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Polychlorinated Biphenyls Contamination of Sludge in India

Khageshwar Singh Patel[1*], Shobhana Ramteke[1], Bharat Lal Sahu[1], Yogita Nayak[1], Saroj Sharma[2], Chin-Chang Hung[3]

[1]School of Studies in Chemistry/Environmental Science, Pt. Ravishankar Shukla University, Raipur, India
[2]Department of Chemistry, Devi Rathi Mahila Mahavidhaya, Rajnandagaon, India
[3]Institute of Marine Environmental Chemistry and Ecology, National Taiwan Ocean University, Taiwan
Email: *patelks_55@hotmail.com

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Abstract

Polychlorinated Biphenyls (PCBs) exist in all compartments of the ecosystem due to wide industrial applications and production during burning processes. The PCBs congeners were quantified in the industrial, municipal and agricultural wastes of the most industrialized area of the country, Raipur city, India. The concentration of ΣPCBs in the sludge, sewage and agriculture wastes (n = 4) was ranged from 497 - 800 µg/kg with mean value of 634 ± 146 µg/kg. The PCBs congeners detected in the waste materials were ranged from 56 - 85 with mean value of 75 ± 13. The highest number of congeners was quantified in the sludge materials due to the existence of the heavy PCBs. The PCBs congeners were found to decrease vertically due to the adsorption by the geo-media. The distribution, sources and toxicities of PCBs congeners in the waste materials are discussed.

Keywords

Polychlorinated Biphenyls, Sewage, Sludge, Contamination

1. Introduction

The persistent organic pollutants (POPs): DDT, aldrin, dieldrin, endrin, chlordane, heptachlor, hexachlorobenzene, mirex, toxaphene, polychlorinated biphenyls (PCBs), dioxins, furans, etc. are toxic chemicals that adversely affect human health and the environment around the world [1]. They frequently occur in the Asian environment due to their large use in industrial and agricultural sectors as well as production during burning processes [2]. Polychlorinated biphenyls (PCBs) are a class of organic compounds having a basic chemical

*Corresponding author.

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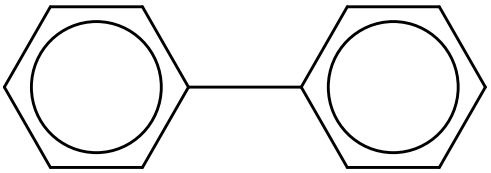
structure "I", which include two benzene (biphenyl) rings and between 1 and 10 chlorine atoms substituted on each of the benzene molecule [3]. PCBs have numerous industrial and other applications as dielectric fluids and insecticides [4]. PCBs have been reported to cause many adverse health effects, such as immune deficiency, nervous system alteration, endocrine disruption, and gastrointestinal system bleeding and liver damage [5]. Many sludge were reported to be contaminated with polychlorinated biphenyls at elevated levels, 6 - 2500 µg/kg [6]-[15]. Limited PCB congeners in soils of industrial cities of the country were reported by Kumar *et al*. [16] [17]. Therefore, in this work, the contamination, distribution and sources of PCBs congeners (>50) in the agricultural, industrial and municipal wastes of the most industrialized area of central India: Raipur, Chhattisgarh, India are described.



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2. Methods and Materials

2.1. Selection of Studies Areas

Iron and steel is one of the largest sectors of industries in the central India. Iron ore is smelted in blast furnace with coke and lime to produce pig iron and generates a huge amount of waste. The one of the Asia biggest steel plant (capacity of 4.8 MT iron/Yr) is running nearby Raipur city at Bhilai with subsequent dumping of the industrial effluents. Several coal based thermal power plants are operated for generation of electricity by dumping waste materials in the nearby area. The Rice is major agricultural crops in central region of the country which generate a huge amount of the biomass waste. Large quantity (≈300 million lit/day) of untreated sewage waste is discharged into the environment. Hence, these wastes were selected for monitoring of the PCBs congeners.

2.2. Sample Collection

Six samples *i.e*. sludge, sewage and burnt agricultural wastes were collected in month of May, 2009 using a stainless-steel scoop. The sampling locations (*i.e*. Bhilai Steel Plant dumping pond, Monet thermal power plant waste, Urla municipal waste and IGAU rice field agricultural waste) are shown in **Figure 1**. They were kept in a 250-mL glass bottle and dried at 30˚C in an oven for overnight. The samples were crushed into fine particles by mortar and sieved out the particles of mesh size <0.1 mm. The samples were stored in aluminum foil by freezing at −4˚C.

2.3. Analysis

The sediment (5.0 g) was extracted with 25-mL distilled water by subsequent decanting out the extract for the pH measurement. The CHNSO-IRMS Analyzer (SV Instruments Analytica Pvt. Ltd.) was used for analysis of the total carbon (TC) and black or elemental carbon (BC or EC). The soil sample (15 mg) was oxidized with O_2 at 1020˚C with constant helium flow by measuring the resulting CO_2 gas with a thermal conductivity detector. The H_3PO_4 (10 drops) treated soil sample was oxidized in a similar way for determination of BC and organic carbon (OC) content. The OC content was analyzed by titration method using $K_2Cr_2O_7$ as oxidant [18]. The carbonate carbon (CC) content in the soil was evaluated by subtracting the BC+OC to the TC content by using the following equation.

$$\mathrm{CC} = \mathrm{TC} - (\mathrm{BC} + \mathrm{OC})$$

The dried waste sample was extracted with mixed solvent: methylene chloride + methanol (1:1, v/v) by using a Dionex accelerated solvent extraction (ASE) system. The extract was evaporated, and re-extracted with solvent, methylene chloride by subsequent addition of the surrogate standards. The extract was purified by silica/alumina column chromatography to isolate the PCB fractions [19]. The quantitative analyses of PCBs were performed by HP 5890-GC (gas chromatography) and HP 5970-MS (mass spectrometer) in the SIM mode [20].

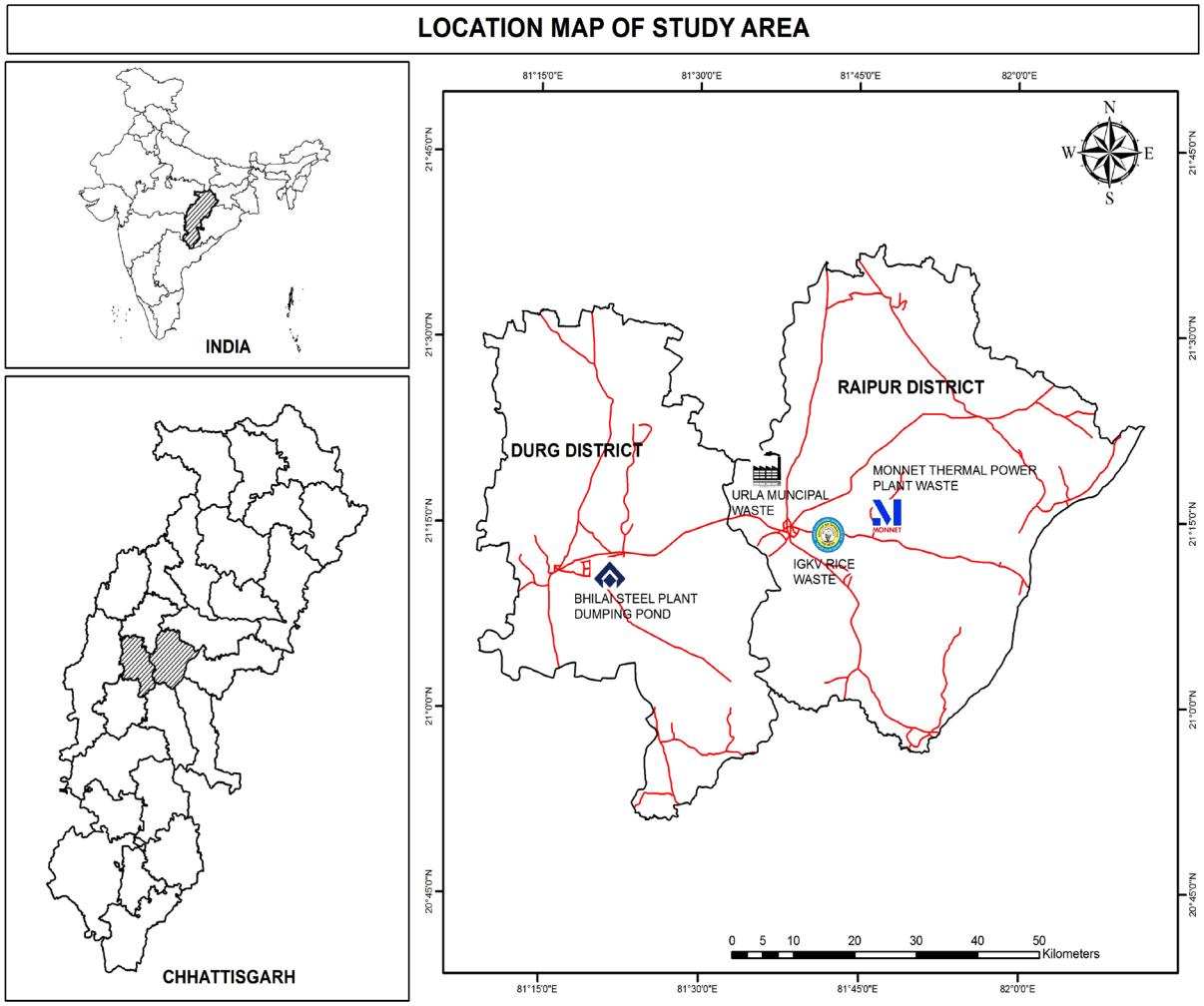

Figure 1. Representation of sampling locations.

3. Results and Discussion

PCBs are mixtures of 209 individual chlorinated compounds with chemical formula of $C_{12}H_{10-n}Cl_n$, where *n* ranges from 1 to 10. The chemical description of PCBs are summarized in **Table 1**.

3.1. pH of Extract

The pH value of the extract (n = 4) was ranged from 7.8 - 8.4 with mean value of 8.1 ± 0.2. All the extracts were found to be slightly alkaline in nature. The lowest pH value of the sludge sample was recorded. The pH value of the sludge extract was found to be increase slightly as the depth profile of the sludge was increased from 0 - 30 cm, **Table 2**.

3.2. Carbon Concentration

The carbon content of the waste materials are presented in **Table 2**. The concentration of BC, OC and CC (n = 4) was ranged from 7.5% - 9.0%, 0.03% - 0.05% and 0.11% - 0.19% with mean value of 8.3 ± 0.8, 0.04 ± 0.01 and 0.15% ± 0.04%, respectively. The BC had fair correlation with the OC and CC content (r = 0.93 - 0.97), indicating their origin from the same sources. The BC concentration was found to decrease with increasing depth profile from 0 to 30 cm unlikely to the OC and CC. The BC content in the waste materials of the Raipur area was found to be higher than other region of the World, probably due to higher coal burning in this region [21]-[23].

Table 1. Description of PCBs.

Compound	Congener number	Abbreviation	Molecular weight	Formulae
Monochlorobiphenyls	3	MCBs	189	$C_{12}H_9Cl$
Dichlorobiphenyls	12	DCBs	233	$C_{12}H_8Cl_2$
Trichlorobiphenyls	24	TCBs	258	$C_{12}H_7Cl_3$
Tetrachlorobiphenyls	42	TeCBs	292	$C_{12}H_6Cl_4$
Pentachlorobiphenyls	46	PeCBs	326	$C_{12}H_5Cl_5$
Hexachlorobiphenyls	42	HCBs	361	$C_{12}H_4Cl_6$
Heptachlorobiphenyls	24	HeCBs	395	$C_{12}H_3Cl_7$
Octachlorobiphenyls	12	OCBs	430	$C_{12}H_2Cl_8$
Nonachlorobiphenyls	3	NCBs	189	$C_{12}HCl_9$
Decachlorobiphenyls	1	DeCBs	233	$C_{12}Cl_{10}$

Table 2. Carbon content in waste materials, %.

Waste material	pH	BC	OC	CC
Sl1	8.4	7.5	0.03	0.12
Sl2	8.6	7.3	0.06	0.15
Sl3	9.1	7.1	0.09	0.18
TPPW	8.1	8.9	0.04	0.17
AW	7.8	9.0	0.05	0.19
MW	8.0	7.6	0.03	0.11

Sl1, Sl2 and Sl3 denote the Bhilai steel plant sludge collected at depth of 0 - 10, 10 - 20 and 20 - 30 cm; TPPW = Monate thermal power plant waste; AW = Agricultural waste; MW = Municipal (sewage) waste.

3.3. Total PCBs Concentration

The PCBs contents in the waste material is summarized in **Table 3**. The sum of total concentration of PCBs in 4 different types of waste materials (Sl1, TPPW, MW and AW) was ranged from 497 - 800 µg/kg with mean value of 634 ± 146 µg/kg. The highest concentration of the PCBs was observed in the AW sample due to the excessive input of the congener 3. The large fraction of the PCBs, 68% - 84% in all samples was contributed by the monochlorobiphenyls (MCBPs), **Figure 2**.

3.4. Spatial Distribution of Congeners

The observed congener frequency of the PCBs in the Sl, TPPW, MW and AW was found to be 85, 77, 81 and 56, respectively, **Figure 3**. The highest concentration of MCBPs was observed in the AW, may be due to chlorination of the biphenyls, **Figure 4(a)**. The highest concentration of DCBPs and TCBPs to HCBPs was marked in the TPPW and MW, Respectively, **Figure 4(b)** and **Figure 4(c)**.

3.5. Vertical Distribution of PCBs

The vertical distribution of ΣPCBs concentration and congeners are presented in **Table 3**. Both concentration and congener numbers of PCBs were found to decrease as the depth profile of the sludge was increased from 0 - 30 cm, may be due to their adsorption by the sludge matrix and biodegradation, **Figure 5** and **Figure 6**. At least 42% estimated congeners in the sludge was found to exist in the sludge, **Figure 5**. The congener frequency was increased up to TeCBPs, and thereafter, decreased slowly up to HeCBs, **Figure 5**.

In general, the concentration of MCBPs, DCBPs, TCBPs, PCBs, HCBPs and HeCBPs was found to decease as the depth profile of the sludge was increased from 0 - 30 cm, **Figure 6**. However, the reverse trend was seen

Table 3. Distribution of PCBs congeners in waste materials, µg/kg.

Type of PCBs	Congener	Sl1	Sl2	Sl3	TPPW	MW	AW
MCBPs	1	111.9	86.5	28.6	127.9	148.4	254.7
	3	374.3	188.6	305.3	226.5	69.7	414.4
DCBPs	4 + 10	7.6	5.8	3.9	7.6	0.0	0.0
	7 + 9	1.3	1.0	1.5	1.2	0.3	0.7
	6	3.8	2.1	0.2	3.6	2.3	3.4
	8 + 5	15.6	12.9	0.4	16.0	18.4	15.1
TCBPs	19	2.8	2.1	1.3	1.2	2.5	2.0
	18	1.8	2.4	1.6	2.1	5.7	1.8
	17	5.8	5.6	4.3	5.1	7.0	0.0
	24	0.0	0.0	0.0	0.0	1.5	0.0
	16 + 32	3.3	2.2	1.9	2.5	2.2	7.2
	29	1.4	1.1	1.9	1.2	0.1	2.0
	26	3.9	1.9	2.1	3.1	1.3	2.3
	25	1.1	1.9	1.5	1.7	1.0	3.0
	28 + 31	3.8	2.8	3.7	3.8	8.2	4.8
	22	4.2	3.0	2.3	3.7	1.9	3.1
	21 + 33 + 53	3.3	2.4	2.8	3.0	3.3	2.3
TeCBPs	51	2.9	2.2	2.2	2.6	0.0	0.0
	45	2.3	1.7	1.0	2.3	1.0	1.2
	46	1.6	2.3	0.0	1.4	2.1	2.4
	62	2.9	2.0	0.6	2.5	4.1	1.6
	49	2.4	1.8	0.0	2.3	8.1	5.1
	47 + 48	2.0	1.6	0.5	1.4	2.3	0.0
	44	0.8	0.6	0.5	2.4	2.0	1.3
	37 + 42	7.1	5.2	2.9	4.7	4.0	2.3
	41 + 64 + 71	4.6	11.6	5.9	7.2	3.2	23.1
	63	0.0	0.0	1.1	1.5	0.0	0.0
	74	2.3	2.6	2.6	2.7	3.2	2.6
	70.76	3.2	2.5	2.2	2.3	7.3	5.2
	66 + 95	2.7	2.5	1.7	2.0	4.0	3.6
	56 + 60	5.2	4.2	24.7	0.0	20.2	10.2
PCBPs	91	0.0	0.0	0.0	0.0	0.0	0.0
	100	2.0	1.6	1.2	1.7	1.7	3.9
	105	6.6	6.2	2.5	5.9	5.9	0.0
	89	4.2	0.0	0.0	1.7	14.4	0.0
	101	3.0	1.5	1.3	1.3	6.9	2.3
	99	2.4	1.2	0.8	0.7	2.1	0.7
	83	1.0	1.9	0.3	0.2	1.9	3.0
	97	1.0	0.6	0.1	0.1	5.9	0.3

Continued

	81 + 87	1.2	0.8	0.3	0.0	0.0	0.0
	85	0.9	0.6	0.3	0.3	1.1	0.0
	77 + 110	10.3	13.3	4.2	8.6	6.7	4.6
	82 + 151	4.7	5.6	0.0	0.0	0.0	0.0
	123 + 149	12.0	11.8	2.9	5.8	8.9	0.0
	118	4.9	6.4	1.7	3.5	3.0	0.0
HCBPs	134	14.7	15.1	0.0	0.0	6.3	0.0
	146	7.5	5.7	0.0	0.0	0.0	0.0
	136	6.3	5.9	1.2	2.4	4.8	0.0
	132	3.8	5.7	1.5	3.0	5.3	0.0
	135 + 144	3.3	2.4	0.9	3.8	4.2	0.0
	153	14.3	11.2	3.0	8.3	17.1	0.0
	157	2.2	0.0	2.2	0.0	1.7	0.0
	141	3.3	2.4	0.6	2.0	1.8	0.0
	137 + 130 + 176	2.2	2.4	0.0	2.2	3.1	0.0
	138 + 158	5.7	3.9	1.6	4.5	31.2	5.5
	163	6.6	5.9	3.5	7.5	0.0	0.0
	129 + 178	5.2	14.1	9.1	10.1	4.0	0.0
HeCBPs	183	2.8	0.0	0.0	0.0	5.4	0.0
	185	0.5	0.0	0.0	0.4	2.6	0.0
	177	1.5	0.0	0.0	0.0	3.2	0.0
	201	0.0	0.0	0.0	0.0	5.0	0.0
OCBPs	208 + 195	0.0	0.0	0.0	0.0	6.9	0.0
	194	0.0	1.4	0.0	0.0	0.8	4.1

MCBPs, DCBPs, TCBPs, TeCBPs, PCBPs, HCBPs, HeCBPs and OCBPs denotes the mono, di, tri, tetra, penta, hexa, hepta and octachloro-derivative of BPs.

with TeCBPs. The following four different scenarios for the PCBs depth profile distribution was observed. The concentration of large number of congeners was found to decrease when the depth profile was increased, may be due to strong adsorption by the sludge matrix (**Table 3**). The concentration of some congeners (*i.e*. 63, 56, 60 and 74) was found to increase as the depth profile was increased, may be due to less adsorption by the sludge matrix. However, the concentration of few congeners (*i.e*. 7, 9, 18, 21, 25, 29, 33, 41, 51, 53, 64, 71, 129 and 178) was unaffected with respect to increasing depth profile, may be due to being nonpolar in nature. Some heavy congeners *i.e*. 183, 185, 177 and 201 was observed to be appeared in the surface sludge.

3.6. Toxicities

The coplanar PCBs (*i.e*. 77, 126 and 169) having dioxin-like properties were reported to be the most toxic congeners [24]. Among them, the congener 77 existed in all samples, and its concentration was not found to be affected with respect to increasing depth profile from 0 to 30 cm. The PCBPs in all waste materials of the studied area was found to be several folds higher than the recommended value of 60 µg/kg soil [25].

3.7. Sources

The most common trade name of PCBs is Aroclor, being mixture of higher chlorinated biphenyls. The content of PCBs (*i.e*. MCBPs, DCBPs, TCBPs and TeCBPs) among themselves had good correlation (r = 0.97) in the

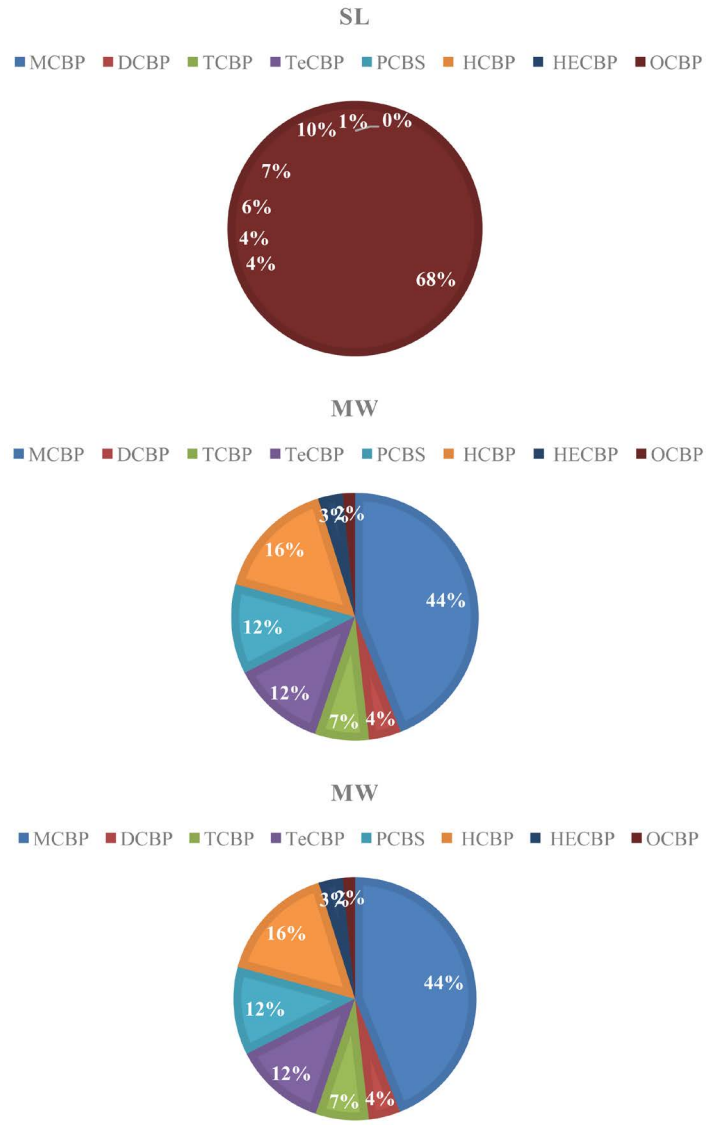

Figure 2. Relative concentration of MCBPs, DCBPs, TCBPs, TeCBPs, PCBPs, HCBPs, HeCBPs and OCBPs in the waste materials.

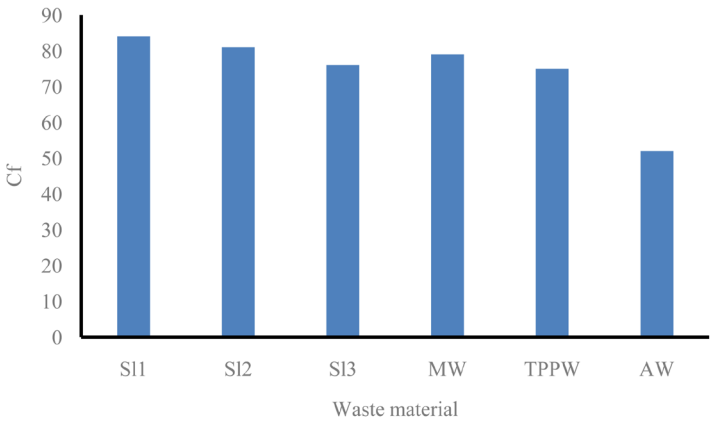

Figure 3. Representation of congener frequency in the waste material.

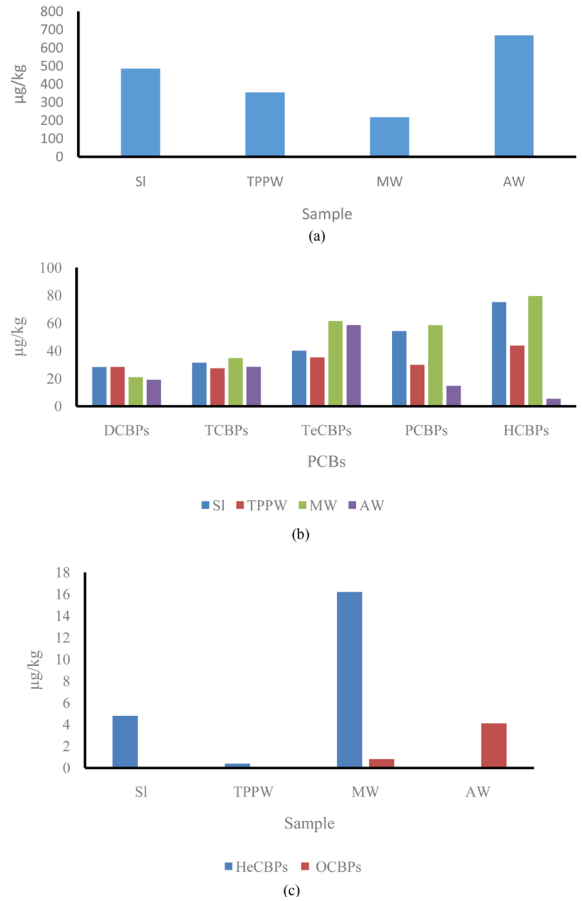

Figure 4. Representation of mono, di, tri, tetra, penta, hexa, hepta and octachloro derivatives of BPs.

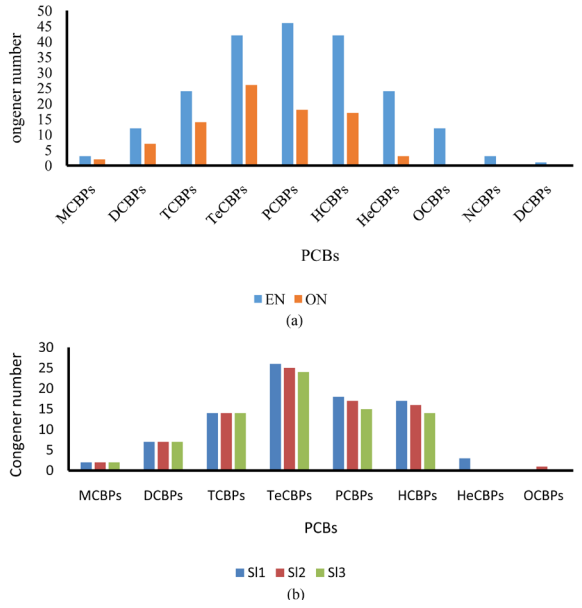

Figure 5. Congener distribution in the environmental samples, EN = Estimated congeners, ON = Observed congeners, Sl1 = 0 - 10 cm, Sl2 = 10 - 20 cm, Sl3 = 20 - 30 cm.

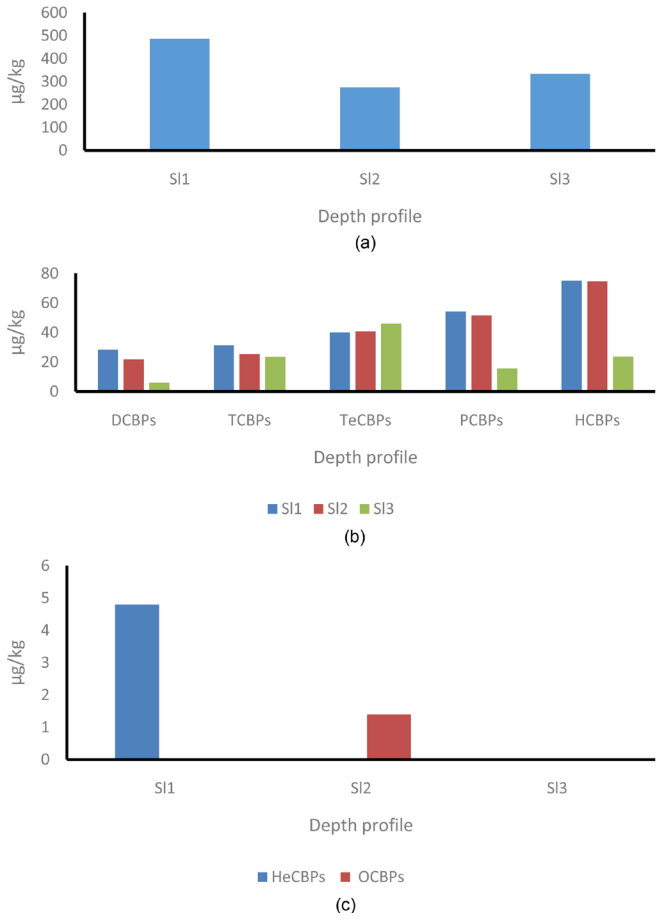

Figure 6. Depth profile studies of PCBs in the sludge, Sl1 = 0 - 10 cm, Sl2 = 10 - 20 cm, Sl3 = 20 - 30 cm.

sludge. However, the higher PCBs (*i.e.* PCBPs, HCBPs, HeCBPs and OCBPs) had fair correlation (r = 0.60), indicating origin from multiple sources. It means the lower PCBs could be generated by the chlorination processes of the BPs. The higher PCBs are released in the environments by leakage from the application sites.

$$C_{12}H_{10} + Cl_2 \rightarrow C_{12}H_9Cl + HCl$$

$$C_{12}H_9Cl + Cl_2 \rightarrow C_{12}H_8Cl_2 + HCl$$

$$C_{12}H_8Cl_2 + Cl_2 \rightarrow C_{12}H_7Cl_3 + HCl$$

$$C_{12}H_7Cl_3 + Cl_2 \rightarrow C_{12}H_6Cl_4 + HCl$$

4. Conclusion

Significantly higher concentrations of PCBs in the industrial and municipal waste materials of Raipur region of the country were observed. The major fraction of the PCBs was contributed by the congeners, 1 and 3. The PCBs concentration and congeners were decreased with respect to depth profile of the sludge. The origin of lower PCBs was expected by the chlorination of the biphenyl. However, origin of higher PCBs was assumed from leakage or evaporation from the application sites. The NCBPs and DCBPs were not detected in waste materials.

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