

DDT and HCH Compounds in Soils, Ponds, and Drinking Water of Haryana, India

B. Kumari, R. Singh, V. K. Madan, R. Kumar, T. S. Kathpal

Centre of Environmental Studies, College of Basic Sciences and Humanities,
CCS Haryana Agricultural University, Hisar 125 004, India

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Soil serves as ultimate sink for pesticides applied directly to the soil or when applied on the crops. Organochlorine insecticides particularly HCH and DDT have been in use in Haryana, India for agriculture as well as public health purposes during the last 2-3 decades. Razik *et al.* (1988) reported residues of BHC, aldrin, dieldrin and p,p'-DDT in Nile river waters. The contamination of pond waters with a number of herbicides, fungicides and insecticides was demonstrated by Frank *et al.* (1990) in Ontario, Canada. The contamination of drinking water as well as ground water with organochlorine insecticides has been reported all over world (Bakra *et al.* 1990; Iwan 1990; Jani *et al.* 1991; Ritter 1991).

In Vietnam and Thailand contamination of soil samples collected randomly from paddy fields, uplands and urban areas (roadsides and gardens) with organochlorine compounds such as DDT, HCH and PCBs has been published by Thao *et al.* (1993). No systematic work has been done in India to find out the level of pesticidal contamination of soil, pond and drinking water with these two persistent insecticides being used extensively since mid fifties. Studies were, therefore, undertaken during 1992-94 to investigate the level of pesticidal contamination of soils and pond waters from three districts of Haryana. Additionally some drinking water samples from different hand pumps and water works of Hisar city were analysed for presence of DDT and DDT residues.

MATERIALS AND METHODS

The studies were conducted during 1992-94 at three different locations (L). Soil as well as pond water samples were collected from rice growing areas in Hisar (L-1) and Sirsa (L-2) districts, and from rainfed areas of Rohtak (L-3). The drinking water samples (12) were collected from hand pumps and water works of different sites of L-1.

Correspondence to: B. Kumari

Forty two samples of soil (L-1 10, L-2 and L-3 16 each) collected randomly from depths upto 10-15 cm from the fields were brought to laboratory and air dried. Representative sample (40 g) obtained after quartering from each location was extracted immediately by column chromatographic technique (Kathpal *et al.* 1988) utilizing 5 g adsorbent mixture (activated charcoal - Celite545 - florisil, 3:1:1). Hexane - acetone (4:1, v/v) mixture (150 ml) was employed as eluant. Residues from pond water samples (250 ml) were extracted thrice with 50, 30 and 30 ml of hexane by partitioning. Hexane extracts were cleaned with activated charcoal employing hexane - acetone mixture (4:1,v/v) as per details given by Kathpal and Dewan (1975). Drinking water samples were extracted in a similar manner followed for pond water. However, there was no need of clean-up. Hexane extract after passing through sodium sulphate was directly used for analysis.

Extracts obtained from soil and water were analyzed for residue content of different isomers of HCH, DDT and its analogues. The analyses were carried out with a Hewlett Packard 5890A gas chromatograph equipped with electron capture detector (^{63}Ni) and glass column (1.5 m x 3.0 mm ID) packed with 3% OV-17 coated on chromosorb WHP (80-100 mesh), and operated at temperatures($^{\circ}\text{C}$) : column 200, injection port 230 and detector 250. Nitrogen (60 ml/mm) was used as carrier gas.

Percent recovery from soil, pond water and drinking water samples spiked at the levels of 0.01 - 0.05 ppm varied from 87 to 98% for HCH isomers, and 85 to 92% for DDT and its analogues.

RESULTS AND DISCUSSION

Analytical data given in Tables 1 and 2 reveal that all 10 soil samples collected from L-1 showed contamination with HCH residues. Total residues of α , β , γ and δ isomers of HCH ($\Sigma\text{-HCH}$) were $0.134 \pm 0.059 \mu\text{g g}^{-1}$. α , β and γ isomers of HCH were present in all the samples in more or less equal amounts ($0.042\text{-}0.046 \mu\text{g g}^{-1}$), whereas δ isomer was found to be least of all isomers. Two out of ten soil samples did not contain detectable amounts of $\delta\text{-HCH}$ at the lowest detection limit of $0.001 \mu\text{g g}^{-1}$.

Average residues of $\Sigma\text{-DDT}$ were $0.039 \pm 0.012 \mu\text{g g}^{-1}$. Among different analogues of DDT : p,p'-DDE, o,p'-TDE and p,p' -TDE were present in most of the samples whereas p,p'-DDT, the parent compound was present only in five out of 10 samples at a level of $0.002 \pm 0.003 \mu\text{g g}^{-1}$. All the samples (16) collected from L-2 were contaminated with residues of $\Sigma\text{-HCH}$ ($0.048 \pm 0.023 \mu\text{g g}^{-1}$). Like L-1 soils, the average amounts of α , β and $\gamma\text{-HCH}$ were almost equal ($0.014\text{-}0.017 \mu\text{g g}^{-1}$) in all the samples.

None of the tested samples was found to contain measurable amounts of δ -HCH, DDT and its analogues with lowest detection limit for the latter being $0.003\mu\text{g g}^{-1}$. In L-3 samples average Σ -HCH residues were $0.162\pm0.087\mu\text{g g}^{-1}$. Unlike samples from L-1 and L-2, the soil samples from L-3 contained all isomers of HCH with maximum average ($0.103\mu\text{g g}^{-1}$) of β -HCH. However, α and γ residues were slightly lower than those detected in L-1 soils. DDT was detectable in the form of o,p'-DDE and p,p'-DDE. The total DDT residues were found to be $0.045\pm0.015\mu\text{g g}^{-1}$.

Table 1. Residues of HCH in soils,

Location	Average residues ($\mu\text{g g}^{-1}\pm\text{SD}$)				
	α - HCH	β - HCH	γ - HCH	δ - HCH	Σ - HCH
L-1	0.046	0.042	0.044	0.002	0.134
(10)	± 0.025	± 0.020	± 0.025	± 0.001	± 0.059
L-2	0.017	0.014	0.017	BDL	0.048
(16)	± 0.006	± 0.005	± 0.006		± 0.023
L-3	0.027	0.103	0.014	0.018	0.162
(16)	± 0.012	± 0.076	± 0.016	± 0.008	± 0.087

BDL - Below detectable limit.

Figures in parentheses denote number of samples analysed.

Table 2. Residues of DDT and its analogues in soils.

Location	Average residues ($\mu\text{g g}^{-1}\pm\text{SD}$)					
	o,p'-TDE	p,p'-TDE	o,p'-DDE	p,p'-DDE	p,p'-DDT	Σ -DDT
L-1	0.008	0.007	BDL	0.020	0.002	0.039
(10)	± 0.003	± 0.008		± 0.006	± 0.003	± 0.012
L-2	BDL	BDL	BDL	BDL	BDL	BDL
(16)						
L-3	BDL	BDL	0.024	0.021	BDL	0.045
(16)			± 0.012	± 0.019		± 0.015

BDL - Below detectable limit.

Figures in parentheses denote number of samples analysed.

It is evident from the data given in the Tables 1 and 2 that the incidence of contamination of soil samples with HCH was higher than with DDT. Locationwise, L-3 soils contained maximum HCH residues followed by L-1 and L-2. The total amount of DDT isomers was almost equal in soils of L-1

and L-3, whereas, soils of L-2 were free from DDT contamination. In the light of the fact that use of DDT has been banned and that of HCH restricted in agriculture since 1993, in India, these residues appeared to have been carried over from previous treatments.

Contamination of soil with DDT and HCH upto the levels of 0.675 and 0.032 $\mu\text{g g}^{-1}$, respectively was reported from the states of northern India, i.e. Uttar Pradesh, Punjab and Delhi during 1965-1978 (Anonymous 1988). Thao *et al.* (1993) demonstrated presence of HCH residues upto 0.0048 $\mu\text{g g}^{-1}$ in Vietnam and 0.0004 $\mu\text{g g}^{-1}$ in Thailand, whereas DDT was shown to be present upto 0.02 $\mu\text{g g}^{-1}$ in former and 0.083 $\mu\text{g g}^{-1}$ in the latter case.

The analytical data pertaining to pond water samples collected from different locations are presented in Tables 3 and 4. In all the 10 samples collected from L-1 average Σ -HCH residues were $2.2 \pm 0.9 \mu\text{g l}^{-1}$. Among different isomers, α was maximum ($1.0 \mu\text{g l}^{-1}$), whereas β , γ and δ isomers were present at a level of $0.4 \mu\text{g l}^{-1}$. DDT residues (Σ -DDT, $0.3 \pm 0.27 \mu\text{g l}^{-1}$) were detected in the form of p, p'-DDE and o, p'-TDE, in amounts varying from 0.1 to $0.2 \mu\text{g l}^{-1}$. None of the water samples contained DDT in the parent form at lowest detection limit of $0.15 \mu\text{g l}^{-1}$.

All the 16 samples from L-2 showed presence of average Σ -HCH residues to the level of $9.0 \pm 4.0 \mu\text{g l}^{-1}$. α and 13 isomers were present in all the samples with average amounts varying from 2.0 to $3.0 \mu\text{g l}^{-1}$. β -HCH was present in 10 samples to a level of $3.0 \pm 3.0 \mu\text{g l}^{-1}$, whereas δ -HCH was present upto $1.0 \mu\text{g l}^{-1}$ in most of the samples. All the samples were found to be contaminated with DDT and its analogues. The residues were mainly present as p, p'-TDE and p, p'-DDE. The cumulative amount of these two was $2.7 \pm 1.0 \mu\text{g l}^{-1}$.

All the 16 samples from L-3 were contaminated with all the four isomers of HCH, and have an average (Σ -HCH) residue level of $2.5 \pm 1.0 \mu\text{g l}^{-1}$. α -HCH was maximum ($0.8 \mu\text{g l}^{-1}$) followed by β -HCH ($0.7 \mu\text{g l}^{-1}$), δ -HCH ($0.6 \mu\text{g l}^{-1}$) and γ -HCH ($0.4 \mu\text{g l}^{-1}$). None of the samples contained detectable amounts of DDT residues with lowest detection limit of $0.1 \mu\text{g l}^{-1}$.

Drinking water samples (12) collected from different sites of L-1 were analysed for possible contamination due to HCH and DDT residues. None of the samples contained residues of these insecticides in detectable amounts.

Contrary to our observations pesticidal contamination of drinking water from Jaipur has been reported by Bakra *et al.* (1990). They found organochlorine

Table 3. Residues of HCH in pond waters.

Location	Average residues ($\mu\text{g l}^{-1} \pm \text{SD}$)				
	α - HCH	β - HCH	γ - HCH	δ - HCH	Σ -HCH
L-1	1.0	0.4	0.4	0.4	2.2
(10)	± 0.5	± 0.6	± 0.3	± 0.3	± 0.9
L-2	2.0	3.0	3.0	1.0	9.0
(16)	± 1.0	± 3.0	± 3.0	± 1.0	± 4.0
L-3	0.8	0.7	0.4	0.6	2.5
(16)	± 0.2	± 0.7	± 0.2	± 0.2	± 1.0

Figures in parentheses denote number of samples analysed.

Table 4. Residues of DDT and its analogues in pond waters.

Location	Average residues ($\mu\text{g l}^{-1} \pm \text{SD}$)				
	o,p' -TDE	p,p' -TDE	o,p' -DDE	p,p' -DDE	Σ -DDT
L-1	0.2	BDL	BDL	0.1	0.30
(10)	± 0.2			± 0.1	± 0.27
L-2	BDL	2.0	BDL	0.7	2.7
(16)		± 1.0		± 0.6	± 1.0
L-3	BDL	BDL	BDL	BDL	BDL
(16)					

BDL - Below detectable limit.

Figures in parentheses denote number of samples analysed.

insecticides including HCH and DDT residues varying from 1.07 to 81.23 $\mu\text{g l}^{-1}$. Similarly drinking water samples from Ahmedabad city were found to contain HCH and DDT residues but within prescribed Maximum Residue Limits of 1.0 and 3.0 $\mu\text{g l}^{-1}$ for DDT and HCH, respectively (Jani *et al.* 1991).

A critical perusal of the data given in Tables 3 and 4 clearly reveals that contamination of pond waters due to HCH was higher than DDT residues. The order of contamination was highest in L-2 followed by L-1 and L-3.

Contamination of lake waters with DDT residues ranging from 6.6 to 9.6 $\mu\text{g l}^{-1}$, from Jaipur, India, has been recorded by Kumar *et al.* (1988). Lindane was present in traces. According to Parveen and Masud (1988) cattle drinking water from Karachi, Pakistan, contained γ -HCH ranging from 1.0 to 16.4 $\mu\text{g l}^{-1}$, and p,p'-DDT and p,p'-DDE in traces. In Ontario, rural pond waters were found to contain DDT residues upto 2.24 $\mu\text{g l}^{-1}$ (Frank *et al.* 1990).

Although the level of contamination of pond waters in our studies is relatively much less, yet the presence of HCH and DDT residues in pond waters is a matter of concern because consumption of contaminated pond waters by milch animals may lead to accumulation of lipophilic DDT and HCH residues in the animal body which ultimately gets secreted in milk (Kalra *et al.* 1986). However, chances of further accumulation of DDT and HCH in soil and water in Haryana do not seem to be high because of restrictions imposed on their use on agricultural crops with effect from early ninties in India. On the contrary the present contamination level is expected to come down due to subtropical meteorological conditions of Haryana which are considered to be favorable for pesticidal dissipation.

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