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Dioxin-Like Polychlorinated Biphenyls in River Sediments

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ABSTRACT

Dioxin like polychlorinated biphenyls (dl-PCBs) was investigated in sediments from Yamuna River in Delhi, India. The concentration of ∑dl-PCBs ranges between 0.04-5.59 with an average of 1.28±0.16 ng g^{-1}. Among the 12 dl-PCB congeners the congener no PCB-118 and PCB-114 were the dominant congeners and accounted for >48% and >24%, respectively. Toxic equivalency in term of toxic equivalent quotient (TEQ) of ∑dl-PCBs was calculated and presented. The TEQ in this study was ranged between <0.01-115.23 pg WHO_{2005}-TEQ g^{-1} with an average of 14.42±2.20 pg WHO_{2005}-TEQ g^{-1}. CB-126 and CB-169 congeners represent the higher TEQ values which both had the high toxic potency. Concentration of dl-PCBs does not exceed sediment quality guidelines.

Keywords: Persistent Organic Pollutants (POPs), PCBs, dioxin-like, Sediment.

INTRODUCTION

Polychlorinated biphenyls (PCBs) 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 [1]. These are persistent organic pollutants (POPs) which are resistance to chemical physical and biological degradation and being ubiquitously found in all environments of the earth [2]. PCBs are long range atmospheric transport (LRAT) pollutants and have been transported world-wide affecting regions far from their original sources such as the Arctic [3,4] and Antarctic [5] and high altitude regions of Mt Everest [6]. Coplanar or dioxin-like PCBs are formed unintentionally in the same way as polychlorinated dibenzo-*p*-dioxins/furans (PCDDs/Fs) [7,8]. Primary sources of dioxin like compounds are chemical- and petrochemical processing plants, ferrous and non-ferrous metal smelting operations, paper and pulp industries, cement production and smaller non-point sources include domestic burning of wood and open burning as well as by natural processes such as vegetation fires [9].

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 [10]. The International Agency for Research on Cancer has determined that PCBs are probably carcinogenic to humans (Group 2A). Their physico-chemical characteristics, which include hydrophobicity and resistance to degradation, make these chemicals ultimately to accumulate in soils, sediments and biota [11]. In the aquatic environment PCBs are bio-concentrated and transferred in the food chains and may return to humans with the aquatic food [12,13].

As a party to the Stockholm Convention, India is obligated to abide by the objectives of the treaty and is encouraged to support research on POPs. In May 2004, Stockholm Convention on POPs entered into force with the intention of reducing, and ultimately eliminating these pollutants. In India, some studies have been conducted into distributions of POPs, such as intentionally released insecticide, little known about the unintentionally released of other POPs, such as PCBs and dioxins. Earlier studies on various environmental matrices in India revealed PCBs contaminations in air, water, sediment [14-22], biota and humans [23,24]. In continuation of support research on POPs, this study was aimed to evaluate persistent dioxin like polychlorinated biphenyls (dl-PCBs) concentration in sediments from Yamuna River in Delhi stretch.

MATERIALS AND METHODS

Description of study area and Sampling

The River Yamuna, a major tributary of River Ganges, originates from the Yamunotri glacier of the lower Himalayas. The river flows through Himachal Pradesh, Uttrakhand and Haryana states, and enters Delhi near Palla village after traversing a route of about 224 Km 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 on the of River Yamuna. The River is tapped at Wazirabad through a barrage for drinking water supply to Delhi and accounts for more than 70% of overall water supply. Generally, no water is allowed to flow beyond Wazirabad barrage in dry season, as the available water is not adequate to fulfill the demand of water supply of Delhi. Whatever water flows in the downstream of Wazirabad barrage is the untreated or partially treated domestic and industrial wastewater contributed through several drains. Delhi alone generates 1,900 million liter per day (MLD) of sewage, against an installed wastewater treatment capacity of 1,270 MLD. Thus 630 MLD of untreated and partially treated sewage enters the river every day. After 22 Km downstream of Wazirabad barrage there is Okhla barrage and water is not allowed to flow through barrage during dry season. The total catchment area of Yamuna River in Delhi stretch is about 1485 Km^2.

Samples were collected from 60 locations on the bank of Yamuna River in Delhi extending between Palla, Wazirabad and Okhla (**Figure 1**). After sample collection pebbles and wood sticks removed and manually mixed thoroughly. Sub-samples of the sediment were subsequently taken and stored in labelled wide mouth amber glass bottles and transported ice-preserved to the laboratory and stored at -20^{0}C till further extraction.

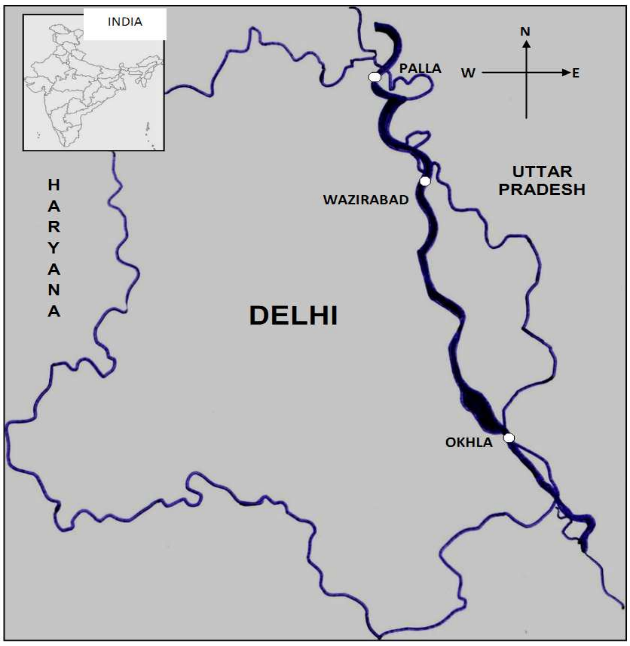

Figure 1: Map showing study stretch of Yamuna River in Delhi

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. Certified reference standards of PCBs were purchased from Dr. Ehrenstorfer (GmbH, Augsburg, Germany).

Sample Extraction

The extraction of pollutants was carried out in Soxhlet apparatus. 10-15 g wet sample was homogenized with sodium sulphate and the mixture was transferred into a clean cellulose extraction thimble and inserted into a Soxhlet assembly and extracted using dichloromethane: acetone (1:1 v/v) as extraction solvent. A 150 ml of extraction solvent was added and the whole assembly was heated for 16 h. After extraction the extract was evaporated to near dryness under reduced pressure in a 40 ^{0}C water bath using Rotary Vacuum evaporator (Eyela, Japan). 20 ml hexane was added to the concentrated extract and evaporated to a small volume (about 1 ml).

Chromatographic column cleanup

The multilayered silica gel column chromatography was performed to separate target analytes from other interfering organic and polar species. Briefly multilayered silica gel column (300 mm x 30 mm) was packed from bottom to up with 2.5 g silica gel, 4.0 g silver nitrate silica gel, 2.5 silica gel, 4.0 basic silica gel, 2.5 g silica gel, 12.0 g acid silica and 5.0 g anhydrous sodium sulphate. The column was pre-rinsed with 50 ml n-hexane before sample was loaded. The concentrated extracts and three 2 ml portions of hexane from rinsing the sample flask were transferred to top of the chromatography column. The elution of analytes was subsequently carried out using 170 ml hexane and concentrated to 2.0 ml. The eluted extract was concentrated using Rotatory Vacuum evaporator and Turbo Vap (Caliper, USA) under gentle stream of pure nitrogen using 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.

Instrumental analysis

A total of 12 dl-PCB congeners (IUPAC No -77, -81, -105, -114, -118, -123, -126, -156, -157, -167, -169, and -189) were analysed and quantified. 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 3^0C 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}.

Analytical quality control

Certified reference standards were used for the instrument calibration and quantification of PCBs. The target analytes were identified in the sample extract by comparing the 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 0.99, and matrix spike recovery 100±20%. Each sample was analysed in duplicate and the average was used in calculations. Moisture content was determined to report data on dry weight basis. 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.

The dioxin-like PCBs (dl-PCBs) are assigned with the toxic equivalent factors (TEFs) based on the relative toxicity with 2, 3, 7, 8-tetrachloro dibenzo-*p*-dioxin (TCDD) [10]. Toxic equivalent quantities (TEQ) of dl-PCBs were calculated using WHO toxicity equivalent factors (TEFs). The results of the analysis are reported in ng g^{-1} and pg WHO_{2005}-TEQ g^{-1}.

RESULTS AND DISCUSSION

Concentration of dl-PCBs

Table 1: Concentration of dl- PCB congeners (ng g-1) in Yamuna River bank sediments

Congener	Concentration				
	Range	Mean	SD	SE	%
Non ortho-PCBs					
dl-PCB-77	<0.01-0.22	0.08	0.05	0.01	2.60
dl-PCB-81	<0.01-0.40	0.15	0.09	0.01	4.17
dl-PCB-126	<0.01-1.15	0.15	0.17	0.02	11.11
dl-PCB-169	<0.01-0.04	0.02	0.01	<0.01	0.36
Mono ortho-PCBs					
dl-PCB-105	<0.01				
dl-PCB-114	<0.01-1.19	0.41	0.041	0.05	24.24
dl-PCB-118	<0.01-2.92	0.72	0.64	0.08	48.78
dl-PCB-123	<0.01				
dl-PCB-156	<0.01-0.14	0.09	0.03	<0.01	1.42
dl-PCB-157	<0.01-0.27	0.19	0.12	0.01	0.75
dl-PCB-167	<0.01-0.03	0.01	0.01	<0.01	0.21
dl-PCB-189	<0.01-0.44	0.09	0.08	0.01	6.35
∑dl-PCBs	***0.04-5.59***	***1.28***	***1.22***	***0.16***	***100***

*Note: <0.01=below detection limit,*standard error=SD/√n*

The concentrations of dl-PCBs in sediments from Yamuna River in Delhi are presented in **Table 1**. The observed concentration of ∑PCBs was ranged between 0.04-5.59 ng g^{-1} (dw) with the mean of 1.28±0.16 ng g^{-1}. Mono ortho PCBs were higher in concentration than non ortho-PCBs and accounted for more than 70% to ∑dl-PCBs in Yamuna River sediments. On the course of River in Delhi stretch, concentration of total dl-PCBs with an average significantly increases from up/stream (Palla) to down/stream (Before reaching Okhla) (**Figure 2**). Recent studies reported significant levels of PCBs in different environmental matrices from Delhi and adjoining areas [16-22]. Similar to this study, other studies worldwide, on PCBs distribution have been reported in the literature from Songhua River, China (ranged from 0.26 to 9.7 ng g^{-1}, with an average of 1.9 ng g^{-1}) [25]; Ghar El Melh lagoon, Tunisia (ND-3.99 ng g^{-1}) [26]. However, other studies on PCBs have reported the higher concentrations than our study in tropical sediments from Bahlui River, Romania (3-26 ng g^{-1}) [27]; Tam Giang Lagoon, Vietnam (2.03–24.7 ng g^{-1}) [28]; Minjiang River Estuary, Southeast China (15.14–57.93 ng g^{-1}) [29]; Hanoi, Vietnam (1.3 to 384 ng g^{-1}) [30]; Wuhan, Central China (0.90-46.14 ng g^{-1}) [31] and Lianjiang River, in Heping town of Guiyu, China (4.7-743 ng g^{-1}) [32].

Polychlorinated biphenyls (PCBs), which have been widely used in industrial applications, may also be present in the electronic waste stream. India is growing at an exponential rate in terms of electronic waste (e-waste), generating approximately 150,000 t/year, much of which is stockpiled or poorly managed. Maximum amount of e-waste generated in the country ends up at New Delhi for recycling purpose though in an informal manner which can be one possible source for PCBs. A statistically significant ($p<0.05$) positive linear correlation has been observed between the amount of e-waste generated in 2005 and the PCB concentration in the atmosphere of Indian cities [33]. Other possible sources are from open biomass burning which is common in agricultural field after crop harvesting, and depositions of emissions from wood processing, paint and dying, chemicals and transformer manufacturing units. These PCBs 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.

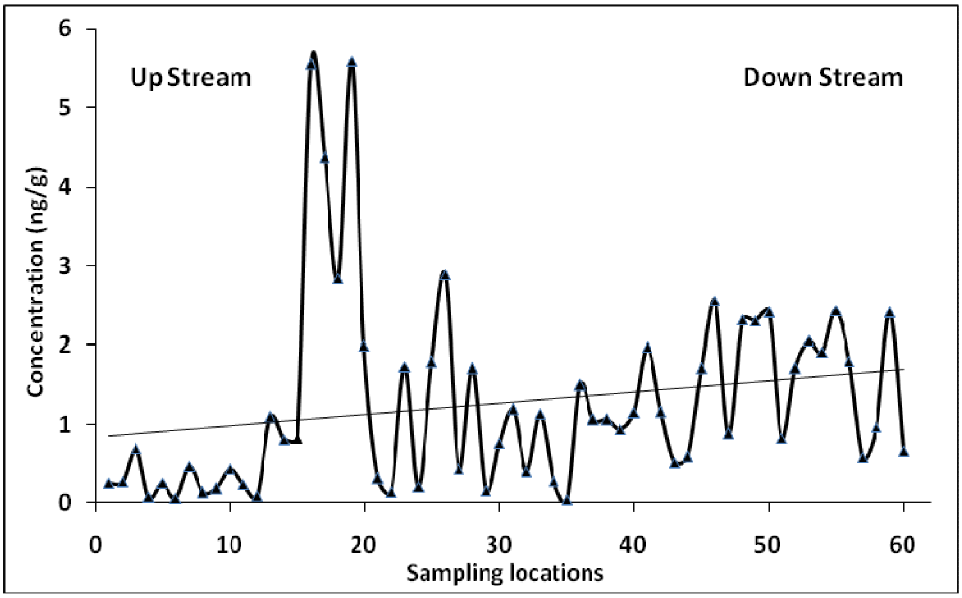

Figure 2: **Distribution pattern of ∑dl-PCBs in Yamuna River bank sediments from different sampling locations (up stream to down stream).**

Congener Profile and Group Homolog

The individual PCB congener profiles and their group homolog in Yamuna River sediments are illustrated in **Figure 3 and Figure 4**, respectively. Among the 12 dl-PCB congeners the congener no PCB-118 was the dominant congener, followed by PCB-114, PCB-126, PCB-189, PCB-81, PCB-77, PCB-156 and PCB-157, however concentration of other congeners were low. PCBs are not used as single compounds but as technical mixtures. 70% of PCBs produced globally were tri-, tetra-, and pentachlorinated biphenyls [34].

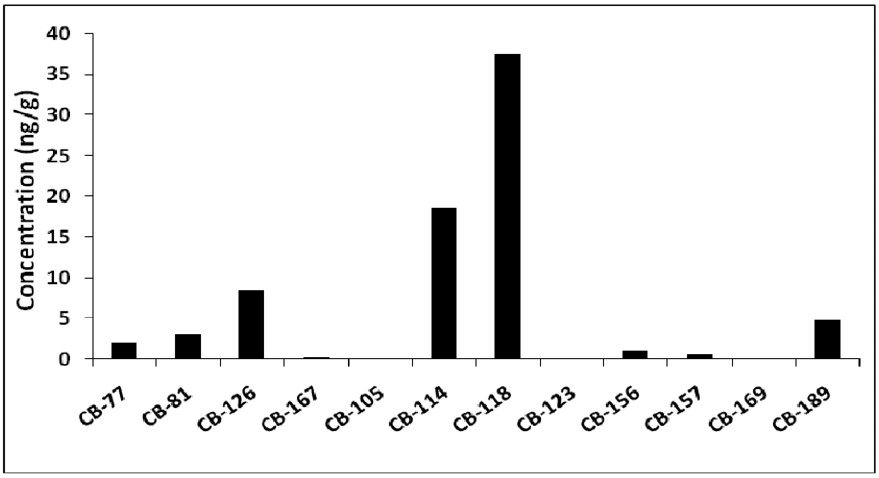

Figure 3: **Total concentration profile of dl-PCBs congeners in Yamuna River bank sediments.**

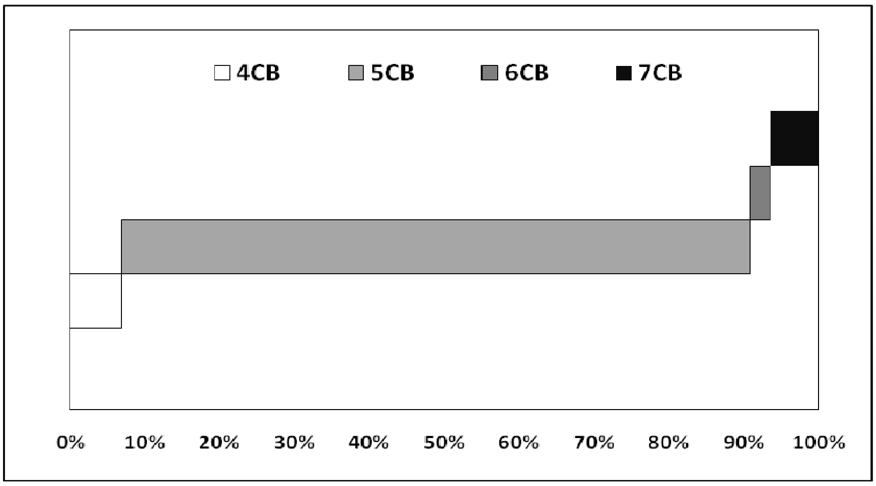

Figure 4: Group homolog of dl-PCBs in Yamuna River bank sediments.

The group homolog of dl-PCBs in Yamuna River sediments was primarily dominated by penta chlorinated biphenyls. Penta-CB homolog was the main contributors to the total dl-PCB homolog in average, and accounted for more than 84% (**Figure 4**). The lighter-weighted molecular PCBs (LWM-PCBs) were lower than those higher-molecular weight PCBs (HMW-PCBs) (penta to hepta-CBs). It is reported that LMW-PCBs were primarily used in power capacitors and transformers, while HMW-PCBs were mainly used as an additive [25]. This indicates that PCBs used in as additives found their way to the environment of Yamuna River in Delhi.

Toxic Equivalency of dl-PCBs

Table 3: Toxic equivalency (TEQ) of dioxin-like PCB congener (pg WHO_{2005}-TEQ g^{-1}) in Yamuna River bank sediments

Congener	TEF	Toxic Equivalency Quotients (TEQs)				
		range	Mean	SD	SE	%
Non ortho-PCBs						
dl-PCB-77	0.0001	<0.01-0.02	<0.01	0.01	<0.01	0.02
dl-PCB-81	0.0003	<0.01-0.12	0.02	0.03	<0.01	0.11
dl-PCB-126	0.1	<0.01-115.0	14.23	17.08	2.20	98.68
dl-PCB-169	0.03	<0.01-1.20	0.14	0.29	0.04	0.97
Mono ortho-PCBs						
dl-PCB-105	0.0003	<0.01	<0.01	<0.01	<0.01	<0.01
dl-PCB-114	0.0003	<0.01-0.05	0.01	0.01	<0.01	0.06
dl-PCB-118	0.0003	<0.01-0.09	0.02	0.02	<0.01	0.13
dl-PCB-123	0.0003	<0.01	<0.01	<0.01	<0.01	<0.01
dl-PCB-156	0.0003	<0.01	<0.01	<0.01	<0.01	<0.01
dl-PCB-157	0.0003	<0.01	<0.01	<0.01	<0.01	<0.01
dl-PCB-167	0.0003	<0.01	<0.01	<0.01	<0.01	<0.01
dl-PCB-189	0.0003	<0.01	<0.01	<0.01	<0.01	<0.01
∑dl-PCBs	-	***<0.01-115.23***	***14.42***	***17.08***	***2.20***	***100***

Note: TEF= WHO_{2005} -Toxic Equivalency Factor

Dioxin like PCBs have toxic responses similar to those caused by 2, 3, 7, 8-tetra-chlorodibenzo-*p*-dioxin (TCDD) the most potent congener within the dioxins groups of compounds [35]. 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 health. The concentrations of dioxin-like PCB congener can be converted into 2, 3, 7, 8-TCDD substituted TEQ concentrations. TEQ concentrations of dl-PCBs with established dioxin-like activity in sediments from Yamuna River was calculated by multiplying the concentration of each dioxin-like PCB congener by its 2, 3, 7, 8-TCDD substituted TEF values (Toxic Equivalency Factors) for human and mammals. The toxicity equivalency (TEQ) for 12 dl-PCBs was presented in **Table 3**. The TEQ of ∑dl-PCBs in this study was ranged between <0.01-115.23 pg WHO_{2005}-TEQ g^{-1} with an average of 14.42±2.20 pg WHO_{2005}-TEQ g^{-1}. The TEQ of non ortho-PCBs (CB-77, CB-81, CB-126 and CB-169) were higher and contributed >99% for ∑TEQ, while 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 for all the samples. CB-126 and CB-169 congeners represent the higher TEQ values which both had the high toxic potency (toxic equivalency factor proposed WHO-TEF=0.1 and 0.03 respectively) thus significantly increasing the ∑dl-PCBs with the contribution of 99% for ∑TEQ. Emissions from coal combustion and industrial waste incineration sources contributed non ortho PCBs and do not solely from commercial PCB mixtures [36].

Eco-toxicological risk assessment

Table 4: **Available sediments quality guidelines for polychlorinated biphenyls (PCBs).**

Guidelines	Concentration (ng g^{-1} dw)				Reference
Threshold Effect Concentration (TEC)	TEL	LEL	MET	ERL	NOAA, 1999; MacDonald et al. 2000
	34.1	70	200	50	
Probable Effect Concentration (PEC)	PEL	SEL	TET	ERM	NOAA, 1999; MacDonald et al. 2000
	277	5300	1000	4000	
CBSQGs	TEC	MEC	PEC	-	WDNR, 2003
	60	368	676	-	

TEL=Threshold effect level, LEL=Lowest effect level, MET=Minimum effect threshold, ERL=Effect level low, PEL=Probable effects level, SEL=Severe effects level, TET=Toxic effect threshold, ERM=Effect range median

Effects on the organisms are usually considered to be an early warning indicator of potential human health impacts. In India, no environmental standards have been established for PCBs in marine or riverine sediments. Therefore, ecotoxicological effects of these pollutants in this study area were roughly evaluated by applying published sediment quality guidelines such as threshold and probable effect level and consensus based sediment quality guidelines (CBSQGs) from National Oceanography and Atmospheric Administration [37] and Wisconsin [38,39]. **Table 4** shows the available sediment quality guideline values from different environmental agencies. Japan established the environmental standards of dioxins for sediment as 150 pg-TEQ g^{-1} [40]. However, Canadian government defined as 21.5 pg TEQ g^{-1} [41]. Concentration of ∑dl-PCBs and their individual congeners in Yamuna River sediment (**Table 1**) were lower than the guideline values and indicate no adverse effects to the biota.

CONCLUSION

As a party to the Stockholm Convention, India is obligated to abide by the objectives of the treaty, and is encouraged to support research on POPs. Distribution of dl-PCBs and their potential ecotoxicological risk were studied in sediments from Yamuna River in Delhi. On comparing with sediment quality guidelines, study indicates levels of dl-PCBs in sediments were lower than guideline values.

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