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Distribution of Polychlorinated Biphenyls in Surface Waters of Various Sources from National Capital Region Delhi India

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Abstract

This paper presents the concentration of twenty eight polychlorinated biphenyls (PCBs) including twelve dioxin-like PCBs (dl-PCBs) congeners in surface water of rivers canals lakes ponds and drains from National Capital Region (NCR) Delhi (India). The total concentrations of 28 PCBs were ranged between 14-1768 ng L^{-1} with a mean of 332±42 ng L^{-1}. The concentration of dl-PCBs was ranged between <1-146 ng L^{-1} with the mean of 40±4 ng L^{-1} and accounted 12% for total 28 PCBs. The toxicity equivalent calculated using WHO toxic equivalency factors (TEFs) was presented and discussed. In terms of the homolog distribution it was dominated by 3–5 chlorinated biphenyls. The tetra-PCBs (65%) dominate the PCB homolog followed by tri-PCB (47%) and penta-PCB (21%). The concentrations were lower than the guideline values; however levels at some locations were higher. Further in-depth study is proposed to determine PCBs its bioavailability and bioaccumulation through tissues of aquatic biota to assess the risk of these contaminants on the ecosystems and human health more thoroughly.

Keywords: polychlorinated biphenyls (PCBs) dioxin-like PCBs surface water Delhi India

1. Introduction

Polychlorinated biphenyls (PCBs) are a group of chemicals that have extremely high boiling points and are nonflammable chemicals which are primarily used in transformers capacitors paints and printing inks and also in many other industrial applications (USEPA 1996; Erickson & Kaley 2011). PCBs are amongst the industrial chemicals and have no known natural sources in the environment but they entered the environment through accidental spills and leaks during the transport of the chemicals or from leaks or fires in transformers capacitors or other products containing PCBs (ATSDR 2000). Coplanar or dioxin-like PCBs are formed unintentionally in the same way as polychlorinated dibenzo-*p*-dioxins/ polychlorinated dibenzo-*p*-furans (PCDDs/PCDFs) (Koppe and Keys 2001; Ishikawa *et al*. 2002; Takasuga *et al* 2004). Potential sources of dioxin like compounds are chemical- and petrochemical plants ferrous and non-ferrous metal smelting operations paper and pulp industries cement production and fuel combustion. Smaller non-point sources include domestic burning of wood landfill fires and open burning as well as by natural processes such as vegetation fires (UNEP 2005).

The environmental contamination of Polychlorinated biphenyls (PCBs) was recognized since more than 45 years ago by Soren Jensen in Sweden (Jensen 1966). Polychlorinated biphenyls (PCBs) are persistent organic pollutants (POPs) resistant to chemical physical and biological degradation and being ubiquitously found in all environments of the earth (Atlas and Giam 1981; de Boer *et al.* 2000; Alison & Jason 2008). These are long range atmospheric transport (LRAT) pollutants and have been transported world-wide affecting regions far from their original sources such as the Arctic (Oehme 1991; Harner *et al.* 1998; AMAP 1998) and Antarctic (George and Frear 1966; Risebrough *et al*. 1976; Hyun *et al*. 2010). Even though the production of these contaminants has stopped but still continues to be detected in rivers and water bodies of many countries (Barra *et al*. 2005; Neamtu *et al*. 2009).

Polychlorinated biphenyls (PCBs) are EPA's most toxic chemicals and are classified as probable human carcinogens (Group B2). The International Agency for Research on Cancer has determined that PCBs are probably carcinogenic to humans (Group 2A). A common way for PCBs to enter your body is by eating meat fish or dairy products or other foods that contain PCBs (ATSDR 2000). In the aquatic environment PCBs are bio-concentrated and transferred in the food chains and may return to humans with the aquatic food (Kuwabara *et al*. 1979; Alcock *et al*. 1998). Polychlorinated biphenyls (PCBs) have a wide range of acute and chronic health effects in humans including cancer neurological damage

reproductive disorders immune suppression birth defects and are also suspected endocrine disruptors (Van den Berg *et al*. 2006).

In May 2004 Stockholm Convention on POPs entered into force with the intention of reducing and ultimately eliminating these pollutants. As a party to the Convention India is legally obligated to abide by the objectives of the treaty and is encouraged to support research on POPs. As per national implementation plan submitted to Stockholm Convention on Persistent Organic Pollutants the PCBs have never been produced in India but used in many industrial applications mainly in electric transformer. The data on the transformers containing PCBs were inventoried and showed that around 9837 tons of PCBs exist in the country (NIP 2011).

Investigations dealing with various matrices in India revealed PCBs contamination in water (Babu Rajendran *et al*. 2005; Kumar Sanjay *et al*. 2008), soils (Minh *et al*. 2006; Kumar *et al.* 2011[A]), sediments (Guzzella *et al*. 2005; Babu Rajendran *et al*. 2005; Binelli *et al*. 2009; Sahu *et al*. 2009; Kumar *et al*. 2008; 2011[B]), atmospheric air (Zhang *et al.* 2008; Chakraborty *et al*. 2010; Pozo *et al*. 2011), food commodities (Kannan *et al*. 1992) and biota including humans (Kannan *et al*. 1995; Tanabe *et al*. 1998; Senthilkumar *et al*. 2000 2001; Kunisue *et al*. 2003; Kannan *et al*. 2005; Reddy *et al*. 2006; Devanathan *et al*. 2008; Someya *et al*. 2009). There is no reported literature on PCBs in surface water from NCR Delhi India. Therefore this study is the continuation of research on POPs in India and focused on measuring PCBs including dioxin like PCBs in surface water of Rivers canals drains lakes and ponds from National Capital Region Delhi (India).

2. Materials and Methods

2.1 *Study Area and Sampling*

National capital territory (NCT) Delhi is the administrative capital city of India with population ~18 million with a total area of 1483 km^2 lies between 28^0 36' 36" N to 77^0 13' 48"E. The National Capital Region (NCR) Delhi comprises by the National Capital Territory (NCT) Delhi and the delineated area of the surrounding states of Haryana Uttar Pradesh and Rajasthan (Figure 1). The entire NCR Delhi region **is** a stretch of about 30242 sq. kms. There are specified industrial zones in Delhi with more than 8000 small to medium industries for food textile chemical paints and dyes metallic non-metallic etc. (DoEF 2010).

Sampling locations in the study area were in three districts each of Delhi Uttar Pradesh and Haryana where agriculture and industrial activities are prominent. Total 67 samples collected from major Rivers irrigation canals lakes ponds and drains during January 2010. Amber glass sampling bottles (1L) were washed successively with detergent tap water and distilled water before sampling. 1 L surface water sample was collected using stainless bucket and transferred to sampling bottle. The glass bottle was filled to the top with the sample water to eliminate air bubbles. After proper labeling, the sample bottles were transported with ice to the laboratory and stored at 4°C followed by an extraction within 7 days.

2.2 *Chemical sand Solvents*

Chemicals and solvents were purchased from Merck India. Silica gel 60 (0.063 – 0.100 mm) was from Sigma-Aldrich. Prior to use silica gel and anhydrous sodium sulphate was cleaned separately with methanol dichloromethane and acetone in Soxhlet extractor for 8 h each and stored air tight at 130^0 C. PCBs congener mixture standard solutions were purchased from Dr. Ehrenstorfer (GmbH Germany).

2.3 *Analytical Methods*

Samples were extracted purified and analyzed according to the methods established by U. S. Environmental Protection Agency (USEPA). Briefly 1L water sample was extracted with dichloromethane (50 ml 25 ml and 25 ml). The dichloromethane extract (lower layer) was drained into a funnel containing 5 cm of anhydrous sodium sulphate. The dichloromethane extract was then evaporated to 2-3 ml on the rotary evaporator (Eyela Japan). Extracts were cleaned using silica chromatography which consisted of 2 cm of anhydrous sodium sulphate (about 1.0 g) overlaid with 5 cm of activated silica gel (about 10.0 g) and topped with another 2 cm of anhydrous sodium sulphate. Once the column was pre-rinsed with 30 ml of hexane the sample was added and then a 50-ml mixture of hexane and dichloromethane (DCM) (1:1 v/v). The eluted extract was concentrated using Rotatory

vacuum evaporator TurboVap (Caliper USA) under a gentle stream of pure nitrogen and solvent-exchanged into hexane to 1.0 ml. The extract was transferred to auto sampler vial and 1 µl was injected onto a gas chromatograph equipped with an electron capture detector (GC-ECD) for quantification.

The separation and quantification of polychlorinated biphenyls (PCBs) was performed by gas chromatography (Shimadzu 2010 Japan) attached with autosampler, and equipped with an Electron Capture Detector (ECD ^{63}Ni) on capillary column (HP-5MS Agilent) 60 m x 0.25 mm x 0.25 µm film. The temperature program of the column oven was set to 170^{o} C for 1 min then increased with 3^{0}C min^{-1} to 270^{o}C kept for 1 min then further ramped with 10^{o}C min^{-1} to 290^{o}C at and kept for 3 min. The injector and detector temperature were maintained at 225^{o}C and 300^{o}C respectively. Purified nitrogen gas was used as carrier at the flow rate of 1.0 ml. min^{-1}.

2.4 *Quality Assurance/Quality Control (QA/QC)*

Certified reference standards from Dr. Ehrenstorfer (GmbH Germany) was used for the instrument calibration and quantification of PCB congeners. The 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 five level calibration curves of the standards. Appropriate quality assurance quality control (QA/QC) analysis was performed including analysis of procedural blanks (analyte concentrations were <MDL 'method detection limit'), random duplicate samples (Standard deviation <5), calibration curves with the r^2 value of 0.999. Each sample was analysed in duplicate and the average was used in calculations. Calculated concentrations were reported as less than the limit of detection if the peak area did not exceed the specified threshold (three times the noise). Concentrations below the limit of detection were assigned zero values for the statistical analysis. PCB congeners are denoted by their International Union of Pure and Applied Chemistry (IUPAC) numbers.

Dioxin-like PCBs are assigned with the toxic equivalent factors based on the relative toxicity with 2378-tetrachloro dibenzo-*p*-dioxin (TCDD) (Van den Berg *et al*. 2006). Toxic equivalent quantities (TEQ) were calculated by multiplying the concentration of individual dl-PCB congener with the corresponding WHO toxicity equivalent factors (TEFs). The results were reported as ng L^{-1} and pg WHO_{2005}-TEQ L^{-1}.

3. Results and Discussion

3.1 *Distribution of PCBs*

Concentrations of total 28 PCBs congeners in surface water samples from Rivers canals lakes ponds and drains of NCR Delhi varied from 14-1969 ng/L with the mean and median of 332±42 ng/L and 275± 42 ng/L respectively (Table 1). The average concentration of total non-dl-PCBs among the 67 sampling sites was 293±41 ng/L with a range of 12-1969 ng/L and total dl-PCBs concentrations varied from <1-146 ng/L with a mean of 40±4 ng/L. Congener no CB-18 CB-44 and CB-70 were the dominant among the non-dl-PCBs with 15%, 29% and 25% respectively. Among dl-PCBs congener no 77, 81, 114 and 123 were the main contributors with 19%, 14%, 14% and 20% respectively. The contamination pattern with average ∑PCBs in water samples from different sources was observed as: Hindon River (477±212 ng/L) > Lakes (417±222 ng/L) > Canals (400±104 ng/L) > Yamuna River (364±52 ng/L) > Drains (298±50 ng/L) > Ponds (269±42 ng/L) (Table 2). River, Lakes and Ponds receives the wastewater though drains, however; Canals cover the agricultural areas and used for irrigation purposes which may be the sources to these water systems for PCB contamination. Significant levels of PCBs in different environmental matrices from Delhi and adjoining areas have been reported by Kumar *et al*. (2008 2011^{AB}) and Chakraborty *et al.* (2010). Other studies on PCBs distribution in tropical environmental water have reported the results similar to our study. Zhang *et al.* (2002) and Chau (2005) reported the PCBs values of 33-1064 ng/L (mean of 295 ng/L) and 91-1353 ng/L respectively, in water from Pearl River, China. Sulej *et al.* (2011) reported the concentration of PCBs in runoff water at Gdansk airport, Poland (average 60-440 ng/L). The higher PCBs levels in the literature have been reported as 6721 ng/L for water samples from water bodies and Rivers in northern Nigeria (Okeniyia *et al*. (2009). Ezemonye (2005^{AB}) reported the PCBs levels of 350-1300 ng/L in Warri River, 1500 ng/L for Ethiope River and 30–2930 ng/L for Benin River in Nigeria. Aydin *et al.* (2004) reported the PCBs levels of 505-2377 ng/L in urban wastewater of Konya, Turkey. Zhang *et al.*

(2003) reported the PCBs levels of 204–2473 ng/L in surface water of Minjiang River, Southeast China. However lower than our concentrations of PCBs have been reported by He *et al*. (2011) in main stream water of Jiangsu section of the Yangtze River, China (<0.21–44.4 ng/L); Wan *et al*. (2011) in Dianchi Lake of Kunming, China (13 to 72 ng/L); Lana *et al*. (2008) for local water bodies from Southern Moravia Region, Czech Republic (5.2 to 190.8 ng/L); Zhang *et al*. (2011) in water from Yangtze River, China (1.23 to 16.6 ng/L); Dai *et al*. (2011) in surface water from Baiyangdian Lake in North China (19.46 to 131.62 ng/ L).

3.2 *PCB Homolog Profiles*

PCBs are not used as single compounds but as technical mixtures. 70% of PCBs produced globally were tri- tetra- and pentachlorinated biphenyls with those trichlorinated ones as dominating homologues (Breivik *et al*. 2002). Trichlorobiphenyl was primarily used in power capacitors and transformers while pentachlorobiphenyl was mainly used as a additive (You *et al*. 2011). PCB homolog patterns in surface waters from NCR Delhi India are presented in Table 3 & Figure 2. In terms of the distribution of homolog congeners, it was clearly dominated by those with 3–5 chlorinated biphenyls. The tetra-PCBs (65%) dominate the PCB homolog in average followed by tri-PCB (47%) and penta-PCB (21%). The PCB patterns show that the percentage (average 85% for tri- to tetra-PCBs) of lighter-weighted molecular PCBs (LWM-PCB) in the water samples were much higher than those higher-molecular weight PCBs (HMW-PCBs) (with only 15%) which suggests that heavier PCBs may be deposit in bottom sediment. In an aquatic environment the PCBs in water may come from industrial and municipal wastewater discharges and air deposition and then tend to be adsorbed on the particles in water and settle in the sediment which could naturally become a sink of PCBs as PCBs are non-ionic compounds and the octanol/water partition coefficient (K_{OW}) is in the range of 104–108.

3.3 *Toxic equivalency (TEQ) of dioxin-like PCBs*

Several PCBs have been shown to cause toxic responses similar to those caused by 2,3,7,8-tetra-chlorodibenzo-*p*-dioxin (TCDD) the most potent congener within these groups of compounds (Giesy and Kannan 1998). As a result the concept of toxic equivalency factors (TEFs) established by the World Health Organization (WHO) has been developed to assess the impact of these compounds on human and environmental health. The reported concentrations of dioxin-like congener can be converted into 2,3,7,8-TCDD TEQ concentrations. TEQ concentrations of PCBs with established dioxin-like activity especially the non- and mono-ortho substituted PCBs in surface waters from NCR Delhi India were calculated by multiplying the concentration of each dioxin-like congener by its 2,3,7,8-TCDD TEF (Van den Berg *et al*. 2006).

In present study the TEQ for 12 dl-PCBs was presented in Table 4. The values were ranged between <1-2314 pg WHO_{2005} TEQ L^{-1} with a mean value of 290±53 pg WHO_{2005}TEQ L^{-1}. In this study the quantity of of twelve dioxin-like PCBs accounts for 12% of total 28 PCBs. Non ortho-PCBs (CB-77, CB-81, CB-126 and CB-169) were higher dl-PCBs and contributed more than 99% of total TEQ of ∑dl-PCBs on the other hand the TEQ of mono ortho PCBs (CB-105, CB-114, CB-118, CB-123, CB-156, CB-157, CB-167 and CB-189) were <1 at all the locations. CB-126 and CB-169 were the dominant dl-PCBs with the contribution of 99% to total dl-PCBs.

The PCBs contamination levels in surface waters from Delhi NCR were compared with guideline values stipulated by national oceanic and atmospheric administration (NOAA 2004) as CMC (Criteria Maximum Concentration) and CCC (Criteria Continuous Concentration). CMC is the highest level for 1h average exposure not to be exceeded more than once every three years and is synonymous with “acute” and CCC is the highest level for a 4 day average exposure not to be exceeded more than once every three years and is synonymous with “chronic”. The U.S. EPA has established a maximum contaminant level (MCL) (500 ng/L) for total PCBs in drinking water (FR 1991; DHS 2008). The Government of British Columbia recommended the 500 ng/L maximum concentrations of PCBs for irrigation water (BC 1992). The observed concentration of PCBs in surface waters from different sources in NCR Delhi were lower than CMC value of 2000 ng/L and MCL of USEPA, and British Columbia however higher than CCC value of 14 ng/L indicates the chronic toxicity to the users.

The higher concentration of PCBs was detected in samples those receives wastewater directly. This indicates that PCB has been used in industries and the chemicals have found their way to the environment. Emissions from coal combustion and industrial waste incineration sources contributed non ortho PCBs and do not solely from commercial PCB mixtures (Chi *et al*. 2007). Contamination of PCBs in Indian environment is restricted to transformer oil rather than technical mixture that used for industries and electrical appliances. It seems likely that the main source of PCBs in NCR Delhi India were from open biomass burning and depositions of emissions from wood processing paint and dying chemicals and from electrical and electronic waste recycling units. These PCB sources also include off gassing from closed system such as older equipments (e.g. transformers that contain large quantities of PCB fluids) and PVC (polyvinylchloride) manufacture.

4. Conclusion

This study has provided data on the levels of PCBs in surface water of different sources from National Capital Region (NCR) Delhi India. The congeners of PCBs containing 3-5 chlorines accounted the most part of PCBs in this study area. The tetra-PCB dominated the PCB homologs in average in NCR surface waters. The quantity of of twelve dioxin-like PCBs accounts for 12% of total 28 PCBs. The average concentrations were lower than guideline values; however PCBs levels at some locations were higher than guidelines and Criteria Continuous Concentration (CCC) which indicates possible chronic toxicological effects on continuous contact. Further work is needed to determine the bioavailability and bioaccumulation through tissues of aquatic biota (for example fishes) to determine the levels of contaminants on these living bodies. Then one could assess the risk of these contaminants on the ecosystems and human health more thoroughly.

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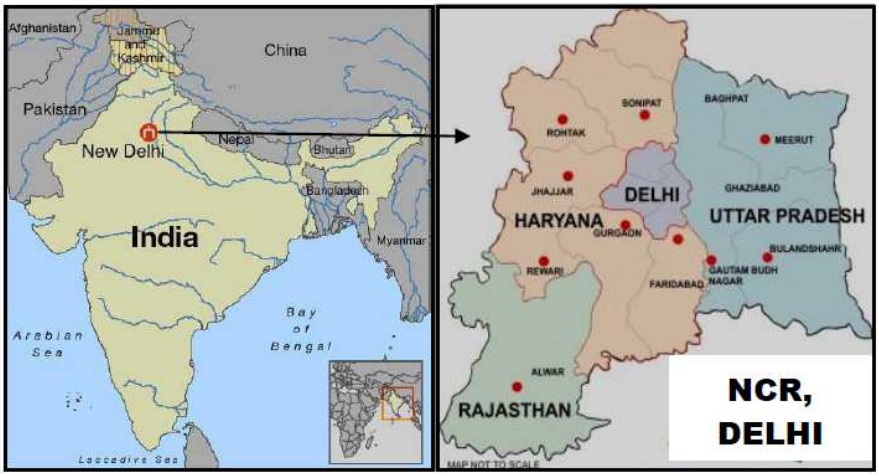

Figure 1: Map showing study area National Capital region (NCR) Delhi India

Table 1: PCBs and dl-PCBs congener concentrations in surface waters from NCR Delhi India

PCBs (ng/L)				**dl-PCBs (ng/L)**			
Congener	Range	Mean±SE	%	Congener No	Range	Mean±SE	%
PCB-18	<1-314	44±7	15	***Non ortho-PCB***			
PCB-37	<1-171	24±5	8	dl-PCB-77	<1-28	8±1	19
PCB-44	<1-594	85±12	29	dl-PCB-81	<1-24	6±1	14
PCB-49	<1-132	12±4	4	dl-PCB-126	<1-22	3±1	7
PCB-52	<1-90	9±2	3	dl-PCB-169	<1-11	<1	<1
PCB-70	<1-	73±35	25	***Mono ortho-PCB***			
PCB-74	<1-50	2±1	<1	dl-PCB-105	<1-14	<1	<1
PCB-119	<1-39	7±1	2	dl-PCB-114	<1-42	6±1	14
PCB-128	<1-34	3±1	1	dl-PCB-118	<1-18	2±1	5
PCB-138	<1-23	3±1	1	dl-PCB-123	<1-41	8±1	20
PCB-151	<1-61	7±1	2	dl-PCB-156	<1-16	3±1	6
PCB-168	<1-19	2±1	1	dl-PCB-157	<1-35	2±1	3
PCB-170	<1-21	2±1	1	dl-PCB-167	<1-19	3±1	7
PCB-177	<1-74	10±2	3	dl-PCB-189	<1	<1	<1
PCB-187	<1-53	5±1	1	***∑dl-PCBs***	***<1-146***	***40±4***	***100***
PCB-207	<1-57	6±1	2		-		
∑PCBs	***12-***	***293±41***	***100***				

∑PCBs+∑dl-PCBs			
Range	**Mean**	**Median**	**SE**
14-1768	332	275	42

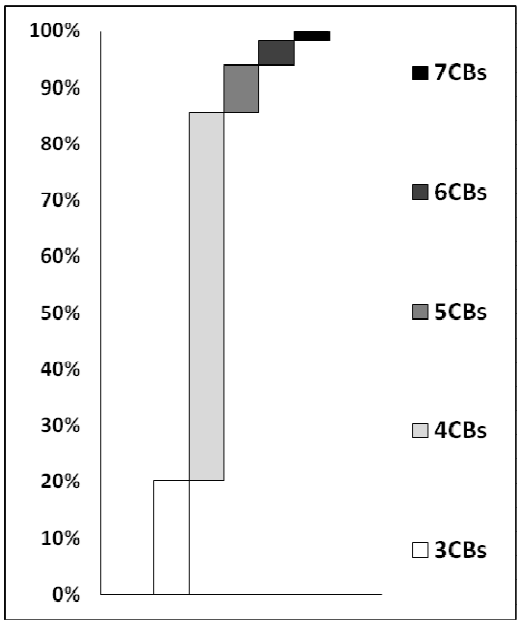

Figure 2: Percent homolog of PCBs (tri to hepta chlorinated) in surface waters of NCR Delhi India

Table 2: Total PCBs and dl-PCBs concentrations (ng/L) in surface waters from NCR Delhi India

Water System (N)*	PCBs	Range	Mean	Median	SE**
Yamuna River (8)	PCBs	92-477	321	335	46
	dl-PCBs	<1-77	43	48	9
	∑PCBs	92-554	364	380	52
Hindon River (4)	PCBs	30-1013	459	397	205
	dl-PCBs	<1-36	18	18	9
	∑PCBs	30-1048	477	415	212
Lake (12)	PCBs	13-917	189	83	73
	dl-PCBs	<1-102	21	4	9
	∑PCBs	15-2708	417	97	222
Ponds (12)	PCBs	39-513	228	217	39
	dl-PCBs	3-146	41	31	11
	∑PCBs	47-537	269	251	42
Canals (23)	PCBs	12-1969	356	239	102
	dl-PCBs	<1-106	45	40	7
	∑PCBs	14-1969	400	288	104
Drains (9)	PCBs	59-430	246	262	44
	dl-PCBs	13-113	52	43	11
	∑PCBs	104-503	298	275	50

*N=number of samples, **SE=standard error (SD/√n)

Table 3: PCBs group homolog (tri to hepta chlorinated) in surface waters (ng/L) of NCR Delhi India

Concentration	Tri-CB	Tetra-CB	Penta-CB	Hexa-CB	Hepta-CB
Mean	71	228	30	16	5
Median	47	142	21	10	2
Range	<1-317	8-2054	<1-112	<1-98	<1-53
% ∑PCBs	20	65	9	4	1

Table 4: DL-PCB congener TEQs (pg WHO_{2005}-TEQ/L) in surface waters from NCR Delhi India

dl-PCB congener	Yamuna River (8)	Hindon River (4)	Lakes (12)	Ponds (12)	Canals (23)	Drains (9)	All samples (67)
dl-PCB-77	<1-2.1 (1)	(<1)	<1-1.8 (<1)	<1-1.9 (1)	<1-2.8 (1)	<1-1.7 (<1)	0-2.8 (<1)
dl-PCB-81	<1-4 (1.6±0.5)*	<1-3.2 (<1)	<1-2.6 (<1)	<1-7 (1.5±0.6)	<1-7 (2.2±0.5)	<1-6 (2.3±0.8)	<1-7.1 (1.7±0.2)
dl-PCB-126	<1-987 (266±157)	<1-418 (138±99)	<1-749 (100±63)	<1-2225 (266±182)	<1-990 (281±66)	<1-1217 (452±164)	<1-2225 (264±51)
dl-PCB-169	<1-83 (27±11)	<1	<1-57 (15±6)	<1-80 (17±8)	<1-327 (25±14)	<1-154 (46±17)	<1-327 (23.6±5.9)
dl-PCB-105	<1	<1	<1	<1	<1	<1	<1
dl-PCB-114	<1	<1	<1	<1	<1	<1	<1
dl-PCB-118	<1	<1	<1	<1	<1	<1	<1
dl-PCB-123	<1	<1	<1	<1	<1	<1	<1
dl-PCB-156	<1	<1	<1	<1	<1	<1	<1
dl-PCB-157	<1	<1	<1	<1	<1	<1	<1
dl-PCB-167	<1	<1	<1	<1	<1	<1	<1
dl-PCB-189	<1	<1	<1	<1	<1	<1	<1
∑dl-PCBs	**<1-1038 (297±161)**	**<1-419 (139±99)**	**<1-812 (117±67)**	**1-2314 (286±188)**	**<1-994 (317±66)**	**1.2-1379 (502±178)**	**<1-2314 (290±53)**

*mean±SE in parenthesis