0.006 and 0.08 μg/kg and agreed with Canadian soil quality standard that is the most stringent guideline at a present.

Also human health risk is determined with calculations of the two possible ways of biphenyls soil exposure - soil ingestion and dermal contact with contaminated soil.

The calculation results showed a lack of cancer menace in cases of contamination with low levels of PCBs, as the determined concentration in our study.

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