Organochlorine pesticide residues in drinking water in the rural areas of Haryana, India

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Abstract Drinking water samples collected from rural areas of three districts of Haryana during pre-monsoon and post-monsoon periods were analysed for the presence of organochlorine pesticide residues. The main source of drinking water in rural areas, i.e. groundwater in Ambala and Gurgaon districts and surface water supply in Hisar district, was found to be contaminated with isomers of HCH and endosulfan and metabolites of DDT, whereas dieldrin remained below detection limits. During the study period, the mean values observed for total HCH, DDT and endosulfan were 87.6, 848.2, and 27.4 ng/L and 99.8, 275.3 and 164.2 ng/L, respectively, for Ambala and Gurgaon districts. In the case of Hisar district, the values

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Present Address: H. R. Sharma Department of Environmental Health, University of Gondar, P.O. Box No. 196, Gondar, Ethiopia e-mail: hrsharma74@yahoo.co.in, sharmahardeeprai@gmail.com were 78.5, 115.9, and 53.0 ng/L, respectively. During the study period, 37% of the samples exceeded the total pesticide level of 500 ng/L indicated in the EECD directive for drinking water. Seasonal variations of pesticide residues were also observed during the study period.

Keywords Organochlorines • Pesticide residues • Groundwater • Surface water • Drinking water • Rural areas

Introduction

Organochlorine pesticides (OCPs) find their use in agriculture against pests. These are also used in household, garbage heaps, waste disposal sites, sewers, godowns, etc. Their use increases many folds during natural calamities, fairs and outbreaks of epidemics. These pesticides may enter into ground and surface water through diffused and point sources. Large intake of water contaminated with low levels of pesticide residues may cause significant effects in consumers (Nair and Pillai 1992), i.e. risk to human health, and alteration of local environment. Lipid solubility and bioaccumulation of low concentrations of OCPs in the body fat of mammals pose potential hazards in the long term.

India is the largest producer of pesticides in South Asia and Haryana state stands in the

third position in India in pesticide consumption (Agnihotri 2000). Haryana state, which is predominantly an agricultural state in India, extends between 27°39'-30°55'N latitude and 74°27'-77°36'E longitude and covers a total geographical area of 442,100 ha, forming about 1.35% of the total area of the country. The population of 21.08 million (Census of India 2001) is distributed in 21 districts. Groundwater from dug wells, hand pumps, tube wells and River Yamuna and some of its canals are the main source of drinking water in different districts of the state. Drinking water from different parts of India has shown the presence of HCH and endosulfan isomers and DDT metabolites (Bakore et al. 2004: Sankararamakrishnan et al. 2005; Singh et al. 2005). Practically limited information is available (Kumari et al. 1996) on the OCPs residues in drinking water in the rural areas of Haryana state where OCPs like DDT and HCH had been extensively used and endosulfan and lindane are still in use.

Thus, the present study was undertaken to assess the OCP levels in drinking water of the rural areas in Ambala, Gurgaon, and Hisar districts of the state known for its intensive agricultural activity in India.

Materials and methods

Selection of the study area

Rural areas of intense agricultural activity (agricultural fields and fallow land) were selected. A total of 19 different sites from different villages, six each from Ambala and Gurgaon and seven in Hisar, were selected for the study (Fig. 1). The districts were selected based on their geographical location, climatic conditions and soil types. These districts are away from each other and cover almost the whole state. The district of Ambala, situated in the northern part of the state, has an area of 1,568.85 km² and a total population of 1,013,660. Hisar, the west central-most district, has an area of 3,787.90 km² and a total population of 1,445,000, and Gurgaon, the southeastern district, has an area of 1,253.07 km² and a population of 870,539 (2001 census). In the district of Ambala, the climatic conditions are different from that of Gurgaon and Hisar. Severe winters with a mean minimum temperature of 5.5°C and very hot summers with a mean maximum temperature of 41.6°C with less rainfall (416 mm) are characteristics of Hisar climate, while high rainfall (1,076 mm) prevails over Ambala district whereas Gurgaon has a somewhat moderate climate as compared to Ambala and Hisar with an annual rainfall of 596 mm (Central Ground Water Board 2007). Udipsamments/Udorthents, loamy sand and Sierozem, arid brown, solonized/desert soils are characteristics of Ambala, Gurgaon and Hisar districts, respectively (Central Ground Water Board 2007).

Water sampling and analysis

The groundwater from hand pumps is the main source of drinking water in Ambala and Gurgaon rural areas, whereas tap water supplied by the Narwana branch of Western Yamuna canal is the main source of drinking water in the rural areas of Hisar. Canal water is usually collected in a reservoir and then supplied to village communities after treatment. A total of 38 samples, in triplicate, were collected on two occasions, i.e. June and October 1999. In Haryana state, monsoon usually extends from July to September. The samples collected in the first and second week of June represented the pre-monsoon/summer season, while the samples of October represented the postmonsoon period. The objective of such sampling was to assess the effect of rains on OCP residues in ground and surface water. The sampling sites for rural drinking water have been selected, keeping in view the possibility of contamination of aquatic bodies due to agricultural runoff. Water samples were collected in the pre-cleaned, ovendried, hexane-rinsed, amber-coloured bottles of 1-L capacity and were sealed with screw caps lined with aluminium foil.

Experimental/chemicals/preparation of extract

The samples were immediately extracted with hexane after these were brought to the laboratory by using conventional liquid–liquid extraction method. Water samples were extracted with hexane, demoisturized with anhydrous Na₂SO₄

Fig. 1 Location of the study areas





and cleaned with alumina column. The samples were analysed on GC equipped with ECD. The samples were analysed using a glass column packed with 1.5% OV-17/1.95% QF-1 on Gas Chrom Q, 100–120 mesh and on an alternate column BP5 capillary 30 m, 0.25 mm I.D. The identities were further confirmed by chemical dehydrochlorination and subsequent gas chromatography as published elsewhere (Kaushik et al. 2010).

Results and discussion

All the samples were analysed in triplicate. Retention time, mean recoveries and minimum detection limits for various pesticides are mentioned in Table 1. Recoveries from fortified samples of water ranged between 80% and 92% for the studied isomers/metabolites and the presented data have been corrected for recoveries. All the ground and surface (canal) water samples were found to be

OCPs	Retention time (min) on glass column	Percent recoveries (%)	Minimum detection limit (ng)
α-HCH	3.83	90	0.5
γ-HCH	4.77	87	0.4
β-ΗCΗ	5.60	92	0.6
δ-ΗCΗ	6.40	85	0.8
o, p'-DDE	10.40	83	0.7
α -Endosulfan	11.50	88	0.3
p, p'-DDE	12.30	86	0.4
Dieldrin	13.11	87	0.14
o, p'-DDT	15.11	82	0.7
o, p'-DDD	15.79	80	0.4
p, p'-DDD	16.15	86	0.5
β-Endosulfan	16.67	87	0.4
p, p'-DDT	17.25	83	0.7
	OCPs α -HCH γ -HCH β -HCH δ -HCH o,p'-DDE α -Endosulfan p,p'-DDE Dieldrin o,p'-DDT o,p'-DDD β -Endosulfan p,p'-DDT	OCPs Retention time (min) on glass column α -HCH 3.83 γ -HCH 4.77 β -HCH 5.60 δ -HCH 6.40 o, p' -DDE 10.40 α -Endosulfan 11.50 p, p' -DDE 12.30 Dieldrin 13.11 o, p' -DDT 15.11 o, p' -DDD 15.79 p, p' -DDD 16.15 β -Endosulfan 16.67 p, p' -DDT 17.25	OCPs Retention time (min) on glass column Percent recoveries (%) α -HCH 3.83 90 γ -HCH 4.77 87 β -HCH 5.60 92 δ -HCH 6.40 85 o, p' -DDE 10.40 83 α -Endosulfan 11.50 88 p, p' -DDE 12.30 86 Dieldrin 13.11 87 o, p' -DDT 15.11 82 o, p' -DDD 15.79 80 p, p' -DDD 16.15 86 β -Endosulfan 16.67 87 p, p' -DDT 17.25 83

contaminated with varying levels of OCP residues. The range and mean \pm S.D. values observed in the three districts during the pre-monsoon and post-monsoon seasons are mentioned in Table 2. The main isomers and metabolites analysed in the groundwater of Ambala and Gurgaon rural areas were β -HCH, p,p'-DDD, p,p'-DDT, γ -HCH, p, p'-DDE and β -endosulfan with a percent traceability of 100%, 83%, 71%, 58%, 54% and 50%, respectively. The other isomers and metabolites were traced in 8–46% of samples while δ -HCH was below the detection limits (BDL) in all groundwater samples analysed. In surface water from Hisar rural areas, the main isomers and metabolites observed were β -HCH, p, p'-DDD, p,p'-DDE, p,p'-DDT and β -endosulfan with a traceability of 93%, 86%, 86%, 57% and 57%, respectively. Other isomers and metabolites were found in 7–36% of samples while α -HCH and δ -HCH were BDL in all the analysed surface water samples. On the basis of mean values of total organochlorine pesticide (ΣOCP) residues $(\Sigma HCH + \Sigma DDT + \Sigma endosulfan)$, maximum contamination was found in water of the rural areas in Ambala district, which was followed by the rural areas in Gurgaon and Hisar districts (Fig. 2).

During the study period, about 50%, 58% and 7% of samples from Ambala, Gurgaon and Hisar rural areas, respectively, exceeded the total pesticide level of 500 ng/L recommended for drinking water (EECD 1998). However, for Σ HCH, 25%, 41% and 14% of samples, respectively, from Ambala, Gurgaon, and Hisar districts and for endo-

sulfan only 01–03% of samples from these areas exceeded the limit of 100 ng/L for individual pesticide (EECD 1998). For Σ DDT, except for 8% of samples each from Ambala and Gurgaon, all the studied samples crossed the prescribed limits. However, in Hisar, 57% of samples exceeded the limits.

During the study period, the concentration of Σ HCH in groundwater from the rural areas of Ambala and Gurgaon ranged between 7.1-298.6 and 8.4-288.6 ng/L. β-HCH was detected in all samples and generally had values more than γ -HCH. In Ambala and Gurgaon groundwater, the percent contribution of α -HCH ranged from 0.4% to 4% while δ -HCH was BDL. In the surface water supply of Hisar rural areas, Σ HCH ranged from BDL to 560.6 ng/L and β -HCH contribution was 96–100% of Σ HCH. Alpha- and δ -HCH were BDL, while γ -HCH was found in only 14% of water samples. The presence of β -HCH as the main isomer contributing to Σ HCH in both groundwater and surface water samples reflects an old source of pollution. It may be due to least reactiveness and environmental persistence of β-HCH among HCH isomers (Wang et al. 2003). Technical HCH is already banned for use in agriculture, but it continues to be used for public health purposes. The restricted use of lindane (γ -HCH) is still there and even the state government recommends its use to control termite, stem borer and root borer of sugarcane (Haryana Government website 2007) due to which γ -HCH residues were found in 42% of the water samples studied.

Table 2 Residues of organochlorine pesticides (ng/L) in rural drinking water from three districts of Haryana

Sr.	Sampling	Ambala		Gurgaon		Hisar	
no.	areas→	Pre-monsoon	Post-monsoon	Pre-monsoon	Post-monsoon	Pre-monsoon	Post-monsoon
	pesticides ↓	(n = 6)	(n = 6)	(n = 6)	(n = 6)	(n = 7)	(n = 7)
1	α-HCH	BDL	BDL-0.7 ^a	BDL-18.1	BDL-17.3	BDL	BDL
			0.1 ± 0.3^{b}	3.0 ± 7.4	2.9 ± 7.0		
2	γ-ΗCΗ	BDL-89.9	BDL-24.5	BDL-63.6	BDL-141.9	BDL	BDL-27.7
		21.0 ± 34.3	6.0 ± 10.2	20.9 ± 25.5	30.2 ± 55.1		5.0 ± 9.1
3	β-НСН	39.2-290.6	7.1-52.3	8.7-188.4	8.4-129.4	2.3-23.2	BDL-537.9
		126.1 ± 100.9	21.9 ± 17.0	99.8 ± 74.4	42.6 ± 50.5	13.1 ± 8.3	138.8 ± 200.5
4	δ-ΗCΗ	BDL	BDL	BDL	BDL	BDL	BDL
5	ΣΗCΗ	40.1-298.6	7.1–76.8	8.7-270.1	8.4-288.6	2.3-23.2	BDL-560.6
		147.1 ± 117.7	28.1 ± 25.6	123.8 ± 90.8	75.7 ± 108.6	13.1 ± 8.3	143.8 ± 207.0
6	α-Endosulfan	BDL	BDL	BDL-404.8	BDL	BDL-90.4	BDL-31.9
				76.7 ± 162.2		37.1 ± 41.7	4.5 ± 12.0
7	β-Endosulfan	BDL-58.8	BDL-164.2	BDL-620.3	BDL-345.1	BDL-52.8	BDL-174.4
		13.1 ± 23.7	41.7 ± 64.6	189.7 ± 292.5	61.9 ± 138.9	14.6 ± 23.1	49.8 ± 61.2
8	Σ Endosulfan	BDL-58.8	BDL-164.2	BDL-913.8	BDL-345.1	BDL-132.9	BDL-206.3
		13.1 ± 23.7	41.7 ± 64.6	266.4 ± 399.5	61.9 ± 138.9	51.7 ± 51.7	54.3 ± 72.2
9	o, p'-DDT	BDL-67.2	BDL-9.7	BDL	BDL	BDL-8.8	BDL-39.2
		15.5 ± 26.1	1.6 ± 4.0			1.3 ± 3.3	7.1 ± 14.7
10	p, p'-DDT	BDL-461.9	BDL-1,050.1	BDL-121.3	13.4-124.5	BDL-50.4	46.1-116.9
		183.4 ± 209.0	565.9 ± 435.0	39.3 ± 56.5	56.7 ± 38.9	7.2 ± 19.0	72.0 ± 23.5
11	o, p'-DDE	BDL-21.1	BDL-53.2	BDL-26.5	BDL-28.8	BDL-6.3	BDL
		3.5 ± 8.6	9.4 ± 21.4	4.6 ± 10.7	4.6 ± 9.0	0.9 ± 2.4	
12	p, p'-DDE	66.0-251.3	BDL-120.4	102.5-341.8	42.2-81.1	BDL-105.5	BDL-97.9
		186.3 ± 68.4	57.2 ± 40.3	202.8 ± 88.0	56.0 ± 13.9	51.4 ± 33.5	54.4 ± 30.7
13	o, p'-DDD	BDL-181.3	BDL-21.0	BDL-6.6	BDL-12.6	BDL-8.8	BDL-35.7
		31.3 ± 73.5	5.8 ± 9.3	1.1 ± 2.7	4.3 ± 5.8	4.4 ± 4.2	8.2 ± 13.9
14	p, p'-DDD	BDL-1,159.6	44.6-279.6	BDL-270.5	BDL-143.9	5.4-27.6	BDL-187.3
		520.2 ± 576.3	116.0 ± 87.4	107.8 ± 114.0	71.8 ± 53.6	18.4 ± 8.0	47.0 ± 64.5
15	ΣDDT	89.3-1,888.4	174.7-1,207.6	115.6-665.1	82.1-308.9	50.1-136.5	90.3-332.2
		940.3 ± 746.8	756.1 ± 443.8	357.1 ± 201.4	193.5 ± 91.2	83.5 ± 30.6	188.7 ± 77.3
16	ΣΟCΡ	868.5-2,187.0	201.9-1,298.0	124.3-1,544.0	119.0-599.1	79.3-197.3	16.9-713.3
		$1,100.6 \pm 770.8$	825.9 ± 461.1	747.4 ± 503.2	331.2 ± 216.5	148.4 ± 46.0	386.9 ± 175.4
17	ΣOCP (mean	201.9-2,187.0		119.0-1,544.0		79.3-713.3	
	of two seasons)	963.2 ± 622.3		539.3 ± 428.5		267.7 ± 174.6	

^aRange

^bMean value \pm S.D.

Kumar et al. (1995) also observed a high concentration of β -HCH as compared to α - and γ -HCH in the rural drinking water sources of northern and northeastern India. The water of River Yamuna and its canals in Haryana was also found to have β -HCH as the main isomer with a mean value of 117.4 and 96.1 ng/L, respectively (Kaushik et al. 2008). The frequent occurrence of β -HCH in higher concentration despite the complete ban on technical HCH use may be due to its high persistence and resistance especially to microbial degradation (Dogra et al. 2004) as compared to α -, γ - and δ -HCH (Johri et al. 1998). Lindane (γ -HCH) also isomerizes in the environment to α and β -HCH (Waliszewski 1993). The occurrence of relatively higher proportions of β - and γ -HCH as compared to α - and δ -HCH is due to the fact that β -HCH is recalcitrant and the restricted use of γ -HCH still continues. Moreover, the loss of various isomers due to volatilization depending upon their vapour pressure (Kaushik 1989) from the time of application and runoff to join water bodies and differential solubilities explain the variation in distribution abundance in relation to



Fig. 2 Residues of different OCP residues in drinking water from three districts (1, pre-monsoon; 2, post-monsoon)

the proportion of their occurrence in the mixture. The pesticide residues which remained in the soils due to earlier application become a source of pesticide pollution of water through agricultural runoff and leaching. These pesticides contaminate drinking water, the fact which had caused a considerable concern as various laboratories detected residues in different brands of mineral water with the total HCH concentration of 24,100.0 ng/L (Prakash et al. 2004) and the highest concentration of lindane, i.e. 4,200.0 ng/L, in various brands of softdrinks (CSE 2003).

Among the studied OCPs, the concentration of DDT was comparatively more in analysed water samples in rural areas of the three districts. In ΣOCP concentration, ΣDDT contributes between 85–91%, 49–56% and 48–58%, respectively, in the drinking water of the rural areas of Ambala, Hisar and Gurgaon. On the main basis of sampling periods also, the concentration of Σ DDT was found to be highest in water of Ambala followed by Gurgaon and Hisar in the pre-monsoon and post-monsoon period (Fig. 2). This may be due to their physical, chemical and biological properties. The half-life of DDT is more than HCH in Indian soils (Kaushik 1991). Among the studied metabolites, the p, p'-DDE had a contribution of 28.94–56.79% and the p,p'-DDT of 11.0-29.3% in groundwater from Gurgaon. These values were 28.83-61.56% and 8.62-38.15%, respectively, in surface water from Hisar rural areas. Singh (2001) reported a 50% contribution of

p, p'-DDE in Σ DDT concentration in groundwater from Agra (India), whereas p, p'-DDD and p, p'-DDT constituted 10% and 40%, respectively. Since p, p'-DDT is known to undergo metabolic conversion and dehydrochlorination (Matsumura 1973), the p,p'-DDE and p,p'-DDD found in drinking water might be due to such metabolic processes. However, in the case of groundwater from Ambala rural areas, p, p'-DDD and p, p'-DDT contribute more during pre-monsoon and post-monsoon sampling, respectively. Mukherjee and Gopal (2002) reported that, except for one groundwater sample, all samples from villages and agricultural fields in the national capital of Delhi were found to be contaminated with p, p'-DDD metabolite only. The groundwater aquifers of Gangetic plains of India, in Unnao district of Uttar Pradesh, were found to have a maximum concentration of p, p'-DDD (BDL-240.2 ng/L) followed by p, p'-DDE (BDL-73.9 ng/L), whereas p,p'-DDT ranged between BDL and 13.6 ng/L (Singh et al. 2005).

p, p'-DDT undergoes slow degradation to p, p'-DDE and p,p'-DDD in natural environment by chemical and biological processes (Baxtor 1990). The ratio of (p,p'-DDE + p,p'-DDD)/p,p'-DDTfurther provides an indication of the extent of recent release of DDT into the environment, with the ratio increasing over time as DDT degraded. In the case of groundwater from Ambala areas, except for one sample, the ratio was 2.6–3.0 during the pre-monsoon season, reflecting the old pollution source. However, during the post-monsoon season, except for one water sample, the ratio lies between 0.13 and 0.79, reflecting the current use of DDT in the area. In the case of Gurgaon water, the ratio ranged between 3.9-7.8 and 1.1-8.8, respectively, in the pre-monsoon and postmonsoon seasons, indicating the old use of DDT, while in the case of Hisar 43% of samples showed the ratio below unity and remaining above unity, indicating both the current and old source of DDT in the canal catchment area.

Water samples were also analysed for endosulfan. Endosulfan isomers were found in 50–57% of water samples. Their concentration is given in Table 2. It may be due to its continued use in agriculture. Endosulfan is recommended for use to control termites in wheat, gram and barley, Fig. 3 Relative

G—Gurgaon and H—Hisar in Haryana

abundance of different

isomers/metabolites of Σ OCP in drinking water from A—Ambala,



Hairy caterpillar in pearl millet, cotton, sunflower, mustard and maize, spotted American and pink ball worms in cotton and leaf fodder in rice crops in the studied districts during different crop seasons (Haryana Government website 2007). In the case of Ambala, α -endosulfan was BDL in all the groundwater samples, while in Gurgaon it was found in only two samples. Endosulfan sulfate could not be analysed. The mean values of Σ endosulfan in water samples were observed in the order: Gurgaon > Hisar > Ambala. Among Σ endosulfan, α -endosulfan contributed a maximum of up to 44.30% and 71.76%, respectively, in groundwater and surface water, while β endosulfan contribution was between 55.7–100% and 28.24–97.71 %, respectively. The concentration of both isomers of endosulfan was BDL in 42% of samples, while in about 48% of samples β endosulfan concentration was higher than that of α -endosulfan. In only 10% of water samples was

 α -endosulfan concentration found to be higher than that of β -endosulfan. Different studies reported the high concentration of β -endosulfan in water samples from agricultural fields (Kumari et al. 2008). From soils, the α -isomer of endosulfan disappears more rapidly as it is more volatile than the β -isomer (UNEP/FAO 2007) and even photo-isomerizes to β-isomer (Walia and Dureja 1993). The present residue levels of endosulfan isomers in the case of Ambala groundwater were lower than that observed in the groundwater of Farrukhabad Gangetic plains, India (Mohapatra et al. 1995); however, in the case of Gurgaon water, the presently observed values were higher. The residue level of BDL-206.3 ng/L in surface water from Hisar rural areas is higher than that of endosulfan residues of ND-24.5 ng/L reported from surface drinking water sources (water works) of the national capital of Delhi, India (Sharma et al. 2003).

The highest concentration of Σ OCP residues was observed in groundwater samples from Ambala, followed by Gurgaon. In the case of surface water samples from Hisar, the values were still lower (Fig. 2). The percentage contribution by different isomers/metabolites analysed during the study period (mean basis) is illustrated in Fig. 3. The levels of pesticide residues observed in groundwater in the present study are lower than the residues (1,559.0 ng/L) reported by Kumari et al. (2008) in groundwater from the agricultural fields from Haryana and from that of tube well water (133,000.0 ng/L) from Jaipur, India (Bakore et al. 2004). However, the present residue level is higher than that reported in groundwater (ND-471.0 ng/L) from Kanpur agricultural areas (Sankararamakrishnan et al. 2005) and that of bore wells (170.0 ng/L) and dug wells (238.0 ng/L) from alluvial groundwater aquifers of Gangetic plains, India (Singh et al. 2005). The OCP residues found in the surface water of rural Hisar (267.7 ng/L) are lower than 250,000.0 ng/L as reported by Sankararamakrishnan et al. (2005) from a reservoir used for drinking purposes in Jaipur, India. However, the present level of OCP residues in surface water (267.7 ng/L) is comparable with the residue levels in its parent canal, i.e. Western Yamuna Canal, of 254.0 ng/L but is lower than Sunder branch, Agra and Gurgaon canals in Haryana state (Kaushik et al. 2008).

Fig. 4 Relative abundance of different organochlorine pesticide residues in drinking water from Ambala (**a**), Gurgaon (**b**) and Hisar (**c**) in Haryana, India



The present residue level of the studied OCP residues in groundwater and surface water of the rural areas of the three districts may be due to intense agricultural activity in the rural areas and their use in vector control programs, epidemics and fairs, during safe storage of grains, in cattle sheds, illegal use and their residues from previous use. The present study further shows that the groundwater was found to be contaminated more than the surface water which may be because the groundwater flows non-turbulently, experiences limited dilutions and has anaerobic conditions as compared to surface water. Soils become reservoir of pesticides from their previous and continual illegal use, thereby steadily transferring them to groundwater. Most OCPs are sparingly soluble in water; however, they leach to the lower soil profiles under the influence of water moving towards the sub-soil (Kaushik 1991).

Seasonal variations

No definite pattern of OCP residues was observed in groundwater samples from Ambala and Gurgaon water during the two seasons (Fig. 4). However, the concentration of p, p'-DDT was found to increase in groundwater and surface water samples, reflecting its use during rainy season. After the rainy season, the percentage contribution of p, p'-DDT increased from 19.5–75%, 11.0– 29% and 8.62–38.15%, respectively, in Ambala, Gurgaon and Hisar rural water. In the case of Hisar surface water samples, the concentration of all isomers and metabolites (except α -endosulfan and o, p'-DDE) were found to increase after the rainy season (Table 2). The presence of these pesticides in wet months and their eventual disappearance in dry months in river water as also observed by earlier workers (Agnihotri et al. 1994; Ramesh et al. 1990) suggest that runoff from agricultural fields in the catchment area of the canal is the cause of such residues.

The findings of the present study show the presence of residues of persistent organochlorine pesticides like HCH and DDT, which are now banned, and endosulfan, which is still in use, in drinking water (ground and surface) from different rural areas of Haryana. Some of the isomers/metabolites detected might be the result of their prolonged persistence in the environment and their illegal use. Their presence along with the residues of other pesticides still in use poses a health risk to local residents consuming that water. Treatment of water should be done before use as 37% of water samples exceeded the total pesticide level of 500 ng/L indicated in the EECD directive for drinking water. There is a need for awareness among farmers so that illegal use could be controlled. Strict action against sellers of banned or spurious pesticides and adoption of integrated pest management practices could be a possible way to overcome/bring down the present residue levels in the future. The present contamination level is expected to come down further in the future due to the subtropical meteorological conditions of Haryana state which are considered as favourable for the dissipation of pesticides. Although different types of pesticides are widely used for agricultural purposes in the state, the present study was limited to only four OCPs. Finally, it must be pointed out that our research was restricted only to rural areas of three districts of the state during two seasons. More comprehensive monitoring studies have to be carried out in different seasons in different districts with a wider spectrum of banned and still-in-use pesticides to get the clear status for the entire state.

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References

- Agnihotri, N. P. (2000). Pesticide consumption of agriculture in India—an update. *Pesticide Research Journal*, 12(1), 150–155.
- Agnihotri, N. P., Gajbhiye, V. T., Kumar, M., & Mohapatra, S. P. (1994). Organochlorine insecticide residues in Ganga River water near Farrukhabad, India. *Environmental Monitoring and Assessment*, 30, 105–112.
- Bakore, N., John, P. J., & Bhatnagar, P. (2004). Organochlorine pesticide residues in wheat and drinking water samples from Jaipur, Rajasthan, India. *Environmental Monitoring and Assessment*, 98, 381–389.
- Baxtor, R. M. (1990). Reductive dechlorination of certain chlorinated organic compounds by reduced haematin

compared with their behavior in the environment. *Chemosphere*, 21, 451–458.

- Census of India (2001). www.censusindia.net/results/index. html.
- Central Ground Water Board (2007). Ground water information booklets of Ambala, Hisar and Gurgaon Districts, Ministry of Water Resources, Government of India, North Western Region, Chandigarh. http:// cgwb.gov.in/District_Profile/Haryana/Ambala.pdf, http:// cgwb.gov.in/District_Profile/Haryana/Gurgaon% 20Broucher.pdf, http://cgwb.gov.in/District_Profile/ Haryana/Hissar.pdf. Accessed 22 January 2011.
- Centre for Science and Environment (2003). www. downtoearth.org.in.
- Dogra, C., Raina, V., Pal, R., Suar, M., Lal, S., Gartemann, K. H., et al. (2004). Organization of lin genes and IS 6100 among different strains of hexachlorocyclohexane-degrading Sphingomonas paucimobilis—evidence for horizontal gene transfer. *Journal of Bacteriology*, 186, 2225–2235.
- EECD (European Economic Community Directive), EEC Council Directive 98/83/EC. (1998). *Official Journal of European Communities*, Brussels, Vol. L330, p 42.
- Haryana Government website (2007). http://haryana. gov.in and http://agriculture.nic.in. Accessed 21 April 2007.
- Johri, A. K., Dua, M., Tuteja, D., Saxena, R., Saxena, D. M., & Lal, R. (1998). Degradation of alpha, beta, gamma and delta-hexachlorocyclohexane by Sphingomonas paucimobilis. *Biotechnology Letters*, 20, 885– 887.
- Kaushik, A., Sharma, H. R., Jain, S., Dawra, J., & Kaushik, C. P. (2010). Pesticide pollution of river Ghaggar in Haryana, India. *Environmental Monitoring and As*sessment, 160, 61–69.
- Kaushik, C. P. (1989). Loss of HCH from surface soil layers under subtropical conditions. *Environmental Pollution*, 59, 253–264.
- Kaushik, C. P. (1991). Persistence and metabolism of HCH and DDT in soil water under subtropical conditions. *Soil Biology and Biochemistry*, 23, 131–134.
- Kaushik, C. P., Sharma, H. R., Jain, S., Dawra, J., & Kaushik, A. (2008). Levels of pesticide residues in river Yamuna and its canals in Haryana and Delhi, India. *Environmental Monitoring and Assessment*, 144, 329–340.
- Kumar, S., Kunwar, P. S., & Gopal, K. (1995). Organochlorine residues in rural drinking water sources of northern and north eastern India. Journal of Environmental Science and Health. *Environmental Science and Engineering & Toxic and Hazardous Substance Control*, 30A(6), 1211–1222.
- Kumari, B., Madan, V. K., & Kathpal, T. S. (2008). Status of insecticide contamination of soil and water in Haryana, India. *Environmental Monitoring and As*sessment, 136(1–3), 239–244.
- Kumari, B., Singh, R., Madan, V. K., Kumar, R., & Kathpal, T. S. (1996). DDT and HCH compounds in soils, ponds and drinking water of Haryana, India. *Bulletin of Environmental Contamination and Toxicology*, 57, 787–793.

- Matsumura, F. (1973). Degradation of pesticide residues in the environment. In: C. A. Edwards (Ed.), *Environmental pollution by pesticides* (pp. 494–513). New York: Plenum.
- Mohapatra, S. P., Kumar, M., Gajbhiye, V. T., & Agnihotri, N. P. (1995). Ground water contamination by organochlorine insecticide residues in a rural area in the Indo-Gangetic plain. *Environmental Monitoring* and Assessment, 35, 155–164.
- Mukherjee, I., & Gopal, M. (2002). Organochlorine insecticide residues in drinking and ground water in and around Delhi. *Environmental Monitoring and Assessment*, 76, 185–193.
- Nair, A., & Pillai, M. K. K. (1992). Trends in amient levels of DDT and HCH residues in humans and the environment of Delhi, India. *The Science of the Total Environment*, 121, 145–157.
- Prakash, O., Suar, M., Raina, V., Dogra, C., Rinku, P., & Lal, R. (2004). Residues of hexachlorocyclohexane isomers in soil and water samples from Delhi and adjoining areas. *Current Science*, 87, 73–77.
- Ramesh, A., Tanabe, S., Iwata, H., Tatsukawa, R., Subramanian, A. N., Mohan, D., et al. (1990). Seasonal variation of persistent organochlorine insectide residues in Vellar river water in Tamil Nadu, South India. *Environmental Pollution*, 67, 289–304.
- Sankararamakrishnan, N., Sharma, A. K., & Sanghi, R. (2005). Organochlorine and organophosphorus pesticides residues in ground water and surface waters of Kanpur, Uttar Pradesh, India. *Environmental International*, 31, 113–120.
- Sharma, H. R., Trivedi, R. C., Akolkar, P., & Gupta, A. (2003). Micro pollutants levels in macroinvertebrates collected from drinking water sources of Delhi, India. *International Journal of Environmental Studies*, 60, 99–110.
- Singh, K. P., Malik, A., Mohan, D., & Sinha, S. (2005). Persistent organochlorine pesticide residues in alluvial groundwater aquifers of Gangetic plains, India. *Bulletin of Environmental Contamination and Toxicology*, 74, 162–169.
- Singh, R. P. (2001). Composition of organochlorine pesticide levels in soil and ground water of Agra, India. *Bulletin of Environmental Contamination and Toxicol*ogy, 67, 126–132.
- UNEP/FAO/CRC.3/13 (2007). Rotterdam convention on te prior informed consent procedure for certain hazardous chemicals and pesticides in International trade. *Chemical Review*, Third Meeting, Rome; March 2007.
- Walia, S., & Dureja, P. (1993). Photochemical and microbial degradation of insecticides. *Pesticide Research Journal*, 5, 133–150.
- Waliszewski, S. M. (1993). Residues of Lindane, HCH isomers and HCB in the soil after Lindane application. *Environmental Pollution*, 82, 289–293.
- Wang, X. T., Chu, S. G., & Xu, X. B. (2003). Oranocholorine pesticide residues in water from Guanting reservoir and Yongding River, China. *Bulletin of Environmental Contamination and Toxicology*, 70, 351– 358.