Congener specific distribution and health risk assessment of polychlorinated biphenyls in urban soils

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Abstract

Polychlorinated biphenyls (PCBs) were primarily used in transformers and capacitors, lubricants, flame retardants, plasticizers, paint, carbonless papers, etc. These are capable of long-range atmospheric transport and have been designated as persistent organic pollutants by the Stockholm Convention. Due to their characteristic properties, PCBs are found worldwide in all environmental matrices (including human) and biota. Soils are usually considered to be the source as well as sink for environmental pollutants, with cumulative effects of long-range atmospheric transport and local sources. Around the world, comparatively higher concentrations of PCBs have been reported in urban soils than suburban or rural soils. Higher amount of PCBs in urban soils may cause toxicological health risks to urban residents through ingestion, inhalation and skin contact. This paper presents the PCB distribution in soils from Delhi, India, and exposure risk estimates for human health through soil ingestion. The concentration of ∑PCBs ranged between 1.08-100.67 ng g^{-1} (mean 21.16 ng g^{-1}±5.24 ng g^{-1}), which was much lower than the Canadian soil quality guideline value of 1.3 mg/kg or 1300 ng g^{-1}. Human health risk estimates through the soil ingestion pathway were made in terms of lifetime average daily dose (LADD), incremental lifetime cancer risks and non-carcinogenic hazard quotient (HQ). The LADD for Delhi adults and children was 3.02×10^{-8} mg kg^{-1} d^{-1} and 1.57×10^{-7} mg kg^{-1} d^{-1}, respectively, which corresponds to toxic equivalent quotients (TEQ) intake of 0.105 pg TEQ kg^{-1} d^{-1} (0.735 pg TEQ kg^{-1} $week^{-1}$) and 0.543 pg TEQ kg^{-1} d^{-1} (3.801 pg TEQ kg^{-1} $week^{-1}$), respectively. The estimated LADD for Delhi residents was lower than the acceptable intake values recommended by the World Health Organization (1 pg TEQ kg^{-1} d^{-1}), the European Commission (14 pg TEQ kg^{-1} $week^{-1}$) and by the Japanese government (4 pg TEQ kg^{-1} d^{-1}). The probability of cancer risk ranges from 6.04×10^{-8} (∑PCBs) to 1.57×10^{-5} (∑TEQ) and 3.13×10^{-7} (∑PCBs) to 8.15×10^{-5} (∑TEQ) for adults and children, respectively, and was within acceptable ranges of 10^{-6} to 10^{-4}. The non-carcinogenic risk in terms of health HQ was 0.105 and 0.330 for adults and children, respectively, which was lower than the acceptable limit of 1. The study found lower concentrations of PCBs than guideline values and low health risk estimates through the soil ingestion pathway within acceptable levels, indicating a minimum risk for Delhi residents.

Introduction

Polychlorinated biphenyls (PCBs) are a group of 209 congeners and have been designated as persistent organic pollutants by the Stockholm Convention. PCBs were primarily used in transformers and capacitors as dielectric and coolant fluids and in lubricants, flame retardants, plasticizers, paint, etc.[1] Even though the production has been banned since the 1970s and usage of these contaminants are restricted in many countries, PCBs continue to be detected in environmental samples from around the world.[2]

PCBs are toxic, bioaccumulative, and can undergo long-range atmospheric transport world-wide to regions such as the Arctic, Antarctic and high altitude regions of Mount Everest.[3-5] PCBs are characterized by high thermal and chemical stabilities, low vapor pressures, high dielectric constants, hydrophobicity, high lipophilicity, and extreme resistant to degradation. Because of these characteristics, PCBs accumulate in soil, sediments and biota.[6]

Soils of urban areas are usually known as the source as well as sink for environmental pollutants, where cumulative effects of long-range atmospheric transport and local sources are the important factors.[7,8] As a result, comparatively higher concentrations of PCBs have been reported in urban soils than suburban or rural soils.[9,10] Humans are exposed to PCBs mainly through the consumption of contaminated food, and occupational exposure to PCBs occurs mainly via the inhalation and dermal routes. Higher amount of PCBs in soils, especially dioxin-like PCBs (dl-PCBs), may cause toxicological health risks to urban residents through ingestion, inhalation and skin contact.[11] In earlier studies, the potential risk to human health from PCBs in urban soils has been reported.[12-13] Therefore, more attention should be given to the status of PCBs in urban soils and the possible risk to human and environmental health.

In earlier investigations, occurrence of PCBs had been reported in environmental and biological samples from India.[14-20] In this study we quantified the levels of PCBs and their risk estimates in urban soils from the Delhi metropolitan area. This is probably the first study to present the 28 PCB-congener levels, including dioxin-like PCBs, in soils from Delhi, and may provide the base-line statistical data needed for health effect assessments. For this purpose, PCBs were extracted from soils using accelerated solvent extraction (ASE) and quantified.

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Key words: polychlorinated biphenyls, urban soil, risk assessment, lifetime average daily dose, health hazard quotient.

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Materials and Methods

Study area and sampling

The sampling area was the national capital territory (NCT), Delhi, which is the capital of India with a population of 1.67 million. The total area of NCT, Delhi, is approximately 1483 km^2 and is located in northern India between 28°24'17"N to 28°53'00"N and between 76°50'24"E to 77°20'37"E. Average annual rainfall is 714 mm. The climate in Delhi is hot and humid. During summer, temperatures rise up to 40-45°C and in winters temperatures fall to 4-5°C.[21]

The soil samples were collected during June 2011 from 14 urban locations on streets and roads near residential areas in Delhi. Approximately 1/2 kg of soil sample was collected from each sampling location, and after removing pebbles, sticks and leaves the sample was mixed thoroughly until homogenized. One part was then transferred to clean, wide-mouthed amber glass containers that were then labeled. After collection, samples were transported to the laboratory and kept at 4°C until analysis.

Chemicals and solvents

Chemicals (sodium sulfate, potassium hydroxide and sulfuric acid) and solvents (high performance liquid chromatography grade acetone, hexane and dichloromethane) were purchased from Merck India. Pre-cleaned silica gel 60 (0.063-0.100 mm) was obtained from Supelco (Sigma-Aldrich Corp., St. Louis, MO, USA) and used as adsorbent in column chromatography. Prior to use, anhydrous sodium sulfate was cleaned separately with methanol, dichloromethane and acetone in Soxhlet extractor for 8 h each, and stored in air-tight conditions at 130°C. Reference standard solutions of PCBs were purchased from Dr. Ehrenstorfer (Dr. Ehrenstorfer GmbH, Augsburg, Germany) and used for the instrument calibration and quantification.

Sample extraction, clean up and analysis

Sample extraction and clean up was carried out according to validated methods.[22] Briefly, a homogenized 20 g sample was dried by mixing with diatomaceous earth (ASE prep DE, Dionex Corp., Sunnyvale, CA, USA) until a free-flowing powder was obtained. The extraction was carried out with accelerated solvent extractor (ASE-350, Dionex Corp.) using acetone/hexane (v/v, 1:1) solvent mixture in two cycles with 5 min static time. The ASE was operated at 1500 psi and the oven temperature was maintained at 100°C. The extracts were concentrated to 2.0 mL using a rotatory vacuum evaporator (Eyela, Tokyo, Japan). Multilayered silica gel column chromatography on a tri-functional column with neutral, basic and acidic silica was performed to remove interfering organic and polar compounds. Details of methodology for clean up and instrumental analysis are reported elsewhere.[20]

Quality control analysis

Appropriate quality assurance quality control analysis was performed, including analysis of procedural blanks (analyzed concentrations<method detection limit, MDL), random duplicate samples (standard deviation, SD<5%), calibration standard verification (SD<15%) and matrix spike recovery (100±20%).[20] PCB congeners were identified in the sample extract by comparing the accurate retention time from the standard mixture and quantified using the response factors from multi-level calibration curves of the standards (r^2 value 0.999).

A signal to noise ratio of 3:1 was used to calculate instrument detection limits by using a valid quantifiable peak. Each sample was analyzed in duplicate and the average was used in calculations. Method detection limits were established by processing 8 aliquots of a sample spiked with a quantity sufficient to produce a detectable response (s/n>3) and multiplying the standard deviation by the $t_{students}$ value (3.0 for 8 replicates). For statistical calculations the non-detect values of PCB congeners have been reported as <0.01 ng g^{-1} (MDL of all 28 individual congeners being less than 0.01ng g^{-1}). Furthermore, it may be noted that our laboratory is ISO 17025 accredited and had been participating in proficiency testing exercises conducted by international agencies, including the Centre d'expertise en analyse environnementale du Québec, and performance scores were satisfactory for PCBs.

Toxic equivalent quotients (TEQ) of each congener were calculated by multiplying the concentration of individual dl-PCB congener with the toxic equivalent factors (TEFs)[11] with reference to 2,3,7,8-tetrachloro dibenzo-*p*-dioxin (TCDD) and reported as pg WHO_{2005}-TEQ g^{-1} dry-weight (dw).

Health risk assessment for polychlorinated biphenyls exposure

In this study, assessment of human health risk is discussed as the calculated estimates of the upper-bound excess probabilistic lifetime cancer risk and non-carcinogenic hazards. Hazard is the exact measure of the magnitude of exposure potential or a quantifiable potential for developing non-carcinogenic health effects after averaged exposure period. Risk is the probability of cancer development in a lifetime after a uniform exposure.[23] We estimated the human exposure to PCBs through soil ingestion and the consequent health risk by using equations presented by United States Environmental Protection Agency.[24-26] For this purpose, the LADD (lifetime average daily dose), non-cancer risk HQ (hazard quotient) and probabilistic incremental lifetime cancer risks (ILCR) were calculated. LADD is an amount a person takes in as a result of exposure to a chemical in contaminated air, water, soil or food. The input parameters used in the health risk estimation for PCBs are given in Table 1. The equations used for estimating LADD, HQ (non-cancer risk) and probable cancer risk were as follows:

$$\text{LADD (mg kg}^{-1}\text{ day}^{-1}) = (\text{Cs x IR x F x EF x ED})/(\text{BW x LT}) \quad (1)$$

$$\text{HQ} = \text{LADD/RfD} \quad (2)$$

$$\text{ILCR} = \text{LADD x cancer slope factor oral} \quad (3)$$

where:
Cs is the total PCBs concentration in soil (μg kg^{-1});
IR is the soil ingestion rate (mg day^{-1});
F is the unit conversion factor (10^{-9});
EF is exposure frequency (days/year);
ED is the exposure duration (year);
BW is the body weight (kg);
LT is the lifetime which is equal to exposure duration x 365 days (days);
RfD is the reference dose for individual dl-PCB congener (mg kg^{-1} day^{-1}).

An RfD is a daily intake rate that is estimated to cause no adverse health effects over a specific exposure duration. Non-carcinogenic risks (HQ) were assessed by comparing exposure with RfD of each dl-PCB and the total was reported for each sampling location. Cancer risk for dioxin-TEQ was calculated for each of the 12 dl-PCB congeners from LADD by multiplying slope factor for dioxin (150,000/mg kg^{-1} day^{-1}) and the total was reported. Cancer risk for non-dl-PCBs was calculated from LADD of 28 PCBs by multiplying the upper bound cancer slope factor (2/mg kg^{-1} d^{-1}). The upper

Table 1. Summary of input parameters used in calculations for health risk assessment.

Symbol	Parameter	Unit	Point estimate
Cs	PCB concentration	μg kg^{-1}	Data from this study
C_{TEQ}	TEQ concentration	μg TEQ kg^{-1}	Data from this study
IR	Soil ingestion rate	mg day^{-1}	100 (adults), 200 (children)
EF	Exposure frequency	Days/year	365 days
ED	Exposure duration	Years	70 (adults), 12 (children)
BW	Body weight	Kg	70 (adults), 27 (children)
LT	Life time	Days	70×365=25,550 (adults) 12×365=4382 (children)
CSF	Cancer slope factor	Per mg kg^{-1} day^{-1}	Dioxin-TEQ: 150,000 Non-dl-PCBs: 2 (upper bound)
F	Unit conversion factor	-	10^{-6}

Cs, concentration in soil; dl-PCBs, dioxin-like polychlorinated biphenyls; TEQ, toxic equivalent quantities.

bound slope factor is used for those exposure pathways for which environmental processes are likely to increase health risk, such as exposure from consumption of foods, sediment or soil ingestion, and dust or aerosol inhalation.

Results

Polychlorinated biphenyls concentrations in soil

The concentrations of ∑PCBs and individual 28 congeners in urban soils from Delhi, India, are presented in Table 2. The observed levels of total PCBs ranged between 1.08- 100.67 ng g^{-1} (dw) with the mean and median of 21.16 ng g^{-1} and 10.16 ng g^{-1} (±5.24 ng g^{-1}), respectively, which is at par with those of other cities around the world. The most common congener numbers among studied PCBs were CB-44, CB-52 and CB-138. The concentration range of 12 dl-PCBs was 0.10-22.57 ng g^{-1} (mean 5.33 ng g^{-1}±0.55 ng g^{-1}, median 2.00 ng g^{-1}±0.55 ng g^{-1}) and accounted for approximately 25% of total PCBs. TEQ of 12 dl-PCBs, calculated using WHO $_{2005}$-TEFs, ranged from less than 1 to 387 pg-TEQ g^{-1} (mean 73±28 pg-TEQ g^{-1}). Non-ortho-PCBs (CB-77, CB-81, CB-126 and CB-169) were the sole contributors and accounted for over 99% of total TEQ (Table 3). Tetrachorinated, pentachlorinated and hexachlorinated biphenyls were the dominant homologues groups of PCBs (Table 4, Figure 1).

Health risk assessment

Adults and children may be exposed to contaminants in soil and may also take chemicals from soil through different intake pathways, such as ingestion, inhalation and dermal contact. Figure 2 shows average of ∑PCBs in urban soil at different locations in Delhi, India. The calculated average daily intake of PCBs through soil ingestion by adults and children in Delhi was 3.02×10^{-8} mg kg^{-1} d^{-1} and $1.57x10^{-7}$ mg kg^{-1} d^{-1}, respectively (Table 5), for a soil PCB concentration of 21.2 μg kg^{-1}. With respect to RfD, the average LADD for adults and children was less than 1% and less than 5%, respectively, of the RfD. The calculated value of TCDD substituted WHO-TEQ daily intake for adults and children was $1.05x10^{-10}$ mg TEQ kg^{-1} d^{-1} and 5.43×10^{-10} mg TEQ kg^{-1} d^{-1}, respectively, when soil TEQ was 0.073 μg kg^{-1} (Table 5). The calculated probability cancer risk estimate for PCB exposure from ingestion of soil was $6.04x10^{-8}$ (∑PCBs) to 1.57×10^{-5} (∑TEQ) and $3.13x10^{-7}$ (∑PCBs) to 8.15×10^{-5} (∑TEQ) for Delhi adults and children, respectively (Table 5). The quantified non-carcinogenic HQ for soil ingestion pathway of PCBs was 0.105 and 0.330 for Delhi adults and children, respectively (Figure 3).

Table 2. Polychlorinated biphenyl congener concentrations (ng/g dw) in urban soils from Delhi, India.

Congeners	Range	Mean	Median	SE*	%
PCB-18	0.17-4.31	1.02	0.51	0.18	3.8
PCB-37	0.21-7.07	1.38	0.76	0.27	5.1
PCB-44	0.24-15.60	2.78	1.13	0.61	11.2
PCB-49	0.22-10.20	1.96	1.01	0.36	7.9
PCB-52	0.18-11.2	2.56	1.00	0.52	11.3
PCB-70	0.08-6.30	1.30	0.43	0.28	5.3
PCB-74	0.16-5.11	1.35	0.59	0.23	5.5
PCB-119	<0.01				
PCB-128	0.09-2.62	0.55	0.34	0.10	2.2
PCB-138	0.06-12.80	2.54	1.02	0.55	11.1
PCB-151	0.12-2.66	0.70	0.21	0.12	1.9
PCB-168	0.08-9.68	1.61	0.49	0.37	7.6
PCB-170	<0.01				
PCB-177	0.09-0.76	0.36	0.22	0.04	1.1
PCB-187	0.11-0.70	0.41	0.41	0.06	0.3
PCB -207	0.09-0.55	0.32	0.32	0.03	0.5
∑PCBs	**0.77-78.93**	**15.83**	**7.30**	**3.29**	**74.8**
dl-PCB-77	0.11-2.25	0.63	0.41	0.10	1.5
dl-PCB-81	<0.01				
dl-PCB-126	0.07-2.63	0.62	0.40	0.10	2.3
dl-PCB-169	0.13-4.12	1.14	0.61	0.18	3.9
dl-PCB-105	0.25-8.5	1.60	1.06	0.32	6.0
dl-PCB-114	0.22-3.55	1.79	1.79	0.30	1.2
dl-PCB-118	0.10-1.65	0.72	0.54	0.07	3.2
dl-PCB-123	<0.01				
dl-PCB-156	0.08-2.87	0.78	0.20	0.15	1.8
dl-PCB-157	0.10-2.70	0.93	0.74	0.14	1.6
dl-PCB-167	0.07-3.41	0.80	0.30	0.17	2.7
dl-PCB-189	0.09-1.30	0.41	0.32	0.05	1.1
∑dl-PCBs	**0.10-22.57**	**5.33**	**2.00**	**0.55**	**25.2**
Total $_{(PCBs+dl-PCBs)}$	1.08-100.67	21.16	10.16	5.24	100

<0.01, below detection limit; *SE, standard error (SD/√n); dl-PCBs, dioxin-like polychlorinated biphenyls.

Table 3. Toxic equivalent quotients of dioxin-like polychlorinated biphenyl congeners in urban soils from Delhi, India.

dl-PCBs	(pg WHO_{2005}-TEQ g^{-1})			
	Range	Mean	SE	%
Non ortho-PCBs	<1-387	73	28	99
Mono ortho-PCBs	<1	<1	<1	<1
∑dl-PCBs	<1-387	73	28	100

SE, standard error; dl-PCBs, dioxin-like polychlorinated biphenyls; TEQ, toxic equivalent quotients.

Table 4. Polychlorinated biphenyl homologues (3-7 CB) in urban soils from Delhi, India (ng/g dw).

Homolog	Tri-CB	Tetra-CB	Penta-CB	Hexa-CB	Hepta-CB
Mean	1.88	9.03	4.94	4.52	0.53
Median	1.05	3.55	1.63	1.31	0.22
Range	<0.01-11.38	0.23-48.93	<0.01-31.30	0.06-24.31	<0.01-2.05
% of ∑PCBs	10.3	49.0	17.3	18.8	4.7

PCBs, polychlorinated biphenyls.

Table 5. Calculated polychlorinated biphenyl exposure from urban soil and risk for adults and children.

Soil ingestion	Concentration (μg/kg)	Adults		Children	
		LADD	Risk	LADD	Risk
Dioxin-TEQ	0.073	1.05×10^{-10}	1.57×10^{-5}	5.43x10-10	8.15×10^{-5}
Non-dl-PCBs	21.2	3.02×10^{-8}	6.04×10^{-8}	1.57×10^{-7}	3.13×10^{-7}

LADD, lifetime average daily dose (mg/kg bw/d); dl-PCB, dioxin-like polychlorinated biphenyls; TEQ, toxic equivalent quotients.

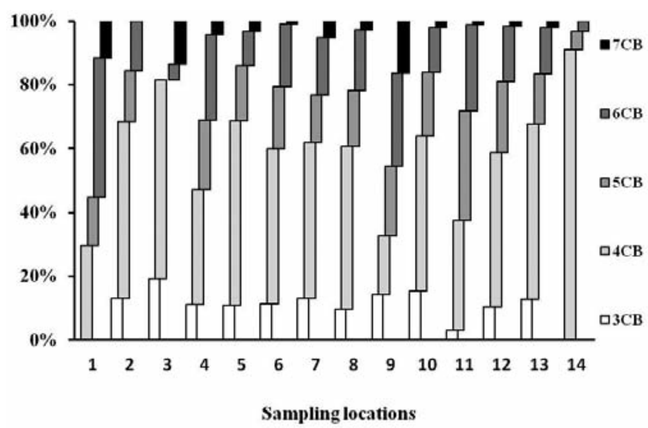

Figure 1. Polychlorinated biphenyl homologs in soil at different locations in Delhi, India.

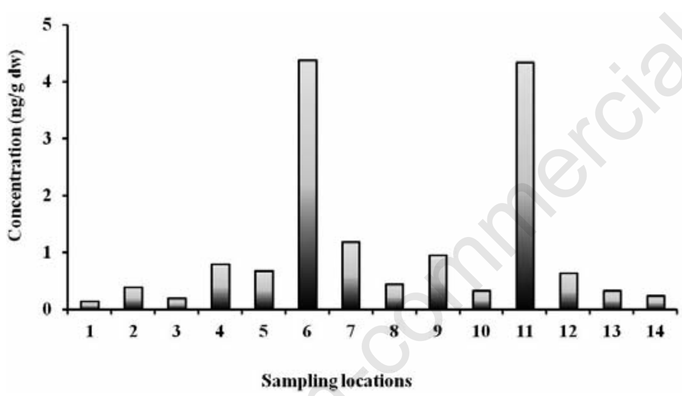

Figure 2. Average of ∑polychlorinated biphenyls in urban soil at different locations in Delhi, India.

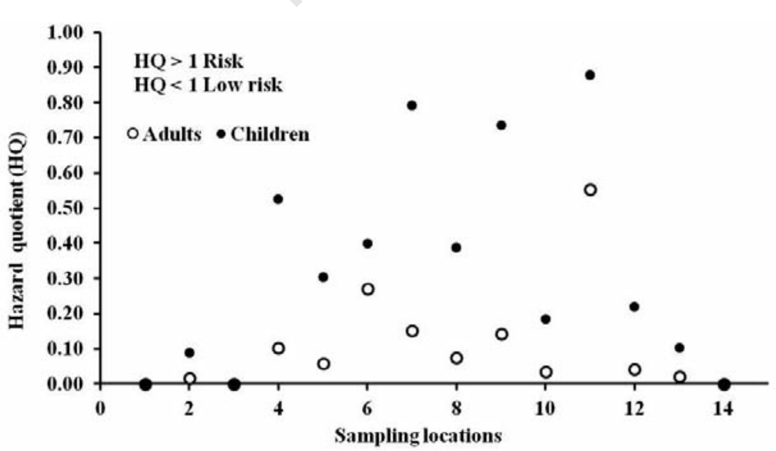

Figure 3. Health risk hazards of polychlorinated biphenyls for adults and children through soil ingestion.

Discussion

Concentrations and possible sources

The PCBs contamination levels in soils from Delhi were much lower than soil quality guidelines (1.3 mg/kg or 1300 ng g^{-1}).[27] The results of this study were similar to observations reported for urban soils from Kathmandu,[7] urban soils of Beijing[28] and from Tibet.[29] However, the concentrations of PCBs in Delhi soils were lower than those of Vietnam,[30] Switzerland,[31] Romania,[32] China,[33] San Felipe, Nuevo Mercurio, Zacatecas, Mexico,[34] Moscow[35] and Guangdong Province, South China,[36] but higher than PCBs reported in soils from Turkey.[37]

The PCB patterns show that the low molecular weighted PCBs were major contributors (60%) compared to high molecular weight PCBs (with 40%). Low molecular weight PCBs were primarily used in electrical equipments while high molecular weight PCBs were mainly used as additives in various applications.[38] PCBs had never been produced in India, but are used in industrial applications.[39] Possible sources of PCBs releases in the study area may be from electronic and electrical waste recycling and industrial emission depositions. During the burning of different mixtures of waste content, garden waste, paper, plastics, PVC (polyvinyl chloride) and painted wood may produce relatively large amounts of dioxin like-PCBs.[40-42]

Risk estimates

To ensure public health safety, a tolerable daily intake (TDI) has been set by international agencies. The TDI is the amount of intake per kg of body weight per day of a chemical substance suspected of having adverse health effects when absorbed into the body over a long period of time. The joint FAO/WHO Expert Committee on Food Additives[43] recommended a TDI of 1.0 pg TEQ $kg^{-1}d^{-1}$ for acute, subchronic and chronic exposures to dioxins. The European Commission[44] established an acceptable weekly intake of 14 pg WHO-TEQ/kg body weight for dioxins and 12 pg dioxin-like PCBs. The Japanese government[45] established the TDI of dioxins at 4 pg-TEQ $kg^{-1}d^{-1}$.

The observed LADD of TEQ intake for adults (1.05×10^{-10} mg TEQ kg^{-1} d^{-1}) and children (5.43×10^{-10} mg TEQ kg^{-1} d^{-1}) are much lower than recommended acceptable daily intake. The acceptable risk distribution is equal or lower than 10^{-6} for carcinogens and may be up to 10^{-4}. The observed ILCR (10^{-8}-10^{-5}) and HQ values less than the acceptable risk level (HQ=1) (Figure 3) suggests minimum risk to the adults and children due to exposure to PCBs through urban soils in Delhi.

Conclusions

Low concentrations of PCBs in Delhi soils, and subsequently their intake values, were lower than those established in international guidelines. Therefore, estimates of cancer and non-carcinogenic risk through soil ingestion were low, indicating no harmful effects on the population of Delhi. In future, it may be useful to conduct more intensive assessments for persistent organic pollutants, in response to human health and environmental concerns.

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