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Distribution of Persistent Organochlorine Pesticide Residues in Gomti River, India

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Organochlorine pesticides (OCPs) have aroused global concern due to their long persistence, low biodegradability and wide range distribution in the environment (Burkow and Kallenborn 2000, Koziol and Pudykiewicz 2001). Recently, these have been classified as persistent toxic substances (UNEP 2002). Organochlorine pesticides are mainly used to control the soil and crop pests in agricultural fields. Pesticides find their way into aquatic systems through discharges of domestic sewage and industrial wastewater, runoff from agricultural fields and direct dumping of wastes into river systems. Combination of their physico-chemical properties such as low aqueous solubility, moderate vapour pressure and octanol-water partition coefficient and persistence in the environment make them capable of long-range transport. In India, the concentration of chlorinated pesticides have been detected in almost all the segments of environment due to their extensive use in past, which have shown potential to biomagnify/accumulate in animal tissue, human blood, adipose tissue and breast milk (Beg et al. 1989). However, most of these pesticides have now been banned in the country (UNEP 2002). Since the pesticides are lipid soluble in nature, cumulative accumulation of low concentrations of these in the body fat of mammals might pose potential hazards in the long run (Metcalf 1997). Endosulfan is very toxic to aquatic organisms (96-hr LC_{50} of 1.5 μg L^{-1} for rainbow trout), lindane is moderately toxic for invertebrates and fish with LC_{50} of 20-90 μg L^{-1}.The reported half-life for aldrin is about 20-100 d, for DDT 1.1-3.4 yr, for lindane and other BHC-isomers generally greater then 1 and 2 yr, respectively, and for endosulfan about 50 d. DDT exhibits high bioconcentration factors (50,000 for fish and 500,000 for bivalves). DDT has been shown to have an estrogen-like activity and possible carcinogenic activity in humans (UNEP 2002). DDT may be metabolized easily to DDE and DDD in the environment and its derivatives are more stable and persistent than parent molecule of DDT (Bossi et al. 1992). In spite of large-scale deposition from agricultural sprays and runoffs (UNEP 2002), the information on pesticides in river sediments is scarcely available. This study was undertaken to examine the levels and distribution patterns of some selected organochlorine pesticides in river system in India.

MATERIALS AND METHODS

The Gomti river, one of the major tributaries of the river Ganga originates from a

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natural reservoir in the swampy and densely forested area (Miankot, elevation of about 200m; North latitude 28°34' and East longitude 80°07') in Pilibhit district of Uttar Pradesh, India. The soil sediments here are silty sands. The river flowing through the districts of Pilibhit, Shajahnpur, Sitapur, Lucknow, Barabanki, Sultanpur, Jaunpur and Ghazipur in Uttar Pradesh drains a catchments area of about 25,000 km^2 and traverses a total distance of about 730 km before finally merging with the Ganga river in Ghazipur district about 30 km north of Varanasi. Throughout its stretch, there are a few small tributaries (Kathna, Sarayan, Reth, Kalyani and Sai) originating within short distances and carrying the wastewater and industrial effluents from different towns and industrial units in the basin. On the banks of the river, Lucknow, Sultanpur and Jaunpur are the three major urban settlements. The river serves as one of the major source of drinking water for the Lucknow City, the State capital of Uttar Pradesh with a population of about 3.5 million. The river, subsequently, receives the untreated wastewater and effluents from Lucknow, Jagdishpur, Sultanpur and Jaunpur directly in its course through more than 40 wastewater drains. The study area covers 8 different locations namely Neemsar, Bhatpur, Gaughat, Mid-Lucknow, Pipraghat, Barabanki, downstream of Sultanpur and Jaunpur on the river spread over about 500 km stretch. Grab samples of water and bed sediments were collected from each of the locations between March-1996 and Feb-1999. Water samples were analysed each month while the sediments samples were analysed once in each season viz. summer, monsoon and winter during the study period. The sediment samples were collected from three points (1/4,1/2 and 3/4) across the river width at each of the eight locations using Ekman-dredge sediment sampler. The samples collected in the polyethylene bags were transported to the laboratory in icebox under low temperature conditions. In the laboratory, pebbles, shells and vegetable matter were removed and the samples were air-dried. The air dried samples then ground with pestle-mortar and sieved to 200 BSS mesh size. The organic carbon content of the sediment was determined using back titration method (Walkely and Black 1934). For organochlorine pesticides analysis 5.0 ± 0.05 g sediment sample from each location were soxhlet extracted with 50 mL of n-hexane for 6 hr. The extracts were demoisturised using anhydrous granular sodium sulphate and concentrated in a rotary evaporator making up final volume to 2 mL. The samples were stored in dark at 4°C till the analysis was performed. The river water samples collected from midstream of the river (30 cm below the surface) in brown glass bottles were transported to the laboratory under ice conditions. For organo-chlorine pesticides analysis, 1L of the unfiltered water sample from each location was triply extracted by liquid-liquid extraction in a separatory funnel using 50, 30 and 30 mL of n-hexane. The combined solvent extracts were demoisturised using anhydrous granular sodium sulphate and concentrated in a rotary evaporator to a final volume of 2 mL. All the sediment and water samples were analysed for selected organochlorine pesticides viz. aldrin, BHC isomers, DDT isomers/meta-bolites and endosulfan on Antek-3000 Gas Chromatograph equipped with Ni^{63} ECD. For all extractions GC grade n-hexane (Spectrochem, India; 99 %) was used. The pesticides standards (99.9% purity) were supplied by Sigma-Aldrich, USA. The minimum detection limit for aldrin, α-BHC β-BHC, γ-BHC, δ-BHC,

op-DDT, pp-DDT, pp-DDD, pp-DDE and endosulfan was found to be 3.6, 3.4, 8.9, 3.1, 2.8, 9.6, 5.7, 6.9, 9.2 and 3.5 ng L^{-1}, respectively Further lower detection was achieved through 500 fold concentration of the samples All analysis were carried out in duplicate and the recoveries of individual pesticides were determined through spiked sample method, which were found between 98-99.5%. Recovery correction factors were applied to the final results.

RESULTS AND DISCUSSION

Concentration of organochlorine pesticides (aldrin, BHC-isomers, DDT and its metabolites, and endosulfan) in water and sediments of the Gomti river, during different seasons of the study period (March-1996 to Feb-1999) are summarized in Table 1 and 2. In Gomti river, no seasonal trend was observed in the distribution of the pesticides, however, the higher levels were found in the monsoon season with a highest concentration of 4997.0 ng L^{-1} in water and 1765.0 ng g^{-1}dw in the sediments. The concentration of individual organochlorine pesticides in water and sediments of the Gomti river at different locations are presented in Table 3 and 4.

In the present study, the levels of total organochlorine pesticides in the river water (0.02-4997.0 ngL^{-1}) are comparatively higher than the levels (214.4-1819.0 ngL^{-1}) reported by Zhang et al. (2003) in the Minjiang river estuary. Similar to the findings of the present study, Wolska et al.(2003) also reported the total organochlorine pesticides concentration from BDL to 5100.0 ng L^{-1} in the Odra river system. In sediments of the Gomti river, the total organochlorine pesticides concentrations (0.2-1765.4 ng g^{-1}dw) are relatively higher than those reported in the Erh-jen river (0.6-23.7 ng g^{-1}dw; Doong et al. 2002), Da-han river (0.2-14.8 ng g^{-1}dw; Doong et al. 2002), Minjiang river estuary (28.8-52.1 ng g^{-1}dw; Zhang et al. 2003) and Odra river (BDL to 33.0 ng g^{-1}dw; Wolska et al. 2003). The detection frequency of different organochlorine pesticides in the river water and sediments are shown in the Fig. 1a. Aldrin was the most scarcely distributed pesticide, whereas, α-BHC was the most frequently occurred one following by other BHC-isomers, both in the river water and sediments. In the Gomti river, aldrin ranged between 0.01 to 205.0 ng L^{-1} in water and 8.2 to 17.0 ng g^{-1}dw in the sediments. Endosulfan was found in the range 0.2 to 1372.0 ng L^{-1} in the water and 4.8 to 70.6 ng g^{-1}dw in the sediments of the river. The levels of total BHC ranged from 0.02–4846.0 ng L^{-1} in the water and 0.1–1650.0 ng g^{-1}dw in the sediments of the river. In water of Gomti river, the total BHC concentration (sum of α-BHC, β-BHC, γ-BHC, and δ-BHC) is relatively higher than the total BHC concentration found in Minjiang river estuary (52.1-515.0 ng L^{-1}; Zhang et al. 2003), Mar Menor Lagoon (30.0-300.0 ng L^{-1}; Perez et al. 2000). However, these values are lower than that of Ganga river (BDL-99517.0 ng L^{-1}; Nayak et al. 1995) and water supply of El-Haram (20700.0-86200.0 ng L^{-1}; Kabbany et al. 2000). Total BHC concentration in the sediments were higher than those found in the sediments of Minjiang river estuary (2.99-16.21 ng g^{-1}dw; Zhang et al. 2003). Several studies indicated that concentration of BHC isomers is a world-wide problem (Fellin et al.1996, Doong et al. 2002, Zhang et al. 2003). Among the

Table 1. Concentration range of OCPs in Gomti river water (ng L^{-1}) during different seasons, March, 96 to Feb, 99.

Pesticide	Sum-96	Mon-96	Win 96-97	Sum-97	Mon-97	Win 97-98	Sum-98	Mon-98	Win 98-99
Aldrin	BDL	BDL-0.7	0.3-205.0	0.03-116.0	0.3-62.0	BDL	0.3-4.0	BDL	BDL
α BHC	0.1-41.0	0.3-118.0	0.3-37.0	0.4-33.0	0.3-48.0	0.3-49.0	0.3-93.0	0.01-52.0	0.4-47.0
β BHC	0.1-95.0	7.0-2128.0	17.0-657.0	8.0-75.0	18.0-4821.0	35.0-4420.0	2.0-369.0	0.02-373.0	36.0-577.0
γ BHC	0.3-6.0	1.0-42.0	1.0-27.0	0.1-25.0	0.1-778.0	2.0-59.0	0.03-52.0	0.01-0.3	0.2-59.0
δ BHC	3.0-442.0	0.3-497.0	1.0-88.0	9.0-301.0	6.0-30.0	BDL	7.0-266.0	0.3-7.0	BDL
Σ BHC	10.0-443.0	4.0-2137.0	1.0-721.0	0.4-347.0	107.0-4846.0	1.0-627.0	1.0-586.0	0.02-375.0	1.0-613.0
op-DDT	BDL	35.0-165.3	BDL-93.0	2.0-3825.0	BDL-187.0	BDL	9.0-1324.0	BDL	BDL
pp-DDT	BDL	BDL-4.7	BDL-93.0	BDL-27.0	BDL	BDL	3.0-1089.0	BDL	BDL
pp-DDD	BDL	BDL	1.0-38.0	BDL-484.0	1.0-2.0	2.0-17.0	2.0-242.0	1.0-2.0	2.0-4.0
pp-DDE	1.0-2.0	1.0-106.7	3.0-21.0	1.0-194.0	0.2-4.0	BDL-2.0	0.1-72.0	BDL-0.3	BDL-1.7
Σ DDT	BDL-2.0	1.0-125.3	1.0-244.0	1.0-4578.0	0.2-151.0	3.0-21.0	1.0-1337.0	BDL-0.3	3.0-5.0
Endo	1.0-3.0	0.3-667.0	0.3-390.0	1.0-15.0	BDL-0.3	BDL	0.3-1372.0	BDL	BDL

Sum=Summer, Win=Winter, Mon=Monsoon; BDL=Below Detection Limit ; Endo= endosulfan

Table 2. Concentration range of OCPs in Gomti river sediments (ng g^{-1} dw) during different seasons, March, 96 to Feb, 99.

Pesticide	Sum-96	Mon-96	Win 96-97	Sum-97	Mon-97	Win 97-98	Sum-98	Mon-98	Win 98-99
Aldrin	BDL	BDL	BDL	BDL	BDL	BDL-8.2	BDL	BDL	BDL-17.0
α BHC	BDL	1.9-6.1	1.1-30.9	0.1-46.6	0.3-24.2	0.1-26.7	0.1-49.4	0.3-27.2	0.2-153.7
β BHC	15.5-42.2	BDL-2.0	13.0-565.0	1.5-6.0	26.5-1620.0	BDL-159.2	4.9-5.4	25.3-243.2	BDL
γ BHC	BDL	BDL-1.6	BDL-448.0	14.8-36.1	0.2-12.2	0.2-0.4	19.0-36.6	0.4-13.7	BDL-0.4
δ BHC	BDL	3.0-70.9	BDL	BDL	BDL	0.9-9.2	BDL-11.3	BDL	1.0-172.5
Σ BHC	15.5-42.2	3.2-73.1	1.0-1024.0	0.1-82.6	27.2-1650.0	0.3-177.0	0.1-97.3	26.1-273.7	0.3-326.0
op-DDT	BDL	BDL	BDL	BDL	1.1-506.8	BDL	BDL	BDL	BDL-21.7
pp-DDT	BDL	BDL	41.0-126.0	BDL	13.5-59.6	11.0-21.0	BDL	BDL	BDL-11.8
pp-DDD	BDL	BDL-28.6	BDL-15.4	BDL	0.7-27.5	5.7-8.5	BDL	BDL	2.9-5.6
pp-DDE	BDL	BDL	BDL	BDL	0.2-22.2	1.8-19.0	BDL	0.2-32.5	10.4-28.2
Σ DDT	BDL	BDL-31.9	17.0-126.0	BDL	0.2-509.0	5.3-27.4	BDL	0.2-32.5	5.6-13.2)
Endo	4.8-12.6	24.9-72.6	BDL	BDL	BDL	18.0-24.5	BDL	BDL	BDL-22.8

Sum=Summer, Win=Winter, Mon=Monsoon; BDL=Below Detection Limit ; Endo= endosulfan

Table 3. Concentration range of OCPs in Gomti river water (ng L^{-1}) at different locations during March, 96 to Feb, 99.

Pesticide	Neemsar	Bhatpur	Gaughat	Mid-Lucknow	Pipraghat	Barabanki	Sultanpur	Jaunpur
Aldrin	0.03-62.0	0.3-2.0	1.0 –3.0	BDL-1.0	1.0-205.0	0.3-38.0	0.2-4.0	0.7-16.0
α -BHC	0.01- 48.0	0.1-52.0	0.1-36.0	0.2-49.0	0.3-93.0	0.3-118.0	0.3-41.0	0.3-39.0
β -BHC	0.02-2569.0	0.2-4821.0	0.1-2266.0	0.2-2048.0	0.2-1005.0	0.3-1972.0	0.1-1242.0	0.2-3674.0
γ -BHC	0.01-7780.0	0.01-283.0	0.04-52.0	0.04-59.0	0.04-39.0	0.02-42.0	0.5-38.0	0.03-240.0
δ -BHC	3.0-132.0	0.3-132.0	0.3-150.0	9.0-266.0	7.0-442.0	3.0 –461.0	8.0-462.0	9.0-497.0
Σ BHC	0.02-2586.0	0.1-4846.0	1.0-2276.0	1.0-2063.0	1.0-1008.0	3.0-1998.0	0.3-1249.0	0.4-2183.0
op-DDT	215.0-573.0	4.0-359.0	23.0-584.0	9.0-35.0	2.0-93.0	26.0-3825.0	76.0-232.0	148.0-1324.0
pp-DDT	51.0-221.0	27.0-244.0	4.7-400.0	3.0-54.0	15.0-1089.0	BDL-14.0	BDL-35.0	BDL-11.0
pp-DDD	14.0-35.0	BDL	BDL-3.0	BDL-17.0	2.0-242.0	1.0-484.0	2.0-10.0	BDL-2.0
pp-DDE	0.2-72.0	0.3-36.7	0.3-105.3	0.1-106.7	1.0-21.0	0.3-194.0	0.3-10.0	1.0-3.0
Σ DDT	0.2-834.0	7.0-567.0	0.3-997.0	3.0-119.0	1.0-244.0	1.0-4578.0	0.3-242.0	1.0-1337.0
Endo	0.3-2.0	0.3-15.0	1.0-27.0	1.0-26.0	3.0-1372.0	2.0-317.0	2.0-329.0	0.3-667.0
Σ OCPs	0.02-2586.0	0.1-4997.0	1.0-2276.0	0.6-2063.0	BDL-1385.0	6.0-4926.0	0.3-1225.0	2.0-1783.0

BDL=Below Detection Limit ; Endo= endosulfan

Table 4. Concentration range of OCPs in Gomti river sediments (ng g^{-1} dw) at different locations during March, 96 to Feb, 99.

Pesticide	Neemsar	Bhatpur	Gaughat	Mid-Lucknow	Pipraghat	Barabanki	Sultanpur	Jaunpur
Aldrin	BDL	BDL	BDL	BDL-8.2	BDL17.0	BDL	BDL	BDL
α -BHC	0.2-3.2	0.1-30.9	0.1-10.9	3.2-49.4	4.9-153.7	0.5-17.2	0.2-2.0	0.1-17.1
β -BHC	104.6-518.0	5.4-501.6	4.7-565.1	2.0-311.8	1.5-1620.3	13.0-55.6	26.5-42.2	39.8-129.1
γ -BHC	2.6-2.7	BDL	0.2-448.2	1.6-36.6	12.2-18.7	0.2-0.4	0.3-0.4	BDL-0.2
δ -BHC	BDL	BDL-21.8	BDL-70.9	3.0-70.9	1.9-172.5	BDL	BDL-16.7	1.0-17.2
Σ BHC	0.2-523.6	0.3-503.6	0.2-1024.2	9.8-339.2	10.9-1650.0	0.5-57.1	0.2-42.2	0.1-134.9
op-DDT	BDL	BDL	BDL	BDL	BDL-1.1	BDL-21.7	BDL	BDL
pp-DDT	BDL	BDL	10.8-11.8	13.5-126.0	BDL-59.6	21.0-41.0	BDL	BDL
pp-DDD	BDL-0.7	BDL	BDL	2.9-8.5	3.1-28.6	BDL	BDL	BDL-5.6
pp-DDE	2.4-3.4	1.2-1.8	0.4-0.5	2.6-19.1	28.2-32.5	0.2-0.2	0.3-0.4	1.8-3.1
Σ DDT	3.4-3.5	1.4-1.8	0.5-11.8	12.4-126.2	10.5-115.9	0.2-41	BDL-0.4	3.1-509.2
Endo	BDL-49.3	BDL-72.6	BDL-56.1	7.8-24.5	10.3-39.0	BDL-12.6	BDL-12.8	12.6-24.9
Σ OCPs	0.2-527.1	0.3-504.9	4.8-1024.0	9.8-368.1	24.7-1765.4	16.2-57.3	0.2-54.9	0.1-640.1

BDL=Below Detection Limit ; Endo= endosulfan

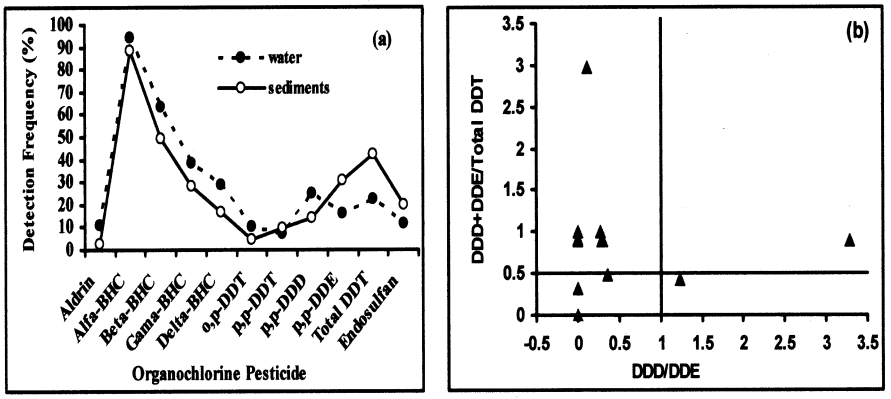

Figure 1. (a) The detection frequencies of OCPs in river water and sediments, and **(b)** relationship between DDD/DDE and (DDE+DDD)/∑DDT in sediments of Gomti river.

isomers of BHC, α-BHC was the most widely distributed showing detection frequency 89 and 94% in the sediments and water, respectively, with concentrations ranging from 0.01 to 118.0 ng L^{-1} in water and 0.1 to 153.7 ng g^{-1} dw in the sediments. γ-BHC ranged from 0.01 to 778.0 ng L^{-1} and 0.2 to 448.0 ng g^{-1}dw in the water and sediments of the river, respectively. The δ-BHC was observed in the range of 0.3-497.0 ng L^{-1} in water and 1.0-172.5 ng g^{-1}dw in the sediments of the river. However, the levels of β-BHC were comparatively higher than other isomers ranging from 0.02–4846.0 ng L^{-1} in water and 1.5–1620.0 ng g^{-1}dw in the sediments of the river. In terms of individual components of the BHC, on an average, β-BHC accounted for 82 and 78% of the total BHC concentration in the water and sediments of the river, respectively. Zhang et al. (2003) have also reported that β-BHC accounted 67% of total BHC concentration in water and 50% of total BHC concentration in the sediments of the Minjiang river estuary. The higher concentration of β-BHC in river may be due to the erosion of the weathered agricultural soils containing BHC compounds. Further, wide distribution of α-BHC and higher levels of β-BHC in the river may be attributed to the tendency and potential of α-BHC for transport to a long distance, and resistance of β-BHC to hydrolysis and environmental degradation (Willett et al. 1998). In Gomti river the detection frequency of BHC compounds showed that the contamination of BHC is widespread in Gomti river. Total DDT concentration was in the range of 0.2–4578.0 ng L^{-1} in the river water, which is higher than that found in Minjiang river estuary (40.6-233.5 ng L^{-1}) reported by Zhang et al. (2003), and lower than that of reported in the water supply of El-Haram (2300.0-61000.0 ng L^{-1}; Kabbany et al. 2000) and Ganga river (135.0-66516.0 ng L^{-1}; Nayak et al. 1995). In the present study, the total DDT concentration ranged from 0.2 to 509.0 ng g^{-1} dw in the sediments which were found higher (1.6-13.1 ng g^{1} dw) in the sediments of Minjiang river estuary (Zhang et al. 2003). Among the DDT metabolites, op-DDT, pp-DDT, pp-DDD and pp-DDE ranged from 2.0-3825.0; 3.0-1089.0; 1.0-484.0 and 0.2-194.0 ng L^{-1} in the water and 1.1-21.7;

3825.0; 3.0-1089.0; 1.0-484.0 and 0.2-194.0 ng L^{-1} in the water and 1.1-21.7; 10.8-126.0; 0.7-28.6 and 0.2-32.5 ng g^{-1}dw in the sediments, respectively. The higher concentrations of DDT metabolites in the river water may be attributed to the particulate matter present in the unfiltered water samples. In the Gomti river, pp-DDD was the most frequently detected metabolite of DDT in the water (detection frequency 25%), while in sediments pp-DDE occurred most frequently (detection frequency 31%). The relative concentrations of the parent DDT compounds and its metabolites can be used as indices for assessing the contamination source. Since, DDT can be biodegraded under aerobic conditions to DDE and under anaerobic condition to DDD, ratio of (DDE+DDD)/∑DDT >0.5 is considered to be subjected to long-term weathering (Hites and Day 1992; Doong et al. 2002). The ratios of DDD/DDE >1.0 indicates the anaerobic degradation of the pp-DDT. Fig. 1b illustrates the relationship between (DDE+DDD)/∑DDT and DDD/DDE in the sediments of the river. Ratios of (DDE+DDD)/∑DDT were in the range of 0.3-3.0. However, the ratios of DDD/DDE ranged from 0.1 to 3.3 with most values lesser than unit. These results indicate that source of DDT contamination is from the aged and weathered agricultural soils with signature of recently used DDT in Gomti river catchments. India has banned DDT for agricultural use in 1989, although, it is used in public health sectors for Malaria control (UNEP 2002). Due to affinity of these compounds to particulate matter, river and marine sediments are thought to be the major sinks for them (Doong et al. 2002). Relatively high concentration of organochlorine pesticides in the sediments of river as compared to water may be due to the high organic contents (0.1-21.4%) in the sediments of the river. The contamination of hydrophobic organic compounds in sediments is dependent on the chemical properties of the ecosystem, the partition coefficients of individual compounds and the organic contents of sediment particles (McKenzie-Smith et al. 1994; Glynn et al. 1995). The total OCPs concentration in the sediments of Gomti river were highly correlated (R=0.81) with the organic carbon content during the summer season, whereas, very poor correlation was found during the monsoon (R=0.16) and winter seasons (R=0.01). This variation may be due to the relative seasonal changes. Heavy river-flow during the monsoon results in the erosion of riverbed sediments and its re-suspension into the river water column and the pollutants sorbed on the sediment particles may be diluted by runoff. Further, poor correlation between total OCPs concentrations and organic carbon content in the winter season may be due to the residual effect of the monsoon, which (July-October) is followed by the winter season (November-February). During the winter season water flow becomes normalized and river system starts to be stabilized with the settling of eroded riverbed sediments. In the summers (March-June), the river system is under the complete stabilized conditions.

The analysis of results revealed that neither temporal nor spatial distribution pattern was found, perhaps due to the presence of multiple and aleatory sources. Gomti river receives intermittent inputs of organochlorine pesticides and BHC isomers, which are the main contributors to the pesticide pollution of the Gomti river. The presence of DDT-metabolites in the river indicates continuous use of

DDT in the catchments. It is notable that in India aldrin is completely banned while the usage of DDT and BHC are restricted (UNEP 2002).

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