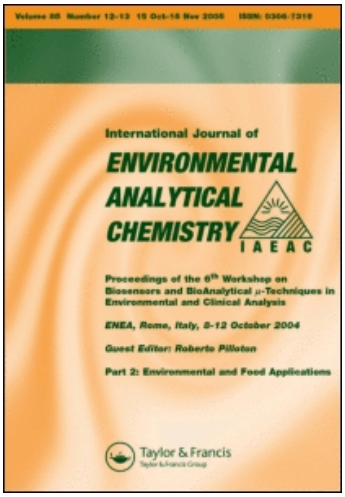

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PERSISTENT ORGANOCHLORINE INSECTICIDE RESIDUES IN SOME PADDY, UPLAND AND URBAN SOILS OF INDIA

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A wide variety of agricultural soils from different regions of India such as paddy, wheat, mustard, potato, cotton, tea, tomato, sugarcane, grape and urban soils were surveyed for the residual levels of persistent organochlorine insecticide residues in 1988 and 1989. DDT and HCH concentrations were found to be higher in upland soils and lower in paddy field soils. These results indicate the large application of HCH and DDT compounds for agricultural purposes in India. In urban soil DDT levels were higher than HCHs reflecting the use of the former insecticide in relatively large amounts for vector control. Among DDT compounds, *p,p'*-DDE showed higher percentage in paddy and upland soils. On the other hand, *p,p'*-DDT contributed to higher levels in urban soils. The β-HCH was detected as the dominant isomer in both agricultural and non agricultural soils. The levels and percentage compositions of DDTs and HCHs revealed the extent of environmental contamination caused by the continuous usage of persistent organochlorine insecticides in large quantities in tropical areas like India.

KEY WORDS: HCH, DDT, persistent organochlorines, paddy soil, upland soil, urban soil

INTRODUCTION

The use of several persistent man-made chemicals such as organochlorine pesticides in developing countries and the health hazards associated with their use has been greatly debated in recent years.[1–3] Inspite of the outcry from researchers and environmentalists, their usage has been steadily increasing in some countries. This is complicated by the fact that few developing countries are manufacturing, consuming and even exporting these pesticides.[4] In this context it should also be mentioned that most of the developing countries are located in the tropical belt whose specific agroclimatic conditions, like high temperature and heavy rainfall, facilitate the rapid removal of these chemicals through air and water. The continued use of these

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pesticides will have far reaching complications as their residues could be traced in remote areas where they are not even used,[5,6] thus contributing to global pollution.

Tropical regions are considered to be very important areas with regard to global pollution by persistent man-made chemicals. To understand the environmental contamination by man-made organics in tropics, India offers an ideal example of a developing country where persistent insecticides like DDT and HCH are still being used for combating agricultural pests and vector control. In this country, annual consumption of DDT amounts to about 19 million kg and that of HCH to 45 million kg.[7] They together account for more than 60% of the pesticides (in quantity) currently used in the country. This has lead to considerable contamination of all the environmental media and biota.[8] As part of our research program aimed at understanding the behaviour and fate of organochlorine insecticides in the tropical environment, our team has already made several studies on the distribution of these residues in air, water, paddy soil, sediments and biota from India.[1,3,9–12] In the present research, a survey was conducted for organochlorine insecticides residue levels in the agricultural and non-agricultural soils in different regions of India.

MATERIALS AND METHODS

Soil samples were collected from 21st Dec. 1988 to 24th Dec. 1989. Sampling locations are shown in Figure 1. Paddy, wheat, mustard, potato, cotton, tea, tomato, sugarcane and grape field soil samples were collected as representative of cultivated soil samples and urban soils (roadside dust and garden soil) as representative of non-cultivated soil samples (Table 1). Samples were collected from at least 4-5 points from each site and then mixed completely. Upper 5 cm of soil were sampled with a vertical corer and placed in polyethylene bags. In the laboratory, the composite samples were air-dried, mixed thoroughly, and shipped to Japan for analyses.

Before analyses, soil samples were ground and passed through a 32 mesh size (500 μm) sieve. Fifty gram of soil was transferred into a 500 ml Erlenmeyer glass stoppered flask. Water free from organochlorines (hexane-washed water) was added to soil samples and kept for 30 min to allow the samples to swell in water. The samples were extracted with 150 ml of acetone by shaking for 1h on a mechanical shaker. The supernatant liquid was decanted into a 3 l separatory funnel containing 200 ml of hexane and 1.3 l of hexane-washed water. This extraction process was repeated twice and the combined acetone extracts were transferred into a separatory funnel. After shaking the funnel, the separated hexane layer was washed thrice with water, dried by passing through anhydrous sodium sulphate and then concentrated to 5 ml using a Kuderna-Danish (KD) concentrator. The KD extracts were subjected to silica gel (Wako gel S-1) or Florisil (Florisil PR) column chromatography. The first fraction eluted with hexane contained *p,p'*-DDE and the second fraction eluted with 20% dichloromethane in hexane contained the HCH (α, β, γ and δ) isomers, *p,p'*-DDD, *p,p'*-DDT and *o,p'*-DDT. Each eluate was concentrated and washed with concentrated sulphuric acid to remove interfering substances. Sulphur in the soil samples was eliminated by treatment with copper chips.

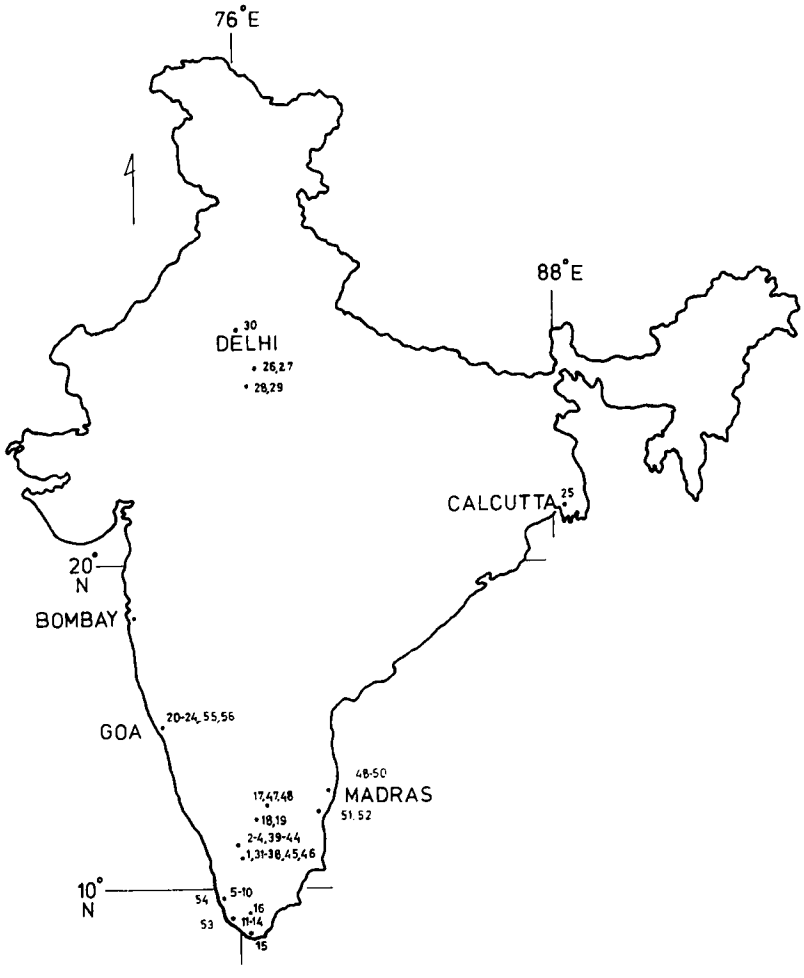

Figure 1 Sampling locations. Numbers represent serial numbers of different soil samples (see Table 1 for details).

Sample extracts were injected into a gas chromatograph (Shimadzu Model GC-7A) equipped with ^{63}Ni electron capture detector and fused silica capillary column (chemically bonded OV-1701, 0.25 mm i.d. × 25 m length). The column oven temperature was programmed as follows: 170° to 230°C at a rate of 1°C min^{-1} with an initial hold of 4 min and a final hold of 8 min. Both injector and detector temperatures were kept at 280°C. Nitrogen was used both as carrier and make-up gas. Confirmation and quantitation of organochlorine insecticides in some representative samples were also carried out using a Hewlett Packard 5890 GC-HP5970B Mass Selective Detector equipped with a fused silica capillary column (chemically bonded OV-1701, 0.25 mm i.d. × 25 m length). The column oven temperature was programmed at 170° to 230°C at a rate of 1°C min^{-1}. Injector and

Table 1 Details of sample and sampling locations in India

Serial No.	*Type of soil*	*Collection date*	*Collection site*
	Agricultural soil		
1	Paddy field soil	23 Dec., 1988	Coimbatore (Tamil Nadu)
2–4	Paddy field soil	24 Dec., 1988	Coimbatore-Nilgiris (Tamil Nadu)
5–10	Paddy field soil	25 Dec., 1988	Coimbatore-Cochin (Tamil Nadu-Kerala)
11–14	Paddy field soil	29 Dec., 1988	Trivandrum-Cape Comorin (Kerala-Tamil Nadu)
15	Paddy field soil	30 Dec., 1988	Cape Comorin-Tirunelveli (Tamil Nadu)
16	Paddy field soil	30 Dec., 1988	Tirunelveli (Tamil Nadu)
17	Paddy field soil	21 Dec., 1989	Nandi (Karnataka)
18, 19	Paddy field soil	21 Dec., 1989	Bangalore (Karnataka)
20, 21	Paddy field soil	23 Dec., 1989	Santacruz (Goa)
22	Paddy field soil	23 Dec., 1989	Sriden (Goa)
23	Paddy field soil	23 Dec., 1989	Zuari (Goa)
24	Paddy field soil	23 Dec., 1989	Miramar (Goa)
25	Paddy field soil	23 Dec., 1989	Calcutta (West Bengal)
26	Wheat field soil	13 Dec., 1989	Bhalabgal (Haryana)
27	Wheat field soil	13 Dec., 1989	Prathala (Haryana)
28	Wheat field soil	13 Dec., 1989	Palwal (Haryana)
29	Mustard field soil	13 Dec., 1989	Prathala (Haryana)
30	Potato field soil	13 Dec., 1989	Delhi
31–38	Cotton field soil	23 Dec., 1988	Coimbatore (Tamil Nadu)
39	Cotton field soil	23 Dec., 1988	Coimbatore-Nilgiris (Tamil Nadu)
40–44	Tea plantation soil	24 Dec., 1988	Nilgiris (Tamil Nadu)
45	Tomato field soil	23 Dec., 1988	Coimbatore (Tamil Nadu)
46	Sugarcane plantation soil	23 Dec., 1988	Coimbatore (Tamil Nadu)
47	Grape orchard soil	21 Dec., 1989	Nandi (Karnataka)
	Urban soil		
48	Roadside dust	21 Dec., 1988	Madras (Tamil Nadu)
49	Roadside dust	20 Dec., 1989	Madras (Tamil Nadu)
50	Garden soil	20 Dec., 1989	Madras (Tamil Nadu)
51	Roadside dust	19 Dec., 1989	Chidambaram (Tamil Nadu)
52	Garden soil	19 Dec., 1989	Chidambaram (Tamil Nadu)
53	Roadside dust	29 Dec., 1988	Trivandrum (Kerala)
54	Roadside dust	28 Dec., 1988	Cochin (Kerala)
55	Roadside dust	24 Dec., 1989	Panaji (Goa)
56	Garden (park) soil	24 Dec., 1989	Panaji (Goa)

'-' indicate that areas sampled between two major collection sites. For e.g. Coimbatore-Nilgiris indicate that samples were collected from different places in between Coimbatore and Nilgiris.

Names mentioned in the parentheses represent the states in which the sampling sites were located.

ion-source temperatures were kept at 250° and 280°C, respectively. Insecticide residues were determined by selective ion monitoring at m/z 217 for HCH isomers, m/z 235 for p,p'-DDD, m/z 246 for p,p'-DDE and m/z 235 for o,p' and p,p'-DDT.

RESULTS AND DISCUSSION

Concentrations of Σ DDT (p,p'-DDE, p,p'-DDD, p,p'-DDT and o,p'-DDT) and Σ HCH (α, β, γ and δ isomers), in paddy field, upland and urban soils are given in Table 2.

Table 2 Concentrations (ng g^{-1} dry wt.) of ΣDDT and ΣHCH in paddy, upland and urban soils in India

Type of soil	*n*	*Σ DDT*[a]				*Σ HCH*[b]			
		Range	*x*[c]	*Median*	*S.D.*	*Range*	*x*	*Median*	*S.D.*
Paddy field soil	25	0.85–2,200	99	2.7	430	0.42–280	34	18	65
Cotton field soil	9	77–4,400	1,000	280	1,400	31–86,000	9,700	79	27,000
Tea plantation soil	5	110–430	260	250	110	20–61	38	29	18
Wheat field soil	3	2.0–5.7	3.7	3.3	1.5	2.9–5.8	4.2	4.0	1.2
Grape orchard field	1	2,100	—	—	—	180	—	—	—
Mustard field soil	1	41	—	—	—	15	—	—	—
Potato field soil	1	30	—	—	—	11	—	—	—
Sugarcane plantation soil	1	1,900	—	—	—	29	—	—	—
Tomato field soil	1	360	—	—	—	640	—	—	—
Roadside dust	7	16–190	65	46	58	0.55–27	20	24	8.7
Garden soil	2	3.4–90	47	—	—	0.6–3.6	2.1	—	—

[a] Σ DDT = *p,p'*-DDE + *p,p'*-DDD + *p,p'*-DDT + *o,p'*-DDT.
[b] Σ HCH = α-HCH + β-HCH + γ-HCH + δ-HCH.
[c] Arithmetic mean.

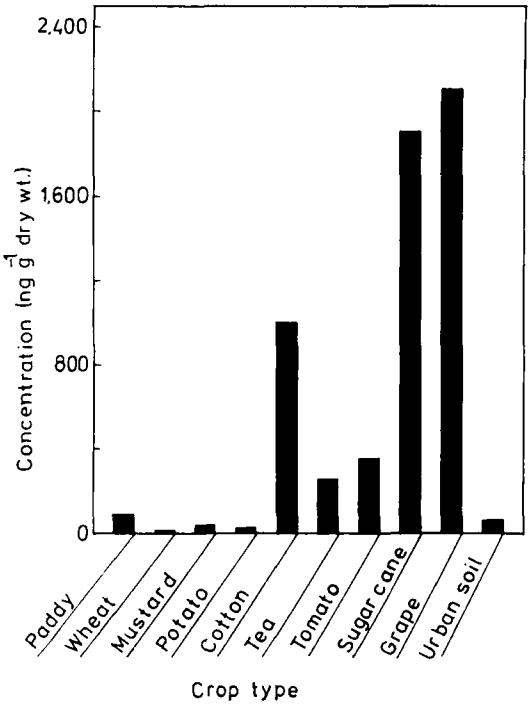

Figure 2 Mean concentration of ΣDDT in various types of soils from India. Urban soil represents road side dust, and garden soil from urban localities.

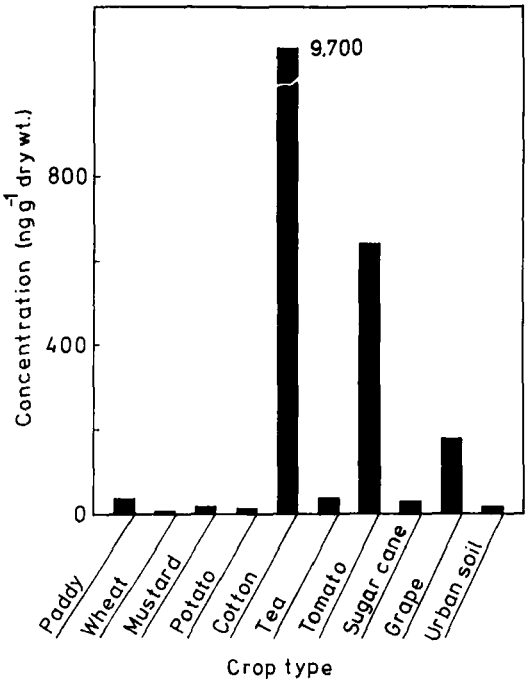

Figure 3 Mean concentration of ΣHCH in various types of soils from India. Urban soil represents road side dust, and garden soil from urban localities.

In paddy soil samples, Σ DDT levels varied from 0.85 to 2,200 ng g^{-1}. On the other hand, Σ HCH levels ranged from 0.42 to 280 ng g^{-1}. In cotton field soils, DDTs and HCHs were detected to be in the range of 77 to 4,400 ng g^{-1} and 31 to 86,000 ng g^{-1}, respectively. In tea field soils, Σ DDT levels ranged from 110 to 430 ng g^{-1} and Σ HCH 20 to 61 ng g^{-1}, respectively. In other agricultural soils (wheat, grape, mustard, potato, sugarcane and tomato), Σ DDT concentrations varied from 2.0 to 2,100 ng g^{-1} and Σ HCH 2.9 to 640 ng g^{-1}, respectively. In urban soil (roadside dust and garden soil), Σ DDT levels ranged from 3.4 to 190 ng g^{-1}. On the other hand, Σ HCH levels in the same samples varied from 0.55 to 27 ng g^{-1}.

The use of pesticides vary according to crop species which may have a bearing on the residue levels. For example, it is much larger for commercially important crops and also crops which are more susceptible to pest attack. Hence the insecticide residue levels recorded in the present study were compared with those of crop types.

The concentrations of Σ DDT and Σ HCH in individual soil types are represented in Figures 2 and 3. Regardless of the small sample size, Σ DDT and Σ HCH levels seem to be relatively high in cotton soils. In this context, information on the relative use of pesticides based on cropped area (RUPA) provides further evidence. Using the estimated data on pesticide usage on different crops in India[13], and on the percentage of total cropped area, RUPA values were calculated. The resulted figures

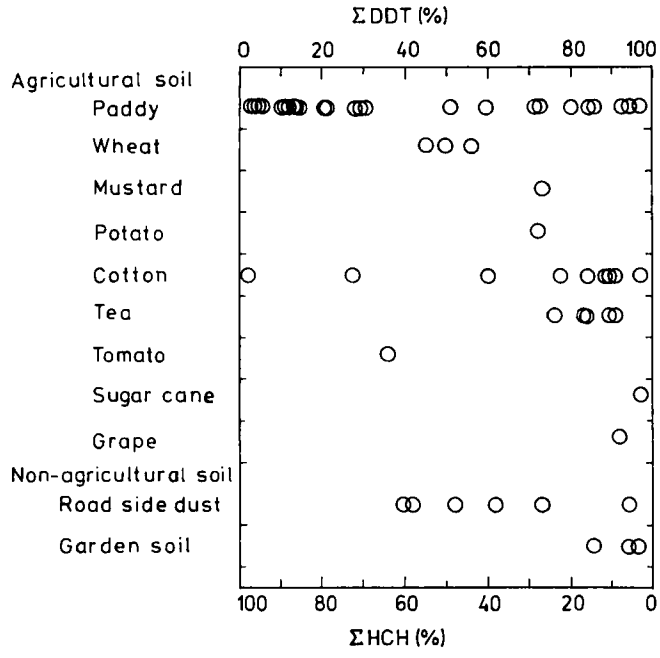

Figure 4 Relative proportion (%) of ΣDDT and ΣHCH residues in paddy, upland, and urban soils from India.

revealed that cotton ranks first (11) followed by vegetables and fruits (4 and 3, respectively), and rice (0.71). Thus, the relatively large use of insecticides on cotton crop account for the higher residue levels of DDT and HCH detected in cotton field soil.

The relative proportions (on percentage basis) of Σ DDT and Σ HCH are given in Figure 4. In the case of Σ DDT, upland soils registered higher levels than those of paddy as well as urban soils. Although high concentrations of DDTs were detected in some samples, in general the levels in paddy soils seem to be relatively lower than upland soils. It could be attributed to the relatively smaller quantity of DDT used for paddy. Our earlier studies[12] also revealed low levels of DDT in paddy soils from the Vellar river watershed, South India. Despite the fact that the use of DDT was restricted for agricultural purposes in India[14], its levels in some upland soils (for e.g. cotton, tea, sugarcane and grape orchard soils) were high. These may be residues from the insecticides applied so far. Only 15% of the DDT produced in India is used for agricultural purposes.[15] Even though the overall agronomic utilisation of DDT is less, upland soils might be receiving a major share of this DDT as evidenced by their residue levels.

The concentrations of HCHs in upland soil seem to be less scattered than those of DDTs and nearly in the same range as those of paddy soils. HCH levels in paddy and urban soils were comparable. The overall pattern of HCH residue levels could

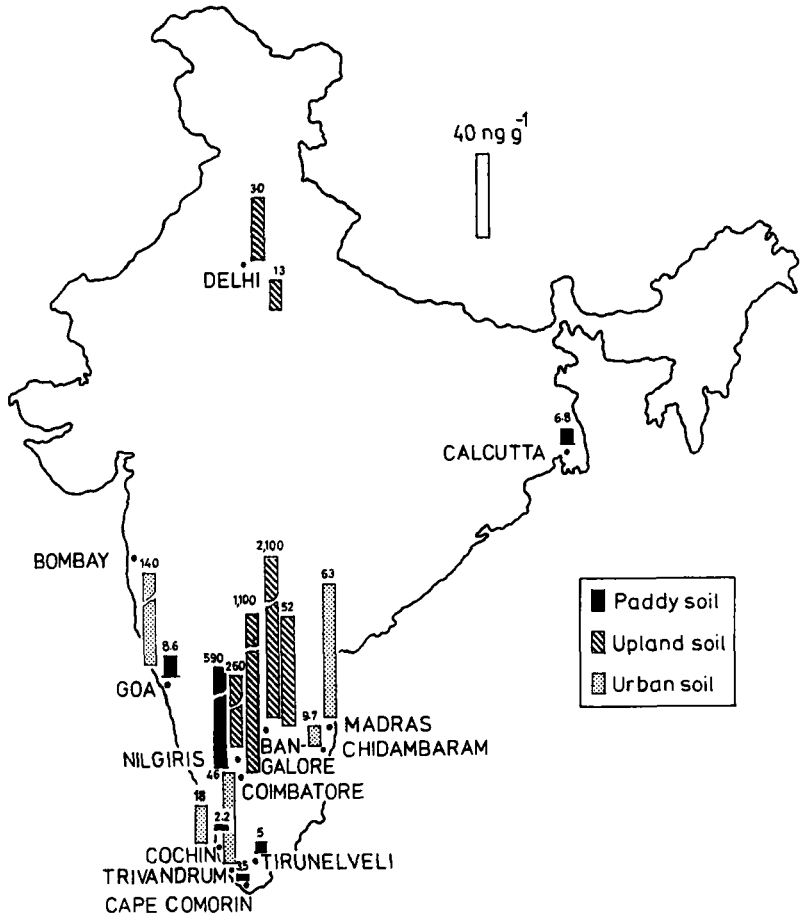

Figure 5 Spatial distribution of ΣDDT concentrations in paddy, upland, and urban soils from India. Numbers on the top of bars indicate the concentrations.

be explained by the fact that this insecticide is used largely for a wide range of agricultural purposes in India,[16] resulting in uniform contamination of the soil. This is significant in the light of our earlier studies which have shown high levels of HCH in rice and other agricultural foodstuffs from upland crops[3] as well as air, water, soil and sediment[9,10,12] and also human breast milk.[1]

Spatial distribution of Σ DDT and Σ HCH concentrations are shown in Figures 5 and 6. Comparison of concentrations on a regional basis may not be possible due to the small number of samples analysed from some localities. Nevertheless, the considerable levels of residues detected in all the places surveyed indicate the extent of environmental contamination by these persistent chemicals.

Though it is difficult to critically evaluate the DDT and HCH contamination of urban soil due to the limited number of samples analysed, these residues detected

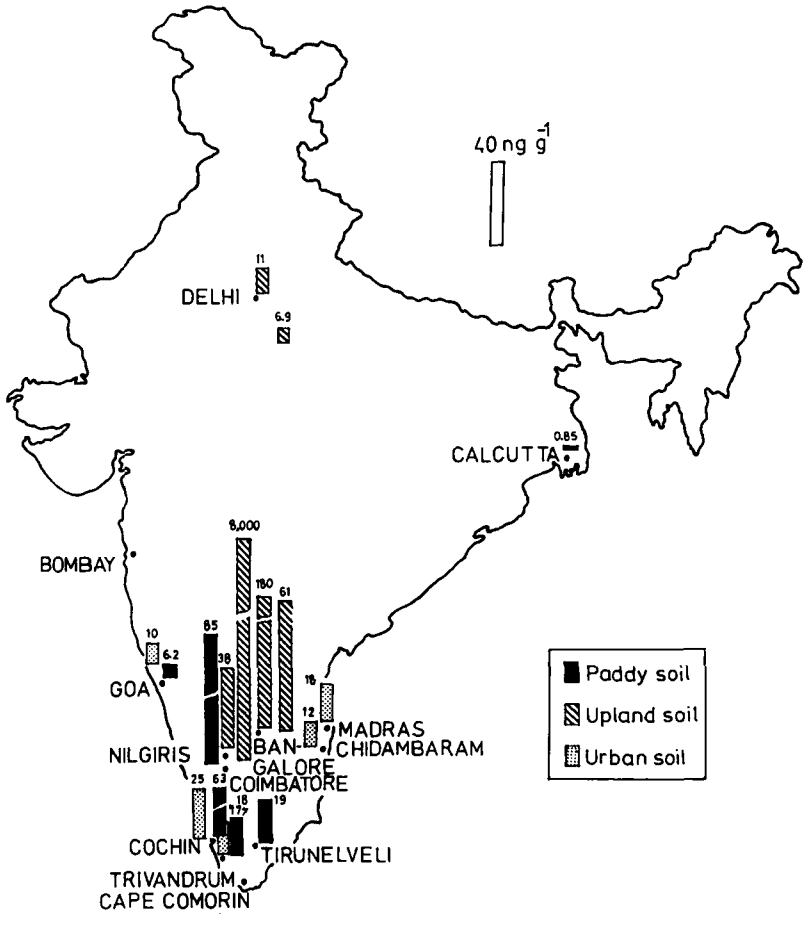

Figure 6 Spatial distribution of ΣHCH concentrations in paddy, upland, and urban soils from India. Numbers on the top of bars indicate the concentrations.

have the same concentration levels suggesting the probable sources such as spray drift of insecticides from agricultural areas and also their application in considerable amounts for vector control purposes in urban and suburban areas. However, DDT levels were relatively higher than those of HCH in many urban soil samples (Figure 4). This could be due to the larger consumption of DDT for vector control purposes, compared to that of HCH.[16,17]

The concentrations of DDTs and HCHs recorded in the present study were compared with the data reported so far from various places in the world (Table 3). Although the mean concentration of DDTs detected in Indian paddy soils were comparable to those detected elsewhere, the maximum concentrations were relatively higher than those measured elsewhere. On the other hand, the mean and maximum concentrations of HCHs in Indian upland soils were relatively higher than those

Table 3 Concentration (range and mean) of ΣDDT and ΣHCH in soils reported from various places in the world

Type of sample	*Location*	*Survey year*	*Concentration (ng g^{-1} dry wt.)*		*References**
			Σ DDT	*Σ HCH*	
Paddy field soil	Japan	1968–70	NA	200–1,000	20
	U.S.A.	1972	10–940[a] (80)[b]	NA	25
	Indonesia	1982	0–20 (1.2)	0.5–20 (8.9)	26
	Taiwan	1981	ND–120 (12)	ND–36 (1.9)	27
	Malaysia	1981	4.7–5.8 (5.3)	2.8–8.8 (5.3)	28
	India	1988	0.25–4.3 (1.9)	1.1–1,100 (120)	12
		1988	0.85–2,200 (99)	0.42–280 (34)	Present study
Upland soil	Japan	1970	Tr.–8,300 (810)	76–8,300 (790)	29
	U.S.A.	1971	10–390,000 (610)	NA	30
	Indonesia	1982	0.6–24 (10)	0.8–11 (3.9)	26
	Taiwan	1980	5.4–25 (15)	1.2–6.0 (3.5)	31
	India	1988	2.0–4,400 (650)	2.9–86,000 (3,800)	Present study
Urban soil	U.S.A.	1975–1976	6–405	NA	32
	India	1974	10–2,600	NA	33
	India	1988	3.4–190 (61)	0.55–27 (16)	Present study

[a] Range (Min.–Max.).
[b] Arithmetic mean.
NA: not analysed.
* Though many published reports are available on organochlorine insecticide residues world-wide, data from representative countries which are still using these insecticides or during their peak usage period was selected for the sake of comparison. For e.g. data quoted here for U.S.A. and Japan refers to the period of maximum usage of these insecticides. In the case of some countries in the tropical and subtropical belt, the figures given here represents the period of relatively active use of these chemicals.

measured in similar soils of other regions. Results of our earlier investigations,[1,9–12] already emphasized the extensive and recent use of HCH for agricultural purposes in India. This trend is likely to continue in this country in the future because an increasing demand for this insecticide is expected. On the other hand, DDT was banned for agricultural purposes in India, recently.[18] Hence, its use in this sector is likely to decrease and there is a possibility that its residue levels also decrease in the future.

Composition of DDT compounds and HCH isomers were investigated in detail. The percentage of *p,p'*-DDT in paddy and upland soils seem to be lower than those of other metabolites. On the contrary, this compound contributed to a higher content among all DDT compounds in urban soil. This trend indicates the continuing input of technical DDT formulation into the environment mainly as a result of vector control operations mentioned earlier.

The percentage composition of DDTs in paddy and upland soils was found to be in general in the order of *p,p'*-DDE > *p,p'*-DDT > *p,p'*-DDD. Our earlier studies in Vellar river watershed, South India[12] also showed higher levels of *p,p'*-DDE in paddy field soils. However, in the present study, some upland soils (for e.g. tea field and grape orchard) recorded relatively higher percentage of *p,p'*-DDT when compared

to those of cotton field soils, indicating presumably a slower degradation rate under colder climate.

As regards the percentage composition of HCH isomers, in wheat, mustard, grape, and urban soils, the proportion of HCH isomers were in the order of β-HCH > α-HCH > δ-HCH > γ-HCH. However, in some samples such as cotton, tea, and some other upland soils, the composition of HCHs was more or less similar to that of the technical grade HCH. These variations in percentage composition could be attributable to their physico-chemical properties and time elapsed after the application. Technical grade of HCH used in India contains 70% of α, 9% of β, 14% of γ and 7% of δ.[9] Although the α isomer constitutes the largest proportion in technical HCH, its high vapour pressure and water solubility might have contributed to its loss from soil through rapid volatilization and water run-off, respectively. On the other hand, β-HCH has lowest vapour pressure and water solubility, but most stable among HCH isomers and relatively resistant to microbial degradation.[19] Such physico-chemical properties of β-HCH account for its high levels in soil.

A specific pattern in residue behaviour could be seen when the data recorded in the present study (representing a tropical area) was compared with those measured in temperate areas. When the concentrations of DDT and HCH residues in paddy field soils from South and North India (monitored in the present study) were compared with those from Japan (during their period of maximum use in the late 1960's and early 1970's,[20]), the levels of DDTs and HCHs in Indian samples were apparently lower. Though the average per hectare consumption of insecticides in India and Japan are similar (for e.g. HCH is applied, respectively, at about 2.5 kg ha^{-1} [21] and 3.1 kg ha^{-1} [22]), the Indian climatic conditions favour rapid volatilization of the residues from the soil. Previous studies conducted in an experimental paddy field in Vellar river watershed, South India, revealed that more than 90% of the HCH applied was volatilized within two weeks after insecticide application.[23]

Though tropical climate favours rapid volatilization of residues, continued usage of these chemicals over the years contribute to higher levels in soil. The residues accumulated in soil will ultimately find their way into crops. Singh *et al.*,[24] noted uptake of organochlorine insecticides by plants in significant proportions. A variety of agricultural products such as cereals, pulses, spices and oils from India have been reported to be contaminated by insecticide residues.[3] In this context, contamination by persistent organochlorines could have harmful consequences unless pertinent legislative measures are taken to phase out the use of these chemicals.

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