Distribution and Sources of Polychlorinated Biphenyls in Air, Dust, and Sediment from India

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Abstract: Persistent organic pollutants, such as polychlorinated biphenyls (PCBs), pose a serious risk for human health and the environment. In this study, PCBs contamination and sources of ambient air, road dust, and sediments in the most polluted city in India, Raipur has been measured over the period 2008–2015. The seasonal variations of particulate matter (PM), elemental carbon (EC), organic carbon (OC), and carbonate carbon (CC) were studied, and maximum concentrations were detected in the December–January period each year. Total PCBs concentrations in the ambient air (associated ~~to particulate matter~~), road dust, and sediments samples during 2008 were in the 186–645 pg m^{-3}, 102–537, and 241–538 ng g^{-1} range, respectively. 2-chlorobiphenyl (PCB-1) and 4-chlorobiphenyl (PCB-3) were the dominant chemical compounds identified. A substantial vertical migration of the PCBs in the sediment was observed. Concentration variations (spatial and temporal), correlations, and sources of PCB are discussed. In particular, an average increment rate of 6.2%, 4.9%, and 5.4% of PCBs concentration in the particulate matter (PM_{10}), road dust, and sediments respectively, was observed over the 2008–2015 period. The reported data points to India's low degree of accomplishment of the Stockholm Convention's requirement to phase out the use of PCBs in equipment by 2025 and ensure elimination of PCBs by 2028.

Author keywords: PCBs; Contamination; Air; Dust & sediment; Toxicity; Sources.

Introduction

Persistent organic pollutants (POPs) (i.e., aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, toxaphene, polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins (PCDD), and polychlorinated dibenzofurans (PCDF), plus the 16 so-called "new POPs" under the Stockholm Convention, are characterized by their high half-lives, low water solubility, and high lipid solubility (Harrad 2009). In particular, PCBs, a group of man-made oily liquids or solids, are widely used in electrical equipment, hydraulic fluids, heat transfer fluids, coolants, lubricants, and plasticizers in paint, paper, and plastics due to their low electrical conductivity and high resistance to heat and thermal degradation (Robertson and Hansen 2001). PCBs are a class of aromatic compounds ~~having~~ two benzene rings with a maximum of 10 substituted Cl-atoms, and are mixtures of up to 209 individual chlorinated compounds, known as congeners, whose chemistry differs from species to species (Hutzinger 1974). PCBs have been reported to cause many adverse effects both on wildlife and on human health, including immune deficiency, nervous system alteration, endocrine disruption, and gastrointestinal system bleeding and liver damage (ATSDAR 2014).

Contamination with PBCs has been reported in air, water, soil, dust, and sediment samples from all over the world (Anh et al. 2019; Biterna and Voutsa 2005; Chakraborty et al. 2013, 2016; Devi et al. 2014; Goel et al. 2016; Kim and Masunaga 2005; Kumar et al. 2011; Nasir et al. 2014; Sakin and Tasdemir 2016; Syed et al. 2013; Wang et al. 2013). In fact, at present, the United Nations Environment Program (UNEP) estimates that 83% of the total amount of PCBs in the world (*ca*. 14 million tons) still remains to be eliminated.

In the particular case of India, the Stockholm Convention was ratified in 2002 and entered into force in 2006. Preliminary investigations on PCBs contamination in sediments and sludge from the Raipur area ~~has~~ been previously reported (Patel et al. 2013, 2015). ~~Findings of the highest concentrations of the monochlorobiphenyl (MCBP) congeners were reported to date.~~ Nonetheless, as noted by the PCB Elimination Network (PEN), "data on elimination of PCB is often incomprehensive, outdated, and incomplete (United Nations Environmental Programme, UNITAR 2017)." Consequently, the aim of present work is to describe the distribution, variation, sources, and fate of PCBs congeners in different environmental samples (air, dust, and sediment) from the most industrialized area of central India, Raipur city (capital of Chhattisgarh state), to strengthen the analyses of India's PCBs situation, in line with the "country specific diagnoses" PEN key theme.

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Materials and Methods

Study Area

The capital ~~city~~ of Chhattisgarh state, Raipur (21°23′ N, 81°63′ E), with a population of *ca*. 2 million inhabitants, was selected for the proposed investigation due to its high degree of industrialization. Raipur city and the nearby region are home to many coal, power, steel, and aluminum industries. For instance, Asia's biggest steel plant is located in Bhilai, and Korba (known as the "power capital of India") is heavily polluted by thermal power plants (Jaiswal et al. 2019a, b).

Environmental Samples Collection

The sampling of the particulate matter was carried out for 13 months (February 2007 to January 2008) in Kota, a residential area (Fig. 1). For spatial variation studies, the sampling was carried out at three additional locations: Pt. Ravishankar Shukla University, Amapara, and Raipura in December 2008. These sampling sites were approximately 2 km distant from each other. The locations at Kota and Amapara ~~are located at~~ commercial and traffic sites. The distance of these sampling sites from the industrial area was approximately 2 km to the east. For temporal variation studies, Kota site was selected, and one PM_{10} sample was collected every January from 2009 to 2015.

A Partisol model 2300 sequential speciation air sampler (Thermo Scientific, Waltham, MA, USA) was used for the collection of ~~particulate matter (~~PM~~)~~ samples. Coarse particles with a diameter <10 μm (PM_{10}) were collected on dried Whatman QM-A 47–mm quartz fiber filters, housed in molded filter cassettes. The sampler was installed on the roofs of buildings at approximately 10 m above the ground level. The sampling was carried out for 24 h (6 a.m.–6 a.m.). Similarly, a relevant field sample blank was prepared, as the filter paper was exposed to the environment during the mounting and dismounting period. The mass difference (w_2-w_1) of the dried blank filter paper before (w_1) and after the exposure period (w_2) was evaluated and subtracted from the sample mass.

The road dust samples were collected using a stainless-steel scoop from eight locations along Raipur city's highway: Tatibandh, Hirapur, Sarora, Khamtarai, Birgaon, Urla, Sankra, and Siltara, during May 2008. These examined sites were situated in an area of approximate 10 km radius. The last four sites listed were located in the industrial area. The Khamtarai site was located in a heavy traffic area. The sample was collected from both sides (left and right) of the road junction in two 250-mL glass bottles. These samples were mixed in equal mass ratio to form a composite sample. Similarly, the sampling was extended up to period 2015, and one sample was taken every year in May from 2009 to 2015 at the Khamtarai site for the temporal variation studies. The dust samples were sundried for one week, and further dried at 50°C in a hot air oven overnight. The samples were then crushed into fine-powder form by sieving out of particles of mesh size >100 μm and finally stored in ~~an~~ aluminum foil for the analyses.

Ten surface sediment samples (0–10 cm depth) were collected in May 2008 from 10 ponds (Rohnipuram, Ashi, Budheshwar, Raja, Pandri, Siltara, Urkura, Birgoan, and Sarora) of Raipur city. These ponds were situated over an area of approximately 25 km^2. The last five listed ponds were located at industrial sites. For depth studies, two more sediment samples at a depth of 10–20 and 20–30 cm were collected in Siltara in 2008. The Raja pond was chosen for the temporal evolution investigation, and one sample was collected every year in May from 2009 to 2015.

A polyvinyl chloride (PVC) stormwater pipe of 5-cm radius (Simpson and Batley 2016) was used for the sediment collection. The samples were collected from five sites of the pond (east, west, north, and south corners, and midpoint). These samples were mixed equally to prepare a composite sample. They were dried, crushed, sieved, and stored in a similar manner to the road dust samples in a dust-free laboratory.

Carbon Speciation

Elemental carbon (EC), organic carbon (OC), and carbonate carbon (CC) were analyzed in the particulate matter, road dust, and sediment samples. An elemental ECS 4010 CHNSO analyzer (Costech Analytical Technologies Inc, USA) was used for the analysis of total carbon (TC). The dust samples were oxidized with O_2 at 1,020°C using ~~a constant~~ helium ~~flow~~ as a carrier. The resulting CO_2 gas was detected by a thermal conductivity detector. The CC content was removed by treating the sample with HCl acid in a CO_2-free atmosphere. ~~The EC and OC contents were determined by the thermal method described.~~ The OC content was analyzed by titration method using $K_2Cr_2O_7$ as oxidant. The EC content in the sample was evaluated by subtracting the CC and OC values from the TC content using

$$EC = TC - (CC + OC) \quad (1a)$$

where EC, CC, OC, and TC stand for the content of elemental carbon, carbonate carbon, organic carbon, and total carbon, respectively.

PCBs Analysis

The analyte sample was dried with sodium sulfate and extracted using a Dionex accelerated solvent extraction (ASE) system. The surrogate standards ($^{13}C_{12}$-labelled PCBs) were added and the samples were extracted with methylene chloride as a solvent. The extract was treated with copper to remove sulfur, and was purified by silica/alumina column chromatography to isolate the PCB fractions (Wade et al. 1988). The quantitative analyses were performed with a HP 5890 gas chromatograph (Agilent Technologies, Santa Clara, CA, USA), and a HP 5970 mass spectrometer in ~~the~~ ~~SIM~~ mode for PCBs, according to the method described by Sericano (2002). The quantification of the PCBs was based on the primary ion with supplementary monitoring of two additional masses for each analyte to verify the peaks identification.

Quality Assurance/Quality Control Analysis

One-way analysis of variance (ANOVA) was performed to analyze the distribution variability of the PCBs concentrations in the particulates, road dust, and sediments. ~~IBM~~ SPSS 20.0 (~~SPSS Inc.~~) software was employed for the factor analysis of source contributions of PCBs in the air, dust, and sediment by extracting factors with an Eigenvalue of >1.0 (Shyu et al. 2011).

The surrogate standard ($^{13}C_{12}$-labelled PCBs) was used to determine the extraction efficiency for the targeted PCB congeners. The standard was added prior to the extraction processes, and the average recoveries of these standards from the different environmental matrices varied from 91.1% to 102%. The calibration curve was prepared by injections of standard solutions containing a mixture of the PCBs at four concentration levels. The blanks used were clear of all the examined PCBs. The limits of detection (LODs) and quantification (LOQs) were determined. Reported values are expressed as an average across three replicate measurements, both for the carbon and PCB analyses.

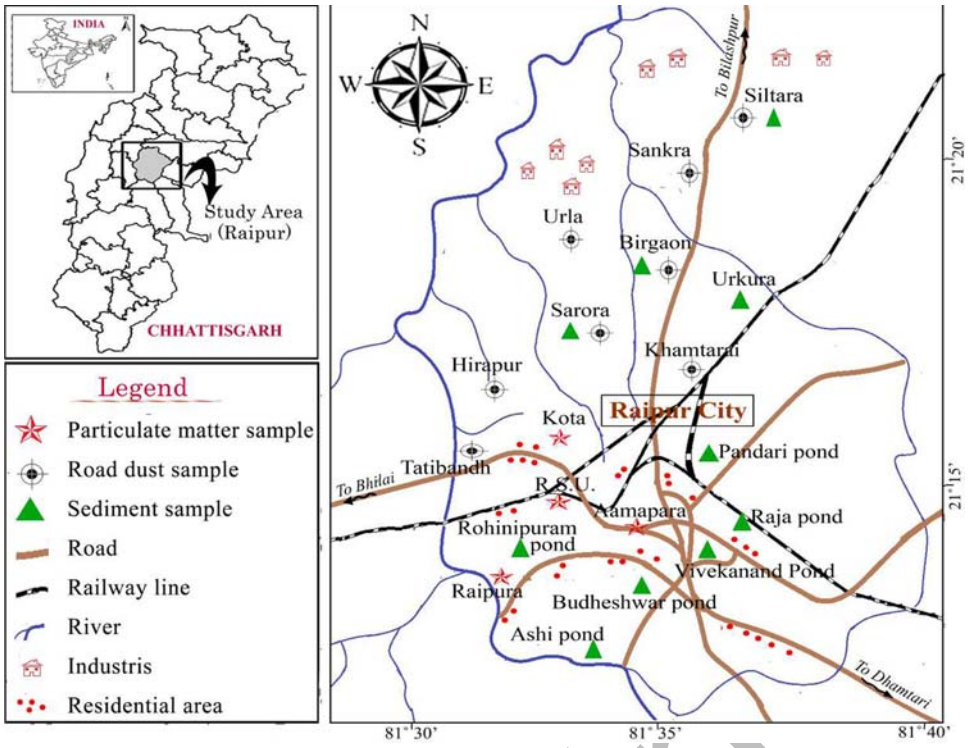

Fig. 1. Sample locations for collection of particulates, road dusts, and sediments in Raipur city. (Map by Yaman Kumar Sahu.)

Results and Discussion

Meteorology and Concentrations of Particulates, Carbon, and PCBs in Air

The meteorology (i.e., temperature, humidity, vapor pressure, wind speed, and sunshine) for the ~~study period~~ February 2007 to January 2008 ranged from 19.7°C to 38.1°C, 20.8% to 92.8%, 7.1 to 24.5 mm, 1.5 to 12 km h^{-1}, and 0.9 to 10 h day^{-1}, respectively. The lowest values of the ambient temperature, vapor pressure, wind speed, and sunshine were observed from December to January. A total rainfall of 96 cm, together with the maximum humidity, was registered from July to September 2007. The winds blew from the northeast and showed speeds of approximately 11 km during the June to August 2007 period. The lowest wind speed from the N direction was observed during the October 2007 to January 2008 period.

The major fraction of the PM was composed of carbons: EC, OC, and CC. The concentration of ($n = 24$) PM_{10}, EC_{10}, OC_{10}, and CC_{10} in the air during the February 2007 to January 2008 period ranged from 116 to 523, from 8.8 to 65.5, from 7.2 to 55.4, and from 6.1 to 58.7 μg m^{-3}, with mean values of 283 ± 138, 28.8 ± 16.9, 23.2 ± 13.6, and 22.7 ± 16.9 μg m^{-3}, respectively. The PM showed a negative correlation with the meteorological factors (i.e., rain–fall, temperature, humidity, vapor pressure, and wind speed) of $r = -0.31$–0.74. Three meteorological factors (rain, wind speed, and wind direction) markedly influenced the PM concentration. The highest mass concentration was observed in the winter season, December–January, expected due to the lowest atmospheric pressure, temperature, and wind speed. In turn in the rainy season (June–August), the lowest concentration was recorded, probably due to PM washout with the rain.

A similar distribution trend of the PM and carbons in ~~the~~ Raipur city was reported by Jaiswal et al. (2019b). In the hilly area of north India, the different distribution trend of the PM and carbons (maximal in the summer season) ~~was~~ likely due to precipitation of the particulates with the snow in the winter season (Ganguly et al. 2019).

Polychlorinated biphenyls are considered as significant environmental air contaminants due to their ecotoxicological and human health implications. The PM in the air is associated with large fraction of EC, OC, and CC. The concentration of ~~the~~ PM_{10}, EC, OC, and CC in the ambient air of Raipur city monitored at four locations (Kota, Pt. R. S. University, Raipura Chowk, and Amapara) were in the 388–845, 33–81, 23–52, and 28–65 μg m^{-3} range, respectively, with average values of 574 ± 222, 53 ± 23, 36 ± 14, and 44 ± 18 μg m^{-3}, respectively. The highest PM_{10} and carbon concentrations in the air corresponded to samples collected in Amapara, most probably due to vehicular emissions.

The concentrations of the PCBs congeners in the ambient air associated ~~to~~ the particulates (PM_{10}) are presented in Table 1. A total 83 congeners ~~in the ambient air~~ were detected. The total concentration of PCBs ($n = 4$) ranged from 72.07 to 429.57 pg m^{-3}, with a mean value of 247.54 pg m^{-3}. The maximum PCBs value was found for ~~the~~ Kota ~~site~~, expected due to vehicular emissions. The dominant congeners (1, 3, and 17) were found at concentrations in the 2.4–130.41, 49.31–147.74, and 1.42–18.8 pg m^{-3} ~~range~~s, with mean values of 46.97, 96.10, and 9.01 pg m^{-3}, respectively.

Table 1 summarizes the concentration of individual congeners in the air. Total concentrations of MCBs, dichlorobiphenyls (DCBs), trichlorobiphenyls (TCBs), tetrachlorobiphenyls (TeCBs), pentachlorobiphenyls (PeCBs), hexachlorobiphenyls (HCBs), heptachlorobiphenyls (HeCBs), octachlorobiphenyls (OCBs), and nonachlorobiphenyls (NCBs) examined in the ambient air varied from 53.16 to 278.15, from 4.44 to 37.50, from 2.57 to 33.49, from 1.81 to 57.46, from 6.65 to 30.85, from 3.02 to 27.24, from 2.19 to 11.87, from 0.16 to 2.45, and from 0.64 to 2.14 pg m^{-3}, respectively, with the highest and the lowest values for MCBs (143.06 ± 99.47 pg m^{-3}) and NCBs (1.12 ± 0.70 pg m^{-3}), respectively. ~~The~~ MCBs accounted for 54.2% of the total PCBs concentration in the air. The other congeners (DCBs, TCBs, TeCBs, PeCBs, HCBs,

Table 1. PCB concentration in ambient air and particulates collected in 2008

			Ambient air, pg m^{-3}					Ambient particulate (PM_{10}), ng g^{-1}				
S. no.	Type	Congener	Kota	RSU	Raipura	Amapara	Mean value ± STD	Kota	RSU	Raipura	Amapara	Mean value ± STD
1	MCBs	1	130.41	2.4	37.01	18.05	46.97 ± 57.40	196.40	6.19	337.53	21.36	140.37 ± 157.25
2		3	147.74	50.76	49.31	136.57	96.10 ± 53.38	222.50	130.82	382.87	161.62	224.45 ± 112.27
3	DCBs	4 + 10	0.49	0.05	0.69	1.96	0.80 ± 0.82	0.74	0.13	1.26	2.32	1.11 ± 0.93
4		6	0	0	0	0.06	0.02 ± 0.03	0.00	0.00	0.00	0.07	0.02 ± 0.04
5		7 + 9	0.01	0.03	0.14	0	0.05 ± 0.06	0.02	0.08	0.00	0.00	0.03 ± 0.04
6		8 + 5	3.51	1.79	2.87	3.45	2.91 ± 0.80	5.29	4.61	9.07	4.08	5.76 ± 2.26
7	TCBs	16 + 32	0	0	0	0.45	0.11 ± 0.23	0.00	0.00	0.00	0.53	0.13 ± 0.27
8		17	18.8	1.42	9.43	6.39	9.01 ± 7.31	28.31	3.66	48.87	7.56	22.10 ± 20.87
9		18	10.45	0	2.86	0	3.33 ± 4.94	15.74	0.00	27.20	0.00	10.74 ± 13.25
10		19	0.06	0.1	0.06	0.05	0.07 ± 0.02	0.09	0.26	0.25	0.06	0.17 ± 0.10
11		21 + 33 + 53	0.2	0.04	1.00	0.57	0.45 ± 0.43	0.30	0.10	0.50	0.67	0.39 ± 0.25
12		22	1.12	0.16	1.11	2.15	1.14 ± 0.81	1.69	0.41	3.02	2.54	1.92 ± 1.14
13		24	2.13	0.33	0.83	1.52	1.20 ± 0.79	3.21	0.85	5.54	1.80	2.85 ± 2.04
14		25	0.21	0	0.09	0.28	0.15 ± 0.12	0.32	0.00	0.50	0.33	0.29 ± 0.21
15		28 + 31	0.18	0.28	1.23	1.13	0.71 ± 0.55	0.27	0.72	0.50	1.34	0.71 ± 0.46
16		29	0.34	0.24	0.26	0.19	0.26 ± 0.06	0.51	0.62	1.01	0.22	0.59 ± 0.33
17	TeCBs	37 + 42	3.61	0	1.34	4.62	2.39 ± 2.10	5.44	0.00	9.32	5.47	5.06 ± 3.83
18		40	2.03	0.07	0.69	2.34	1.28 ± 1.08	3.06	0.18	5.29	2.77	2.83 ± 2.09
19		41 + 64 + 71	2.26	0.07	0.07	2.32	1.18 ± 1.28	3.40	0.18	5.79	2.75	3.03 ± 2.31
20		44	0.78	0	0.27	1.64	0.67 ± 0.72	1.17	0.00	2.02	1.94	1.28 ± 0.94
21		45	0.41	0.06	0	0.47	0.24 ± 0.24	0.62	0.15	1.01	0.56	0.59 ± 0.35
22		46	0.71	0	0.5	1.37	0.65 ± 1.49	1.07	0.00	1.76	1.62	1.11 ± 0.80
23		47 + 48	1.34	0.2	0.97	3.66	1.54 ± 1.49	2.02	0.52	3.53	4.33	2.60 ± 1.69
24		49	0.72	0.48	0	2.05	0.81 ± 0.88	1.08	1.24	1.76	2.43	1.63 ± 0.61
25		51	0	0	0	0.48	0.12 ± .24	0.00	0.00	0.00	0.57	0.14 ± 0.29
26		56 + 60	14.32	0.63	2.99	13.07	7.75 ± 6.95	21.57	1.62	37.03	15.47	18.92 ± 14.68
27		62	0.71	0.14	0.20	1.42	0.62 ± 0.59	1.07	0.36	1.76	1.68	1.22 ± 0.65
28		63	1.8	0.11	1.20	2.47	1.40 ± 1.00	2.71	0.28	4.79	2.92	2.68 ± 1.85
29		66 + 95	5.31	0	2.43	5.24	3.25 ± 2.55	8.00	0.00	13.85	6.20	7.01 ± 5.70
30		70.76	11.28	0.03	5.66	11.47	7.11 ± 5.44	16.99	0.08	29.22	13.57	14.97 ± 11.98
31		74	4.72	0.02	2.55	4.84	3.03 ± 2.27	7.11	0.05	12.34	5.73	6.31 ± 5.05
32	PeCBs	77 + 110	1.06	0.9	3.17	2.96	2.02 ± 1.21	1.60	2.32	2.77	3.50	2.55 ± 0.80
33		81 + 87	0.55	0	0.48	1.96	0.75 ± 0.84	0.83	0.00	1.51	2.32	1.17 ± 0.99
34		82 + 151	1.96	1.69	10.5	1.03	3.80 ± 4.49	2.95	4.36	5.04	1.22	3.39 ± 1.69
35		83	1.25	0	0.77	1.59	0.90 ± 0.69	1.88	0.00	3.27	1.88	1.76 ± 1.34
36		85	1.43	1.03	4.23	0.78	1.87 ± 1.60	2.15	2.65	3.78	0.92	2.38 ± 1.19
37		89	6.28	0	0.97	4.74	3.00 ± 2.99	9.46	0.00	16.37	5.61	7.86 ± 6.88
38		91	0.75	0.81	1.01	3.45	1.51 ± 1.30	1.13	2.09	2.02	4.08	2.33 ± 1.25
39		92 + 84	2.23	0.02	0.07	1.88	1.05 ± 1.17	3.36	0.05	5.79	2.22	2.86 ± 2.39
40		97	1.19	0	1.59	1.86	1.16 ± 0.82	1.79	0.00	3.02	2.20	1.75 ± 1.28
41		99	2.83	0	0.26	2.73	1.46 ± 1.53	4.26	0.00	7.30	3.23	3.70 ± 3.01
42		100	1.7	0.96	0.54	1.71	1.23 ± 0.58	2.56	2.47	4.53	2.02	2.90 ± 1.12
43		101	5.22	0	1.57	4.86	2.91 ± 2.54	7.86	0.00	13.60	5.75	6.80 ± 5.62
44		105	1.2	0.09	1.61	1.03	0.98 ± 0.64	1.81	0.23	3.02	1.22	1.57 ± 1.17
45		107	0.54	0.99	0.65	0.02	0.55 ± 0.40	0.81	2.55	1.51	0.02	1.22 ± 1.07
46		118	0.32	0.16	0.32	0.22	0.26 ± 0.08	0.48	0.41	0.76	0.26	0.48 ± 0.21
47		123 + 149	1.63	0	0.07	0.03	0.43 ± 0.80	2.45	0.00	4.28	0.04	1.69 ± 2.07
48	HCBs	132	0.86	0	1.22	0.85	0.73 ± 0.52	1.30	0.00	2.27	1.01	1.15 ± 0.93
49		134	0.82	0	0.88	0.57	0.57 ± 0.40	1.23	0.00	2.02	0.67	0.98 ± 0.86
50		135 + 144	0	0	1.5	3.04	1.14 ± 1.45	0.00	0.00	0.00	3.60	0.90 ± 1.80
51		136	0.82	1.71	5.1	4.01	2.91 ± .98	1.23	4.41	2.02	4.75	3.10 ± 1.74
52		137 + 130 + 176	2.29	0	2.26	1.44	1.50 ± 1.07	3.45	0.00	6.05	1.70	2.80 ± 2.58
53		138 + 158	7.37	0.37	7.36	8.03	5.78 ± 3.62	11.10	0.95	19.14	9.50	10.17 ± 7.46
54		141	0.31	0.05	1.59	1.34	0.82 ± 0.76	0.47	0.13	0.76	1.59	0.74 ± 0.62
55		146	0.33	0	1.27	0.07	0.42 ± 0.59	0.50	0.00	0.76	0.08	0.34 ± 0.36
56		153	3.62	0.89	4.97	3.18	3.17 ± 1.70	5.45	2.29	9.32	3.76	5.21 ± 3.03
57		157	0.9	0	0	3.07	0.99 ± 1.45	1.36	0.00	2.27	3.63	1.82 ± 1.53
58		163	0.47	0	1.09	0	0.39 ± 0.52	0.71	0.00	1.26	0.00	0.49 ± 0.61
59	HeCBs	172 + 197	0.92	0.31	0.19	0.72	0.54 ± 0.34	1.39	0.80	2.27	0.85	1.33 ± 0.68
60		174	0.7	0.11	0.14	0.67	0.41 ± 0.32	1.05	0.28	1.76	0.79	0.97 ± 0.62
61		177	1.4	0.07	0.28	1.36	0.78 ± 0.70	2.11	0.18	3.53	1.61	1.86 ± 1.38

Table 1. (*Continued*.)

S. no.	Type	Congener	Ambient air, pg m^{-3}					Ambient particulate (PM_{10}), ng g^{-1}				
			Kota	RSU	Raipura	Amapara	Mean value ± STD	Kota	RSU	Raipura	Amapara	Mean value ± STD
62		178 + 129	1.03	0.57	1.16	0.86	0.91 ± 0.25	1.55	1.47	2.77	1.02	1.70 ± 0.75
63		180 + 193	0.88	1.01	3.58	1.57	1.76 ± 1.25	1.33	2.60	2.27	1.86	2.02 ± 0.55
64		183	2.86	0	0.52	1.07	1.11 ± 1.24	4.31	0.00	7.30	1.27	3.22 ± 3.27
65		185	0.82	0	0.13	1.03	0.50 ± 0.51	1.23	0.00	2.02	1.22	1.12 ± 0.83
66		202 + 171 + 156	3.26	0.12	0.73	1.71	1.46 ± 1.37	4.91	0.31	8.56	2.02	3.95 ± 3.61
67	OCBs	194	0.12	0	0.04	0.03	0.05 ± 0.05	0.18	0.00	0.25	0.04	0.12 ± 0.12
68		196 + 203	0.55	0	0.77	0.63	0.49 ± 0.34	0.83	0.00	1.51	0.75	0.77 ± 0.62
69		198	0.12	0	0.3	0.25	0.17 ± 0.14	0.18	0.00	0.25	0.30	0.18 ± 0.13
70		199	0.25	0	0	0	0.06 ± 0.13	0.38	0.00	0.76	0.00	0.29 ± 0.36
71		201	1.41	0.16	0.5	0.55	0.66 ± 0.53	2.12	0.41	3.78	0.65	1.74 ± 1.56
72	NCBs	206	0.67	0.64	1.03	2.14	1.12 ± 0.70	1.01	1.65	1.76	2.53	1.74 ± 0.62

Note: RSU = Ravishankar University; MCBs = monochlorobiphenyls; DCBs = dichlorobiphenyls; TCBs = trichlorobiphenyls; TeCBs = tetrachlorobiphenyls; PCBs = pentrachlorobiphenyls; HCBs = hexachlorobiphenyls; HeCBs = heptachlorobiphenyls; OCBs = octachlorobiphenyls; and NCBs = nonachlorobiphenyls.

HeCBs, OCBs, and NCBs) contributed the remaining 45.8% of the PCBs content.

A lower PCBs content was observed in Raipur city (248 ± 152 pg m^{-3}) than the one reported for the ambient air in Kanpur city (254–432 pg m^{-3}) (Goel et al. 2016), although it was considerably higher than the contents reported (4–389 pg m^{-3}) in other locations, such as Japan, France, Atlantic Ocean, Korea, or Pakistan (Nasir et al. 2014; Syed et al. 2013; Baek et al. 2010; Gioia et al. 2008; Blanchard et al. 2006; Kim and Masunaga 2005).

Concentration of Carbon and PCBs in Particulate, Dust, and Sediments

EC, OC, and PCBs are emitted during various combustion and industrial processes (Brunciak et al. 2001) and are distributed in various environmental compartments (viz. air, water, dust, and soil) in urban and industrial areas (Malina and Mazlova 2017). Along roads, they are predominantly emitted by vehicular emissions (Liu et al. 2019). The environmental PCBs are transported to water reservoirs by rain, runoff water, industrial and municipal waste, etc. (Froese et al. 1997). Tables 1–3 summarize the environmental contamination of PCBs in the ambient particulates, road dust, and sediments of the study area.

The concentration of EC, OC, and CC in the coarse particulate matter (PM_{10}) ranged from 8.4% to 9.61%, from 5.45% to 8.81%, and from 7.54% to 9.43%, respectively, with mean values of 9.15% ± 0.55%, 6.87% ± 1.20%, and 8.74% ± 0.61%, respectively. The comparable EC concentration in the road dust and sediments varied from 5.8% to 6.61% and from 7.29% to 7.77%, with average values of 6.26% ± 0.31% and 7.56% ± 0.21%. However, very low OC (0.39% ± 0.09% and 0.49% ± 0.06%) and CC (0.15% ± 0.03% and 0.12% ± 0.02%) concentrations in the dust and sediment samples were found, as compared to the particulate samples. A higher EC concentration in the studied area than in other locations reported in the literature, both in India and in the rest of world, was observed (Zong et al. 2016; Guha et al., 2015; Han et al. 2015; Ozdemir et al. 2014; Han et al. 2009). This may be tentatively ascribed to massive coal burning in the area, given that two of India's largest coal-fired power stations are operating in Chhattisgarh state.

The concentration of the individual PCBs congeners in the ambient air and particulates is presented in Table 1. The total concentration of PCBs in the ambient particulate matters of four locations ranged from 185.72 to 1,110.82 ng · g^{-1}, with an average value of 574.05 ± 405.12 ng g^{-1}. The concentrations of MCBs, DCBs, TCBs, TeCBs, PeCBs, HCBs, HeCBs, OCBs, and NCBs were the 137.01–720.4, 4.82–10.33, 6.62–87.39, 4.66–129.47, 17.13–78.57, 7.78–45.87, 0.41–6.55, and 1.01–2.53 ng g^{-1} intervals, respectively, with the highest and the lowest values for MCBs (364.82 ± 267.29 ng g^{-1}) and NCBs (1.74 ± 0.62 ng g^{-1}), respectively. Similarly, the major portion of the PCBs in particulate samples was contributed by ~~the~~ MCBs congeners.

As regards PCBs in the road dust samples, 98 congeners were detected. Their concentrations in eight locations of highway are summarized in Table 2. The total concentration of PCBs ranged from 102 to 537 ng g^{-1}, with an average value of 241 ng g^{-1}. The concentrations of MCBs, DCBs, TCBs, TeCBs, PeCBs, HCBs, HeCBs, OCBs, and NCBs were in the following intervals: 37.0–197.4, 7.2–32.4, 11.7–40.6, 0–170.5, 9.4–99.1, 15.4–29.8, 1.5–6.2, 0.7–8.8 and 0–1.3 ng g^{-1}, respectively, with average values of 93.4 ± 67.38, 16.26 ± 8.13, 23.68 ± 10.33, 51.90 ± 56.70, 28.30 ± 30.19, 21.55 ± 4.92, 3.09 ± 1.60, 2.75 ± 2.90 and 0.16 ± 0.46 ng g^{-1}, respectively. The highest total concentration of PCBs was detected at the Khamtarai site, expected due to higher vehicular emissions. The concentration of the ~~prominent~~ congeners was detected in the following increasing order: NCBPs < OCBPs < HeCBPs < DCBPs < HCBPs < TCBPs < PCBPs < TeCBPs = MCB~~P~~s. The total PCBs concentration showed a fair correlation with the congener frequency ($r = 0.61$). It is worth noting that the concentration of PCBs (241 ± 146 ng g^{-1}) in the area of study was higher than those reported for Guangzhou, India; Hong Kong; Chennai, India; and Northern Vietnam (in the 0.25–228 ng kg^{-1} range) (Anh et al. 2019; Chakraborty et al. 2016; Wang et al. 2013).

In pond sediments, 84 PCBs congeners were detected. Their concentrations in the 10 ponds are given in Table 3. The total concentration of PCBs ranged from 241 to 538 ng g^{-1}, with a mean value of 328 ng g^{-1}. The highest PCB content was found in ~~the~~ Raja pond, ascribed to increased human activities in the area. The concentrations of MCBs, DCBs, TCBs, TeCBs, PeCBs, HCBs, HeCBs, and OCBs were in the following ranges: 123.80–372.70, 7.30–29.90, 13.40–46.80, 5.90–74.50, 0–80.20, 0–4.10, and 0–11.10 ng g^{-1}, respectively, with mean values of 201.31 ± 80.97, 17.24 ± 7.0, 28.88 ± 10.84, 30.14 ± 19.91, 25.25 ± 19.73, 22.41 ± 24.68, 1.04 ± 1.36, and 1.31 ± 3.47 ng g^{-1}, respectively. A different trend of occurring ~~of~~ PCBs congeners was found in this case: HeCBs < OCBs < DCBs < HCBs < PCBs < TCBs < TeCBs < MCBs. No NCBs were detected in the surface sediment.

Table 2. Concentration of PCBs in road dust collected in 2008, ng g^{-1}

S. no.	Type	Congener	TB	HP	S	KT	B	U	SK	ST	Mean value ± STD
1	MCBs	1	18.4	15.5	62.3	141.1	125.7	43.7	37.0	44.7	61.05 ± 47.25
2		3	18.6	23.7	43.3	52.7	71.7	35.0	0	13.8	32.35 ± 23.13
3	DCBs	4 + 10	0.0	2.7	0	1.6	0	0	0	0	0.54 ± 1.04
4		6	1.0	1.8	2.1	1.5	2.9	1.0	1.7	1.7	1.71 ± 0.61
5		7 + 9	0.4	0.7	0.5	0.5	0.7	0.6	0.6	0.5	0.56 ± 0.11
6		8 + 5	5.8	11.6	15.3	9.4	28.8	7.3	9.6	19.8	13.45 ± 7.66
7	TCBs	16 + 32	1.6	0.0	2.3	4.2	1.9	1.7	5.0	2.5	2.40 ± 1.57
8		17	7.0	8.7	5.3	5.1	1.9	1.8	1.8	4.8	4.55 ± 2.57
9		18	1.1	1.7	4.3	0.7	0.8	0.8	3.6	3.5	2.06 ± 1.49
10		19	0.5	1.8	1.4	1.1	1.9	0.5	0.9	0.9	1.13 ± 0.54
11		21 + 33 + 53	0.0	0	1.6	4.6	1.2	1.3	0.8	5.9	1.93 ± 2.16
12		22	0.0	0	2.6	4.2	5.4	1.1	4.3	6.0	2.95 ± 2.38
13		24	1.1	0.9	1.7	0.8	1.1	0.5	1.9	0.9	1.11 ± 0.47
14		25	1.4	0	0.9	2.4	1.5	0.5	1.1	1.2	1.13 ± 0.71
15		26	0.4	0	0.6	1.7	1.3	0.6	0.9	1.4	0.86 ± 0.57
16		28 + 31	1.3	0	4.5	6.7	4.0	2.3	2.4	12.9	4.26 ± 4.05
17		29	1.0	0	3.1	2.7	0.7	0.6	1.7	0.6	1.30 ± 1.10
18	TeCBs	37 + 42	0	0	11.2	14.4	4.9	1.6	2.0	22.4	7.06 ± 8.16
19		40	0	0	0.0	8.5	0	0	0	1.7	1.28 ± 2.98
20		41 + 64 + 71	0	0	4.8	10.0	2.6	0.9	2.0	14.9	4.40 ± 5.37
21		44	0	0	2.0	9.5	2.1	0.4	0.9	3.0	2.24 ± 3.13
22		45	0	0	0.6	6.3	1.2	0.4	0.4	1.3	1.28 ± 2.09
23		46	0	0	7.9	14.9	6.9	1.1	3.0	1.6	4.43 ± 5.18
24		47 + 48	0	0	1.0	9.4	1.0	0.7	0.0	1.9	1.75 ± 3.16
25		49	0	0	25.4	17.3	9.0	6.6	13.1	3.8	9.40 ± 8.85
26		51	0	0	0.7	2.4	0.0	0.5	0.0	0.0	0.45 ± 0.83
27		56 + 60	0	0	2.9	5.3	5.5	2.5	2.9	6.4	3.19 ± 2.42
28		62	0	0	4.5	7.1	2.0	0.9	1.3	3.7	2.44 ± 2.49
29		63	0	0	0.6	3.3	0.8	0.0	0.0	0.8	0.69 ± 1.12
30		66 + 95	0	0	1.0	21.8	0.7	1.2	0.0	4.3	3.63 ± 7.48
31		70.76	0	0	10.3	33.3	4.6	1.2	2.8	8.4	7.58 ± 1.06
32		74	0	0	2.0	7.0	0.7	0.6	0.7	5.9	2.11 ± 2.76
33	PeCBs	77 + 110	6.5	0.8	6.4	1.8	4.4	1.8	2.6	1.4	3.21 ± 2.26
34		81 + 87	0	0	0	1.1	0.8	0.4	0.7	0.2	0.40 ± 0.42
35		82 + 151	1.0	3.8	0	0.0	0.0	0.8	1.2	0.0	0.85 ± 1.30
36		83	0.4	0.4	1.7	1.2	0.8	0.4	0.5	0.7	0.76 ± 0.47
37		85	0	0	0	0.7	0.5	12.8	0.5	0.1	1.83 ± 4.44
38		89	0.4	3.1	1.2	2.8	3.5	0.9	2.1	1.4	1.93 ± 1.12
39		91	0.0	0.0	9.9	82.5	0.0	15.4	0.0	0.0	13.48 ± 28.51
40		97	0.1	0.2	0.3	1.0	0.9	0.3	0.7	0.7	0.53 ± 0.34
41		99	0.0	0.1	0.2	0.2	0.1	0.8	0.1	0.1	0.20 ± 0.25
42		100	0.0	0.0	0.5	4.1	1.5	0.4	0.6	1.9	1.13 ± 1.38
43		101	0.7	1.0	1.1	1.3	1.6	0.8	0.9	0.1	0.94 ± 0.44
44		105	1.1	1.3	1.0	1.1	1.3	1.7	1.5	1.4	1.30 ± 0.23
45		118	0.7	0.5	0	0.4	0	0.8	0.8	0.5	0.46 ± 0.32
46		123 + 149	1.5	1.4	0.9	0.9	0.8	2.4	1.6	0.9	1.30 ± 0.55
47	HCBs	129 + 178	0	0	0.7	1.2	0	0.4	1.3	1.2	0.60 ± 0.58
48		132	0.7	0.3	0.6	0.5	0.9	0.8	1.1	0.8	0.71 ± 0.25
49		134	0	3.2	0.0	0.7	0.0	1.3	1.4	0.6	0.90 ± 1.09
50		135 + 144	1.1	2.6	1.5	2.4	0.0	2.0	3.1	1.7	1.80 ± 0.97
51		136	0.3	0.8	0	1.3	1.3	0.9	1.9	0.3	0.85 ± 0.64
52		137 + 130 + 176	0	0	0	0.6	0.8	0.4	0.9	0.6	0.41 ± 0.37
53		138 + 158	19.2	1.0	0	0.0	14.2	0.0	0.0	0.0	4.30 ± 7.78
54		141	0.6	0.8	0.8	1.1	1.3	0.7	1.3	1.0	0.95 ± 0.27
55		146	1.1	0	0	1.0	1.3	1.2	1.7	0.9	0.90 ± 0.60
56		153	2.3	3.6	2.2	4.3	4.0	3.5	0.0	3.3	2.90 ± 1.39
57		157	0.5	2.3	0.9	1.2	0.0	0.0	0.0	1.6	0.81 ± 0.85
58		163	0.0	8.2	23.1	5.4	0.0	4.2	3.6	6.8	6.41 ± 7.35
59	HeCBs	170 + 190	0	0	0	0	0	0	0	2.0	0.25 ± 0.71
60		177	1.1	0.5	1.3	1.7	1.4	0.5	1.3	1.6	1.18 ± 0.46
61		183	0.6	0.0	0.5	1.0	0.9	0.5	1.3	1.1	0.74 ± 0.42
62		185	0.2	0.0	0.1	1.8	1.3	0.5	0.2	1.5	0.70 ± 0.72
63		187 + 182	0	1.8	0	0	0	0	0	0	0.23 ± 0.64

Table 2. (*Continued*.)

S. no.	Type	Congener	TB	HP	S	KT	B	U	SK	ST	Mean value ± STD
64	OCBs	194	0	0	0	0	0	0	0	0.5	0.06 ± 0.18
65		196 + 203	0	2.8	0	0	0	0	0	1.4	0.53 ± 1.04
66		198	0	2.2	0	0	0	0	0	1.0	0.40 ± 0.81
67		201	2.1	3.8	1.1	1.4	1.2	0.7	1.1	2.7	1.76 ± 1.04
68	NCBs	208 + 195	0	0	0	0	0	0	0	1.3	0.16 ± 0.46

Note: TB = Tatibandh; HP = Hirapur; SA = S; KT = Khamtarai; B = Birgaon; U = Urla; SK = Sankra; and ST = Siltara.

When the PCBs concentration in the pond sediment samples was analyzed as a function of depth, it was observed that the concentrations of MCBs, DCBs, TCBs, TeCBs, PeCBs, and HeCBs congeners increased steeply with depth, probably due to poor adsorption on the sediment particles (Fig. 2). However, the concentration of HCB decreased as the depth increased, suggesting an adsorption by the top-layer sedimentary particles (Fig. 2). A noticeable vertical distribution of congeners 1, 3, 4 + 10, 6, 7 + 9, 8 + 5, 17, 18, 19, 77 + 110, and 85 was observed.

For comparison purposes, the PCBs concentration in six ponds in Bhilai and Korba varied from 201 to 648 ng · g^{-1} and from 404 to 773 ng g^{-1}, with average values of 480 ± 150 and 561 ± 155 ng g^{-1}, respectively (Patel et al. 2013). The concentrations of aforementioned congeners in these two cities (in which huge quantities of coal are burnt for steel and electricity production) were even higher than those found in Raipur. It is also worth noting that the concentration of PCBs (328 ± 99 ng g^{-1}) in the pond sediments from the area of study was higher than those observed (<0.01–126.49 ng g^{-1}) in soil/sediment of other locations (Jin et al. 2012; Li et al. 2012; Kumar et al. 2011), except for Harbor Island East, with concentrations of up to at 1,387 ng g^{-1} (Neira et al. 2018).

Comparison of PCBs Concentrations in Environmental Samples

Comparable total PCBs concentrations were observed in the pond sediments and road dust samples, while their concentration in the particulate samples was markedly increased [Fig. 3(a)]. The highest concentrations of MCBs, HeCBs, OCBs, and NCBs were found in the particulates; those of TeCBs and PeCBs in the road dusts (Fig. 3); and those of DCBs, TCBs, and HCBs in the sediments (Fig. 3). Remarkably higher contents of MCBs and HeCBs were detected in the PM samples, which could be an indicator of air pollution.

A high concentration of congeners 3, 17, 89, and 138 + 158 was identified in the PM and sediment samples, indicating emissions ~~by~~ multiple sources. The ~~dominant~~ concentration of congeners 1, 8 + 5, 49, and 91 was registered in the road dust samples, ~~showing~~ emissions mainly ~~by~~ vehicles.

As per the ANOVA test, the uncertainty (F) value for the PCBs concentration in the particulates, road dust, and sediments were found to be 7, 65, 535, and 29, indicating multiple emission sources of the PCBs in the road geo-media.

Temporal Evolution of PCBs Concentration

The total concentrations of PCBs in particulate matter (Kota), road dust (Khamtarai), and sediments (Raja pond) were monitored over an eight-year period (2008–2015), as shown in Fig. 4, and registered a gradual increase from 428 to 670 pg m^{-3}, from 537 to 752 and from 538 to 775 ng g^{-1}, ~~with~~ respectively, probably due to increased vehicular and industrial emissions (Fig. 4). These data represented an average annual increment rate of 6.2% ± 3.2%, 4.9% ± 2.1%, and 5.4% ± 1.1% in PCBs concentrations, respectively.

Toxic Equivalency Factor of PCBs

The toxic equivalency factor (TEF) denotes the toxicity of individual congeners in terms of the most toxic form of dioxin: 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD), for which a value of 1.0 is assigned (USEPA 2010). Among the congeners found, PCB-126 (3,3′,4,4′,5-pentachlorobiphenyl) was the most toxic (USEPA 2010), with a TEF of 0.1, while the TEF factors for PCB-77 (3,3′,4,4′-tetrachlorobiphenyl), PCB-81 (3,4,4′,5-tetrachlorobiphenyl), and others are 0.0001, 0.0003, and 0.00003, respectively. Among the aforementioned "dioxin-like" congeners, 77, 81, 105, 118, 123, and 157 were detected in most of the particulate matter, road dust, and sediment samples. The normalized total mean TEFs for the PM, road dust, and sediments from Raipur city were estimated at 0.00066, 0.00058, and 0.00047, respectively.

Correlations and Sources

Several chemical species, including EC, OC, polycyclic aromatic hydrocarbons (PAHs), and biphenyls (BPs), are emitted during the combustion processes of fuels (Laroo et al. 2012; Hesterberg et al. 2008; Na et al. 2004).

The main PCBs emission sources are the chlorination of biphenyls during the combustion process of fuels and other materials, vaporization; leakage from application sites of Aroclors; and burning, disposal, and dumping of PCBs-containing materials (Meijer et al. 2003).

The combustion of fuels, industrial/metallurgical activities and power generation processes have been reported as possible sources of PCBs, due to reaction of carbon and chlorine at the combustion source (Biterna and Voutsa 2005; Dyke et al. 2003; Weber et al. 2001).

The total PCBs content showed a fair correlation (at $r = 0.73$–0.75) with PM, EC, OC, and CC, indicating that PCBs would be partly originated from burning processes (Table 4). MCBs, DCBs, and TCBs had a good correlation with TCBs, TeCBs, HeCBs, and OCBs ($r = 0.79$–0.93); TeCBs, PeCBs, HCBs, HeCBs, and NCBs ($r = 0.76$–0.91); and MCBs, HeCBs, and OCBs ($r = 0.91$–0.96), respectively, indicating that they would be originated by the chlorination process of lower congeners (1*b*)–(1*g*) as follows (Biterna and Voutsa 2005; Dyke et al. 2003; Weber et al. 2001)

$$C_{12}H_{10} + Cl_2 \rightarrow C_{12}H_9Cl + HCl \quad (1b)$$

$$C_{12}H_9Cl + Cl_2 \rightarrow C_{12}H_8Cl_2 + HCl \quad (1c)$$

$$C_{12}H_8Cl_2 + Cl_2 \rightarrow C_{12}H_7Cl_3 + HCl \quad (1d)$$

Table 3. Distribution of PCBs in sediments of Raipur city collected in 2008, ng g^{-1}

S. no.	Type	Congener	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	Mean value ± STD
1	MCBs	1	66.9	68.7	79.2	103.5	64.6	81.0	116.7	89.3	124.3	95.2	88.94 ± 20.86
2		3	65.8	165.1	44.6	54.5	87.6	58.7	93.7	283.4	174.9	95.4	112.37 ± 74.67
3	DCBs	4 + 10	0	4.8	0	0	5.7	0.0	4.1	5.9	10.7	0	3.12 ± 3.72
4		6	2.4	0.1	1.6	2.0	1.5	2.4	4.7	4.7	4.8	3.1	2.73 ± 1.59
5		7 + 9	0.7	0.0	0.6	0.7	0.8	0.8	0.4	2.1	1.7	0.7	0.85 ± 0.61
6		8 + 5	17.1	2.4	14.9	10.2	4.0	6.4	12.8	11.3	12.7	13.6	10.54 ± 4.81
7	TCBs	16 + 32	2.0	2.4	5.2	4.6	4.0	2.3	4.3	3.1	4.4	2.6	3.49 ± 1.14
8		17	0	0	4.3	7.2	5.8	3.6	4.4	4.3	7.0	9.5	4.61 ± 3.01
9		18	1.8	0.1	2.9	2.7	2.5	1.8	1.3	1.5	2.4	1.4	1.84 ± 0.83
10		19	2.0	0.3	1.7	2.4	3.4	3.5	2.6	1.4	6.8	2.8	2.69 ± 1.73
11		21 + 33 + 53	2.2	1.3	2.5	2.0	4.6	1.9	1.2	1.7	4.2	1.6	2.32 ± 1.17
12		22	2.8	1.9	3.2	2.4	8.7	2.0	1.8	2.2	4.6	0.7	3.03 ± 2.24
13		24	0.0	0.8	1.0	1.3	1.3	0.9	0	0	1.7	1.1	0.81 ± 0.61
14		25	1.7	1.0	2.0	2.1	2.7	1.2	1.6	1.5	2.3	0.9	1.70 ± 0.58
15		26	0.9	1.6	2.1	2.0	5.2	3.0	1.4	1.5	4.9	0.7	2.33 ± 1.57
16		28 + 31	4.1	3.2	5.5	4.8	6.4	3.5	3.8	3.7	5.6	3.3	4.39 ± 1.12
17		29	1.9	0.8	2.4	2.6	2.2	1.9	1.5	1.8	1.5	0.1	1.67 ± 0.75
18	TeCBs	37 + 42	2.0	1.9	3.2	0.9	10.4	2.0	1.6	0.0	6.5	2.2	3.07 ± 3.10
19		41 + 64 + 71	2.7	1.5	12.4	1.4	5.1	1.2	1.6	1.2	6.8	0.9	3.48 ± 3.69
20		44	1.0	0.8	0.6	0.8	4.6	0.7	0.7	0.7	3.6	0.4	1.39 ± 1.46
21		45	1.1	0.8	1.2	2.0	4.2	0.6	0.8	0.7	3.0	0.6	1.50 ± 1.21
22		46	1.1	0.7	1.3	1.1	3.8	0	1.7	1.0	2.6	0.8	1.41 ± 1.08
23		47 + 48	1.6	0	1.1	0	3.9	0	1.8	0	2.8	0.8	1.20 ± 1.35
24		49	2.7	1.9	3.2	4.0	4.8	0	1.9	1.4	3.4	2.7	2.60 ± 1.37
25		51	2.4	0	2.2	0	4.7	1.9	0.0	0	2.3	0.0	1.35 ± 1.61
26		56 + 60	4.0	4.4	4.2	0	10.4	3.5	4.4	0	8.0	8.6	4.75 ± 3.42
27		62	1.7	1.2	1.6	1.4	4.3	0	1.8	1.2	3.3	1.5	1.80 ± 1.19
28		63	1.2	0	1.0	0	1.5	1.1	0	0	2.4	0	0.72 ± 0.85
29		66 + 95	2.7	1.5	2.2	1.8	5.0	3.4	1.6	1.1	4.7	1.4	2.54 ± 1.39
30		70.76	3.1	1.8	2.5	1.8	5.7	2.3	2.1	1.5	4.1	2.5	2.74 ± 1.28
31		74	2.0	1.0	1.2	0.9	3.3	1.3	1.0	0.8	3.5	0.9	1.59 ± 1.01
32	PeCBs	77 + 110	5.8	0.4	1.8	1.1	3.2	2.3	2.2	1.0	7.2	4.1	2.91 ± 2.20
33		81 + 87	0.8	0	0	0	1.2	0.6	0	0	0	0.4	0.30 ± 0.43
34		82 + 151	2.3	0	0	0	2.8	0	0	0	0	5.1	1.02 ± 1.79
35		83	0.9	0	0.1	0.5	0.7	1.1	0.3	0.5	1.0	1.5	0.66 ± 0.47
36		85	8.2	0	7.5	4.1	3.8	15.3	0.1	11.4	0	0.3	5.07 ± 5.39
37		89	2.4	0.9	0	1.1	6.5	3.5	1.1	0.1	3.9	2.3	2.18 ± 2.01
38		91	0	15.0	0	0	46.8	0	0	0	0	0	6.18 ± 15.03
39		97	0.6	0.1	0.1	0.3	1.3	0.4	0.2	0.2	0.2	0.9	0.43 ± 0.39
40		99	0.8	0.7	0.3	1.3	1.1	1.0	0.5	0.7	1.4	0.6	0.84 ± 0.35
41		100	1.6	0.7	1.7	1.4	2.0	2.3	0.5	0.8	1.1	0.9	1.30 ± 0.60
42		101	2.0	1.3	0.7	1.7	2.9	1.6	1.0	0.4	3.4	1.9	1.69 ± 0.93
43		105	4.3	0	0	0	2.2	1.6	0	0	3.1	2.1	1.33 ± 1.57
44		118	3.5	0	0	0	0	0	0	0	0	0	0.35 ± 1.11
45		123 + 149	5.9	0	1.1	0	0	0	0	0	0	2.9	0.99 ± 1.96
46	HCBs	129 + 178	1.8	0	0	0	1.4	0.7	0	0	3.3	0	0.72 ± 1.13
47		132	2.6	0	0	0	0.8	0.7	0.9	0	0	1.6	0.66 ± 0.88
48		134	8.3	0	0	0	0	0	0	0	0	3.3	1.16 ± 2.71
49		135 + 144	2.1	0	1.4	0.7	3.5	0.8	1.4	0	1.8	2.9	1.46 ± 1.16
50		136	4.4	0	0	0	3.8	0.9	0	0	0	2.1	1.12 ± 1.72
51		137 + 130 + 176	2.1	0	0	0	0	0	0	0	2.0	0	0.41 ± 0.86
52		138 + 158	13.6	0	5.6	5.5	6.4	14.6	7.8	5.6	71.2	12.6	14.29 ± 20.49
53		141	2.2	0	0.3	0	1.3	0.9	0.6	0	1.9	0	0.72 ± 0.83
54		146	2.3	0	0	0	0	0	0	0	0	0	0.23 ± 0.73
55		153	7.8	0	0	0	0	0	0	0	0	5.5	1.33 ± 2.86
56		157	1.7	0	0	1.4	0	0	0	0	0	0	0.31 ± 0.66
57	HeCBs	177	0.0	0	0	0	0.7	0	0	0	1.8	0	0.25 ± 0.59
58		183	1.2	0	0	0	0	0	0	0	1.6	1.0	0.38 ± 0.63
59		185	0.8	0	0	0.7	1.2	0	0.1	0	0.7	0.6	0.41 ± 0.44
60	OCBs	194	0.8	0	2.8	0	0	0	0	0	1.2	0	0.48 ± 0.92
61		208 + 195	0.0	0	8.3	0	0	0	0	0	0	0	0.83 ± 2.62

Note: S1 = Siltara; S2 = Urkura; S3 = Birgoan; S4 = Sarora; S5 = Rohnipuram; S6 = Ashi; S7 = Budheshwar; S8 = Vivekanand; S9 = Raja; and S10 = Pandri.

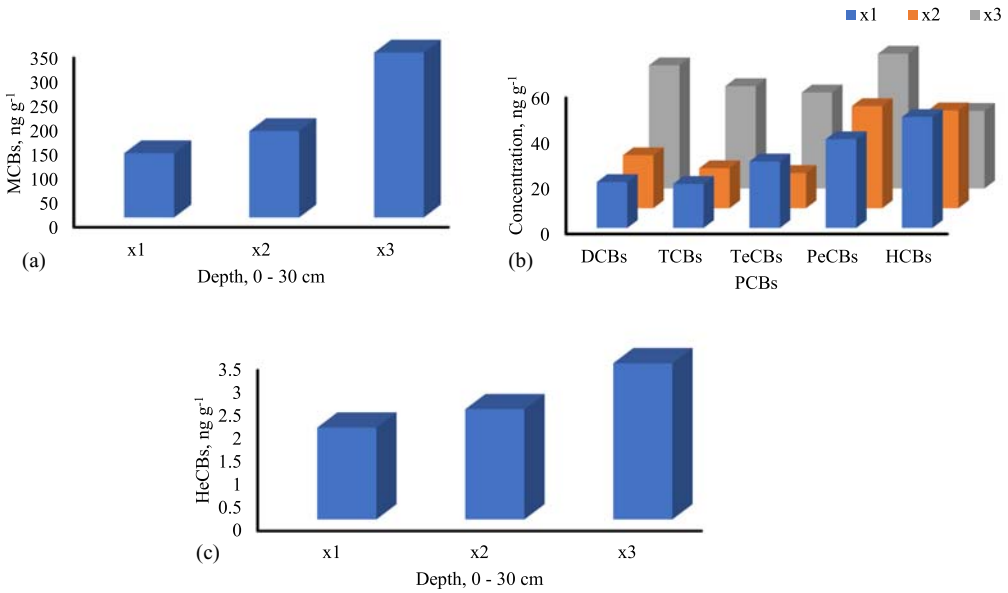

Fig. 2. Vertical concentration variation of PCBs in Siltara sediment. $x1 = 0–10$ cm, $x2 = 10–20$ cm, $x3 = 20–30$ cm: (a) monochlobiphenyls (MCBs); (b) mono-, di-, tri-, tetra-, penta-, and hexachlorobiphenyls (MCBs, DCBs, TCBs, TeCBs, PCBs, and HCBs); and (c) heptachlorobiphenyls (HeCBs).

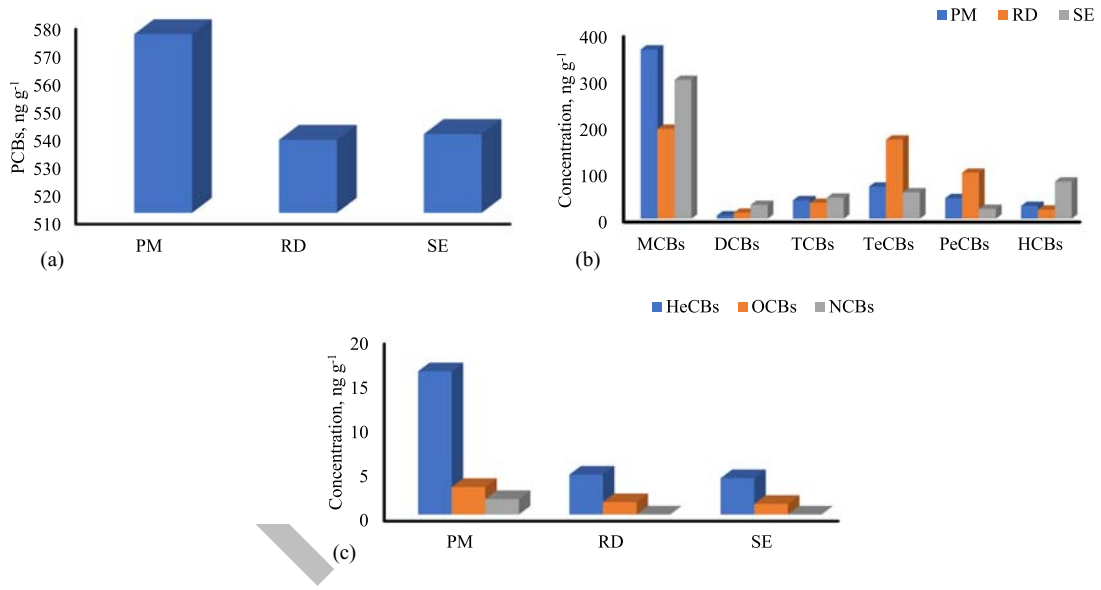

Fig. 3. PCBs concentration variation in particulate matter (PM), road dust (RD), and sediment (SE): (a) polychlorobiphenyls (PCBPs); (b) di-, tri-, tetra-, penta- and hexachlorobiphenyls (DCBs, TCBs, TeCBs, PCBs and HCBs); and (c) hepta-, octa-, and nonachlorobiphenyls (HeCBs, OCBs, and NCBs).

$$2C_{12}H_7Cl_3 + 2Cl_2 \rightarrow 2C_{12}H_6Cl_4 + 2HCl \quad (1e)$$

$$2C_{12}H_6Cl_4 + 2Cl_2 \rightarrow 2C_{12}H_5Cl_5 + 2HCl \quad (1f)$$

$$2C_{12}H_5Cl_5 + 2Cl_2 \rightarrow 2C_{12}H_4Cl_6 + 2HCl \quad (1g)$$

The Varimax rotation method, a commonly used orthogonal rotation method, was selected during the analysis. ~~If the numbers of the both compounds and samples in the data set less than 50% of the total data, these compounds and samples were excluded from the analysis.~~ Furthermore, prior to the analysis, the values lower the minimum detection limit (MDL) were replaced with half of MDL (Shyu et al. 2011).

For the PCBs emission in the ambient air, three factors were extracted out (Table S1). Factor-I contributed 49.762% of the total variance and was correlated with congeners 8 + 5, 21 + 33 + 53, 77 + 110, 82 + 151, 85, 105, 118, 132, 134, 136, 137 + 130 + 176, 138 + 158, 141, 153, 178 + 129, 180 + 193, 196 + 203, and 198, identified as anthropogenic sources associated with the use of commercial Aroclor mixtures (Ikonomou et al. 2002). Factor-II, which accounted for 26.658% of the total variance, was highly correlated with congeners 1, 28 + 31, 56 + 60, 89, 101, 174, 177, 183, 185, 202 + 171 + 156, 201, and 206, which mainly originate from coal and wood combustion (Dumanoglu et al. 2017). Furthermore, most higher chlorinated congeners 151, 135, 144, 141, 179, 182, 187, 183, 174, 181, 177, 180, 193, 170, 190, 199, 203, 196, and 194, are also accepted to correlate with Aroclor 1260 (Jin et al.

2012). Therefore, Factor-II would represent mixed sources that originate both from wood and coal combustion and Aroclor 1260 uses. Factor-III accounted for 23.98% of the total variance, related to congeners 81 + 87, 44, 47 + 48, and 62, which are associated with combustion and industrial thermal processes (Ikonomou et al. 2002; Mao et al. 2019).

In the case of road dust samples (Table S1), Factor-I ~~contributing~~ 33.388% of the total variance and was strongly loaded with congeners 21 + 33 + 51, 25, 26, 28 + 31, 37 + 42, 41 + 64 + 71, 44, 45, 46, 47 + 48, 56 + 60, 62, 63, 66 + 95, 70 + 76, 74, 97, 100, and 101. The main ones, 49, 52, 28, 44, 101, 110, and 118, are emitted from wood and coal combustions (Dumanoglu et al. 2017; Lee et al. 2005). Factor-II (17.616%) mostly correlated with higher congeners: 16 + 32, 22, 82 + 151, 132, 136, 137 + 130 + 176, 141, 146, 153, 177, 183, and 185, some of which are the predominant components of Aroclor mixtures-1260 and 1254 (Du et al. 2008). Factor-III (14.452%) was strongly loaded with congeners 1, 3, 6, 7 + 9, 8 + 5, 17, 19, and 89. Congeners 10, 4, 15, and 17 are associated with Aroclor-1248 (Jin et al. 2012). The last factor, which explained 13.103% of the total variance, was found to be highly loaded with congeners: 18, 24, 29, 49, 77 + 110, 83, and 163. The planar congener 77 contamination ~~to ambient attributable~~ is likely due to commercial formulations (Tanabe et al. 1987). Therefore, the pattern of the road dust data was found to be close to emissions originated from the Aroclor mixtures that have been widely used in electrical transformers, dielectric fluids, heat transfer and hydraulic systems, paints, polymers, lubricants, plasticizers, fire retardants, immersion oils, sealants, and caulking compounds (Kodavanti et al. 2017; Anh et al. 2019).

For the pond sediment contaminations (Table S1), Factor-I accounted for 42.499% of the total variance, and was composed of

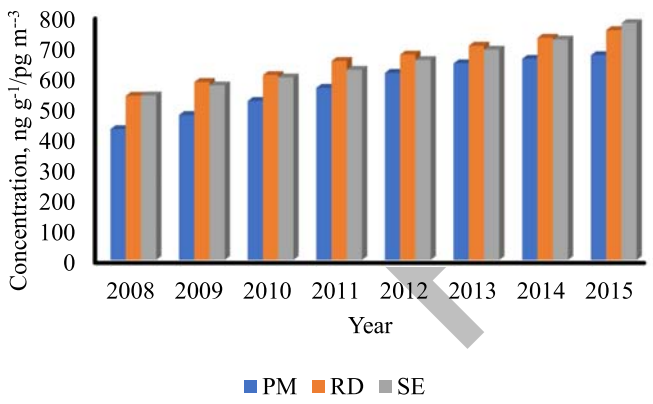

Fig. 4. Temporal variation of PCBs concentration in the particulate matters (PM), road dust (RD), and sediment (SE).

congeners 28 + 31, 29, 41 + 64 + 71, 44, 46, 49, 62, 66 + 95, 70 + 76, 83, and 101. Hence, it would be related to combustion processes, as congeners 49, 28, 44, and 101 are mainly originated from coal and wood combustion (Dumanoglu et al. 2017). Factor-II, which explained 19.628% of the data variance, included congeners 8 + 5, 16 + 32, 21 + 33 + 53, 24, 26, 37 + 42, 44, 45, and 47 + 48. ~~The~~ congeners 44, 47, 51, and 68 are a tracer of the combustion and industrial thermal process. Especially, municipal waste incinerators have an important effect on the emissions of these congeners in the atmosphere (Ikonomou et al. 2002; Mao et al. 2019). Therefore, Factor-II was associated with ~~the~~ industrial thermal process. Factor-III explained 12.531% of the total variance, and was loaded with higher chlorinated PCBs, including 1, 6, 7 + 9, 18, 74, 135 + 144, 138 + 158, and 141. Higher chlorinated PCBs, 135, 141, 144, and 138, mostly originate from Aroclor 1260 (Jin et al. 2012). Therefore, the contribution of this factor may be attributed to the use of Aroclor 1260.

Comparison with PCBs Concentrations Reported for Other Regions

A comparison with PCBs environmental contamination in various sites around the world is presented in Table 5. Total PCBs concentrations in the ambient air and particulate phase reported for Atlantic Ocean, India coastal area, East Africa, Turkey, France, Pohang (Korea), Karachi and Lahore, Panjab (Pakistan), Kanpur (India), London, and Yokohama (Japan) were found to be in the range: 4–220, 216–1,077, 65.6–244 pg m^{-3}, 52–293 ng m^{-3}, 10–270, 15–166, 48–61, 34–389, 254–432, 1,000–2,000 and 62–250 pg m^{-3}, respectively (Arinaitwe et al. 2018; Goel et al. 2016; Sakin and Tasdemir 2016; Nasir et al. 2014; Syed et al. 2013; Baek et al. 2010; Gioia et al. 2008; Zhang et al. 2008; Blanchard et al. 2006; Kim and Masunaga 2005). Hence, the values reported herein would be among the highest (comparable to those from India and Pakistan).

With regard to the total concentrations of PCBs in the dust, electronic waste, and sediments reported in other locations: Guangzhou and Hong Kong, Chennai, Northern Vietnam, Nakdong River (Korea), San Diego Bay, China, and Delhi (Anh et al. 2019, Chakraborty et al. 2016, Jin et al. 2012, Kumar et al. 2011, Li et al. 2012, Neira et al. 2018, Wang et al. 2013), it ranged from 4.02 to 114 ng kg^{-1}, from 1.6 to 53, from 0.25 to 14, from 0.124 to 79.2, from 23 to 1,387, and from <0.01 to 99.40 ng g^{-1}, respectively. Again, the values detected in this study would be among the highest reported in the literature.

Thus, the values found for Raipur would be higher than those found in Korea, China, and Delhi, and would only be exceeded by those detected in San Diego Bay, California.

Table 4. Correlation coefficient, r ($p = 0.05$), of PCBs in ambient air particulates

Type	MCBs	DCBs	TCBs	TeCBs	PeCBs	HCBs	HeCBs	OCBs	NCBs
MCBs	1.00								
DCBs	0.51	1.00							
TCBs	0.91	0.42	1.00						
TeCBs	0.79	0.91	0.62	1.00					
PeCBs	0.65	0.89	0.72	0.84	1.00				
HCBs	0.27	0.83	0.42	0.60	0.90	1.00			
HeCBs	0.93	0.76	0.90	0.89	0.89	0.61	1.00		
OCBs	0.86	0.64	0.96	0.74	0.89	0.66	0.96	1.00	
NCBs	0.00	0.82	−0.18	0.61	0.50	0.61	0.25	0.08	1.00

Note: MCBs = monochlorobiphenyls; DCBs = dichlorobiphenyls; TCBs = trichlorobiphenyls; TeCBs = tetrachlorobiphenyls; PCBs = pentrachlorobiphenyls; HCBs = hexachlorobiphenyls; HeCBs = heptachlorobiphenyls; OCBs = octachlorobiphenyls; and NCBs = nonachlorobiphenyls.

Table 5. Comparison of PCB contamination

Location	Sample type	Concentration	Reference
London and Manchester, UK	Ambient air	1,000–2,000 pg m^{-3}	Jaward et al. (2004)
Coastal area, India	Ambient air	216–1,077 pg m^{-3}	Zhang et al. (2008)
Uludag University, Turkey	Ambient air	293 ± 257 and 52 ± 56 ng m^{-3}	Sakin and Tasdemir (2016)
Lake Victoria, East Africa	Air	65.6–244 pg m^{-3}	Arinaitwe et al. (2018)
Yokohama, Japan	PM	62–250 pg m^{-3}	Kim and Masunaga (2005)
France	PM	10–270 pg m^{-3}	Blanchard et al. (2006)
Atlantic Ocean	PM	4–220 pg m^{-3}	Gioia et al. (2008)
Pohang, Korea	PM	15–166 pg m^{-3}	Baek et al. (2010)
Panjab, Pakistan	PM	34–389 pg m^{-3}	Syed et al. (2013)
Karachi, Lahore, Pakistan	PM	48–61 pg m^{-3}	Nasir et al. (2014)
Kanpur, India	PM	254–432 pg m^{-3}	Goel et al. (2016)
Guangzhou and Hong Kong	Outdoor dust	4.02–228 ng kg^{-1} 7.75–114 ng kg^{-1}	Wang et al. (2013)
Chennai, India	Electronic waste: waste dismantling sites, nearby highways, sub urban and industrial site	53, 3.6, 1.7 and 1.6 ng g^{-1}	Chakraborty et al. (2016)
Northern Vietnam	Street dust	14, 11 and 0.25 ng g^{-1}	Anh et al. (2019)
Nakdong River, Korea	Sediment	0.124–79.2 ng g^{-1}	Jin et al. (2012)
San Diego Bay, Shelter Island Yacht Basin (SIYB), Harbor Island West (HW) and Harbor Island East (HE)	Sediment	23–153, 31–294, and 151–1,387 ng g^{-1}	Neira et al. (2018)
China	Sediment	126.49 ng g^{-1}	Li et al. (2012)
Delhi	Soil	<0.01–99.40 ng g^{-1}	Kumar et al. (2011)
Raipur	PM	248 ± 152 pg m^{-3}	Present work
	Road dust	241 ± 146 ng g^{-1}	
	Sediment	328 ± 99 ng g^{-1}	

These findings corroborate those reported by Chakraborty et al. (2013) in a study on atmospheric PCBs levels (gaseous and particulate phase) in ~~the~~ Indian cities: New Delhi, Agra, Kolkata, Mumbai, Goa, Chennai, and Bangalore. In view of the detected concentrations, the authors urge increasing control over the release of PCB sources in India and ask for measures to protect human health and the environment.

Conclusions

Very high concentrations of PCBs (mainly congeners 1 and 3) were detected in Raipur area: 143 pg m^{-3} (365 ng g^{-1}), 194, and 299 ng g^{-1} for particulate matter, road dust, and sediments, respectively. These contents are among the highest ever reported in the literature, and clearly exceeded the recommended value of 60 ng g^{-1}. The normalized total mean TEFs for the PM, road dust, and sediments from Raipur city were estimated at 0.00066, 0.00058, and 0.00047, respectively. In view of the correlations with particulate matter, elemental carbon, organic carbon, and carbonate carbon, massive coal burning and vehicular emissions in Chhattisgarh region can be ascribed as the main sources of PCB pollution. Temporal evolution, tracked over an eight-year period, showed an average annual increment rate of approximately 5.4% in PCBs concentration, while vertical profile analyses showed substantial PCBs concentrations ~~at~~ deeper sediments. Industrial uses, and coal and biomass combustion were apportioned as the major sources of PCBs contamination in the studied area. The collected data points to a dramatic situation, which calls for urgent action to meet the Stockholm Convention goals.

Data Availability Statement

All data, models, and code generated or used during the study appear in the published article.

Acknowledgments

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Supplemental Materials

Table S1 is available online in the ASCE Library (www.ascelibrary.org).

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