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Temporal Variation of Persistent Organochlorine Residues in Soils from Vietnam

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Abstract—A program to monitor the residual levels of some important groups of organochlorine compounds (DDTs, HCHs and PCBs) in soils from different locations in Vietnam has been carried out since 1990. So far, 480 soil samples have been collected and analyzed to understand contamination levels of the soils by these compounds.

In general, concentrations of the analyzed compounds are in the following order of ∑DDTs > ∑PCBs > ∑HCHs. Concentrations of DDTs ranged from 0.02 ng g^{-1} to 1300.40 ng g^{-1}; PCB concentrations ranged from 0.61 ng g^{-1} to 9960.00 ng g^{-1} whereas that of HCHs spanned from 0.05 ng g^{-1} to 71.00 ng g^{-1}. Distributions of the chemicals varied by chemical and soil type. Higher concentrations of DDTs were recorded in agricultural soils, whereas higher levels of PCBs were found in soils from industrial areas. The concentrations of HCHs appeared not to vary much by soil type.

Investigation of the ratios of DDT and HCH isomers in the soil samples indicated that there have been no recent inputs of these chemicals in the areas studied. Specific analysis of PCB isomers and congeners showed that six groups of PCBs (from *tri*-chlorinated to *octa*-chlorinated) were detected in the analysed samples. The mean percentage proportions of six selected PCB congeners followed the order of PCB138 > PCB 153 > PCB101 > PCB52 > PCB180 > PCB28.

It is worth noting that the concentrations of DDTs and HCHs have gradually decreased over time from 1990 to the present. Concerning PCB levels in soil, an increasing trend over time can clearly be seen, which is further confirmed by high PCB residues detected in recently collected (2005–2007) soil samples.

Keywords: organochlorine compounds, soil contamination, DDT, HCH, PCB

INTRODUCTION

Of all the chemical compounds with potential environmental and human health

Table 1. Details of samples and sampling locations.

Location and type of soil	Number of samples with six sampling periods						Sub total
	1990–1992	1993–1995	1996–1998	1999–2001	2002–2004	2005–2007	
Habac	5	10	10	10	12	10	57
Hanoi	30	10	10	10	20	20	100
Haihung	5	10	10	10	10	8	53
Quangtri	12	6	6	6	8	7	45
TT-Hue	12	6	6	6	8	7	45
HCM City	5	10	10	10	15	10	60
Songbe	5	10	10	10	10	10	55
Tayninh	30	10	10	5	5	5	65
Total							480

impacts, those that are chlorinated have received the most attention. Organochlorine insecticides, chlorobiphenyls, chlorophenols, dioxins and furans are all examples of chlorinated compounds frequently listed as priority pollutants due to their stability, lipophility and bioaccumulation.

Like most parts of the world, Vietnam has imposed a total ban and/or restricted the use of organochlorine insecticides for special purposes (MARD, 1994). But only recently, PCBs have been listed as priority pollutant.

Long term careless use of toxic chemicals including organochlorine insecticides and PCBs has definitely lead to negative effects on human health and the environment. This study was conducted to understand the extent of soil contamination in Vietnam by these compounds.

MATERIALS AND METHODS

Four hundred and eighty samples of various soil types from different locations throughout Vietnam were randomly collected during the period from 1992 to 2007. Details of the samples, collection time and sampling locations are given in Table 1. In most cases, 5 cm surface soil layers were taken and used for chemical analysis. Soils from at least five points in a field were dug and mixed thoroughly making up one sample. The samples were placed into chemically precleaned polyethylene bags and brought to the laboratory. In the laboratory, the soils were air dried, grinded, sieved and kept in precleaned glasses under cool temperature until analysis was performed. The soils were solvent extracted. The extracts were further cleaned up and concentrated as required. The compounds of interest were separated and determined.

RESULTS AND DISCUSSION

Tables 2–4 show concentrations of ∑DDT, ∑HCH and PCBs. The order of residue levels of these chemicals was mainly ∑DDT > PCBs > ∑HCH in soils from Vietnam although elevated levels of PCBs were found in some samples. Relatively high levels of ∑DDT and PCBs were recorded in many samples from

Table 2. Concentrations (min. to max.) of DDTs.

Location and type of soil	Concentration range (ng g^{-1}) of DDTs					
	1990–1992	1993–1995	1996–1998	1999–2001	2002–2004	2005–2007
Habac:						
Rural soil	1.7 to 307.3	1.5 to 329.2	2.0 to 297.5	1.1 to 198.7	2.5 to 155.6	1.1 to 163.5
Urban soil and dust	1.2 to 53.4	1.2 to 48.6	1.3 to 58.6	0.9 to 42.4	1.3 to 41.3	
Hanoi:						
Rural soil	530.8 to 930.5	434.5 to 940.5	545.8 to 870.5	430.8 to 850.5	378.6 to 627.7	251.2 to 420.9
Urban soil and dust	59.7 to 970.6	159.7 to 1,070.6	49.7 to 987.6	51.7 to 789.6	43.1 to 546.6	61.2 to 379.8
Haihung:						
Rural soil	1.3 to 204.3	2.4 to 247.3	0.8 to 197.6	0.9 to 214.3	0.8 to 211.2	0.9 to 198.3
Urban soil and dust	1.2 to 33.4	1.1 to 43.5	1.4 to 23.9	1.3 to 39.4	1.3 to 35.4	1.2 to 40.4
Quangtri:						
Rural soil	1.1 to 1,300.4	1.2 to 1,050.	1.6 to 1,289.4	1.1 to 1,300.4	1.6 to 1,210.4	1.6 to 872.1
Urban soil and dust	1.7 to 307.3	1.5 to 267.4	0.9 to 312.5	1.4 to 295.6	1.2 to 252.6	1.9 to 195.5
Thuathien-Hue:						
Rural soil	1.5 to 938.2	ND to 1,038.4	1.3 to 878.5	1.2 to 1,051.4	1.4 to 951.4	1.2 to 804.3
Urban soil and dust	0.8 to 197.3	1.8 to 242.1	1.6 to 292.1	1.9 to 285.1	1.7 to 255.1	1.2 to 185.7
Ho Chi Minh City:						
Rural soil	49.5 to 1,067.6	56.7 to 987	64.7 to 1,042.6	29.5 to 870.6	18.5 to 789.6	19.5 to 690.6
Urban soil and dust	151.8 to 480.6	130.6 to 330	210.5 to 510.4	170.8 to 530.5	168.8 to 460.4	180.6 to 390.5
Songbe:						
Rural soil	230.7 to 830.5	180.7 to 860.4	246.7 to 630.5	156.7 to 720.7	136.5 to 620.8	135.5 to 580.8
Urban soil and dust	59.7 to 361.6	107.7 to 406.6	79.5 to 475.6	82.5 to 368.6	62.5 to 468.7	60.3 to 508.7
Tayninh:						
Rural soil	1.4 to 297.5	2.5 to 206.3	1.8 to 311.4	2.4 to 234.1	2.1 to 247.2	1.5 to 197.6
Urban soil and dust	1.6 to 73.4	1.3 to 64.1	1.2 to 69.8	1.5 to 81.4	1.3 to 61.6	1.2 to 65.9

Table 3. Concentrations (min. to max.) of HCHs.

Location and type of soil	Concentration range (ng g^{-1}) of HCHs					
	1990–1992	1993–1995	1996–1998	1999–2001	2002–2004	2005–2007
Habac:						
Rural soil	0.15 to 24.23	0.23 to 28.41	0.16 to 19.87	1.12 to 28.96	0.86 to 13.85	0.35 to 12.88
Urban soil and dust	9.42 to 37.28	12.26 to 42.70	6.79 to 48.32	5.38 to 39.17	5.72 to 38.42	5.73 to 29.32
Hanoi:						
Rural soil	8.21 to 36.15	5.61 to 40.73	9.03 to 33.75	3.28 to 24.62	3.35 to 19.64	4.31 to 17.64
Urban soil and dust	17.91 to 71.00	23.48 to 66.57	14.32 to 69.51	11.37 to 69.84	9.67 to 56.34	8.66 to 49.64
Haihung:						
Rural soil	0.16 to 17.12	0.23 to 12.87	0.27 to 9.54	0.31 to 5.69	0.37 to 4.62	0.82 to 3.69
Urban soil and dust	5.21 to 21.98	1.32 to 12.70	4.31 to 18.28	3.89 to 17.85	2.91 to 15.82	2.56 to 13.32
Quangtri:						
Rural soil	0.15 to 6.98	0.61 to 12.26	0.21 to 13.09	0.22 to 9.08	0.12 to 8.48	0.11 to 6.42
Urban soil and dust	0.45 to 18.30	2.43 to 21.08	0.36 to 16.42	3.12 to 146.97	2.16 to 124.23	2.17 to 114.12
Thuathien-Hue:						
Rural soil	2.31 to 16.92	5.26 to 34.19	2.75 to 23.01	1.74 to 22.91	1.62 to 12.81	1.44 to 17.71
Urban soil and dust	4.52 to 29.87	13.92 to 36.95	11.84 to 55.52	15.98 to 54.15	12.93 to 34.11	11.68 to 44.15
Ho Chi Minh City:						
Rural soil	7.59 to 32.86	18.96 to 40.93	9.08 to 33.97	12.19 to 38.98	11.49 to 28.97	11.62 to 23.47
Urban soil and dust	19.12 to 69.78	22.31 to 70.88	17.79 to 59.77	15.53 to 66.71	13.43 to 46.79	12.46 to 41.25
Songbe:						
Rural soil	8.81 to 35.02	15.95 to 4.65	7.99 to 31.29	11.13 to 50.27	9.13 to 39.29	8.12 to 29.89
Urban soil and dust	22.93 to 70.18	19.93 to 67.58	14.59 to 54.74	19.71 to 56.92	13.74 to 46.84	10.76 to 39.88
Tayninh:						
Rural soil	0.18 to 13.36	2.12 to 19.94	1.33 to 8.06	2.01 to 13.61	2.17 to 11.65	1.97 to 9.66
Urban soil and dust	0.66 to 27.70	1.84 to 34.28	5.16 to 25.47	2.23 to 21.48	1.93 to 12.41	1.43 to 11.51

Table 4. Concentrations (min. to max.) of PCBs.

Location and type of soil	Concentration range (ng g^{-1}) of PCBs					
	1990–1992	1993–1995	1996–1998	1999–2001	2002–2004	2005–2007
Habac:						
Rural soil	0.61 to 13.04	0.69 to 15.71	0.79 to 17.86	2.12 to 15.39	4.11 to 25.29	5.13 to 29.37
Urban soil and dust	1.60 to 25.74	2.57 to 24.95	3.72 to 35.83	4.05 to 39.27	5.85 to 44.22	7.15 to 49.23
Hanoi:						
Rural soil	72.03 to 179.06	57.08 to 181.06	61.07 to 229.01	72.03 to 281.06	88.03 to 321.43	67.38 to 369.71
Urban soil and dust	1.60 to 8,817.00	9.70 to 9,512.0	19.60 to 8,675.00	21.60 to 9,167.00	29.66 to 9,082.13	52.71 to 8,992.15
Haihung:						
Rural soil	1.04 to 12.75	0.88 to 14.67	1.33 to 245.71	1.76 to 26.08	3.98 to 36.82	9.62 to 56.07
Urban soil and dust	2.01 to 21.98	3.05 to 22.94	3.87 to 59.73	4.41 to 67.95	4.77 to 89.91	6.74 to 87.97
Quangtri:						
Rural soil	0.68 to 6.62	0.78 to 13.41	1.02 to 8.47	2.28 to 14.05	2.49 to 24.12	3.39 to 37.15
Urban soil and dust	1.01 to 16.78	2.89 to 27.11	4.46 to 34.72	5.33 to 36.96	6.34 to 45.21	9.11 to 55.73
Thuathien-Hue:						
Rural soil	0.88 to 5.67	1.43 to 9.85	1.79 to 11.93	1.03 to 15.37	2.83 to 25.77	1.88 to 30.02
Urban soil and dust	2.01 to 18.35	3.42 to 19.06	1.95 to 29.97	2.37 to 31.22	4.76 to 44.13	6.61 to 51.15
Ho Chi Minh City:						
Rural soil	3.20 to 179.37	6.03 to 150.92	6.21 to 170.37	4.09 to 191.24	5.91 to 201.22	4.94 to 231.76
Urban soil and dust	4.45 to 9,960.71	5.08 to 7,698.21	3.48 to 7,584.21	9.26 to 6,987.25	8.62 to 6,829.53	8.44 to 6,865.29
Songbe:						
Rural soil	1.17 to 121.86	3.15 to 145.62	2.73 to 221.65	5.12 to 119.45	4.66 to 91.34	4.43 to 82.89
Urban soil and dust	1.69 to 879.04	3.49 to 1,579.09	8.92 to 1,975.08	5.94 to 479.41	6.73 to 503.81	7.65 to 521.42
Tayninh:						
Rural soil	2.03 to 320.00	2.31 to 20.00	1.45 to 280.18	3.14 to 29.00	5.23 to 32.17	5.41 to 33.49
Urban soil and dust	3.31 to 29.91	2.12 to 41.96	3.31 to 59.99	1.45 to 39.26	2.67 to 43.15	3.26 to 49.69

Fig. 1. Map showing sampling locations.

Vietnam indicating the irregular distribution of these compounds in soils of the country. On the contrary, low and uniform levels of ∑HCH were obtained from the soils. With regard to residue distribution in different soil types, DDT and HCH concentrations in soils from paddy fields were higher than those in upland and urban (garden and roadside) soils, whereas PCB concentrations in urban soils were higher than those in other soils.

DDTs and HCHs

∑DDT in Vietnamese paddy fields, upland soil and urban and street dust samples ranged widely from 0.8 to 1,300.4 ng g^{-1}. Although most of the agricultural areas revealed relatively low residue levels of DDT, very high concentrations were found in some samples, suggesting the sporadic application of DDT for agricultural purposes or the sampling locations were near where DDT was used for sanitation purposes. In addition, these high levels were probably due to the ignorance of farmers or the use of DDT for combating plague in the absence of other insecticides. DDT concentrations were mostly higher in paddy fields than in upland and street dust samples indicating that DDT is used mainly to protect

rice, the most important crop in Vietnam, from insects.

In the case of ∑HCH, the concentrations in paddy fields, upland soil and street dust samples of Vietnam ranged from 0.15 to 71 ng g^{-1}. Similar to DDT, HCH concentrations from paddy soil samples were also higher than those in upland samples. But the levels were almost in the same range with street dust samples. A detailed comparison would be difficult as the residual levels of HCH were all in the low range. However, HCH residues seemed to be less scattered than those of DDTs. Though the levels were not very high, the presence of HCHs in all types of samples was clearly marked.

Among the two insecticides, HCHs showed lower residue levels than those of DDTs in soil samples from Vietnam. This could be due to differences in their physico-chemical properties (Tanabe and Tatsukawa, 1981). The dissipation rates of HCHs are higher than those of DDTs, because of higher vapour pressure and water solubility of the former compared to that of the latter. Besides, HCH may have been used to a lesser extent as compared to DDT.

The concentrations of ∑DDT and ∑HCH from Vietnam obtained in this study were briefly compared with data reported earlier from several other countries. The concentrations of DDTs in soils from paddy fields in Vietnam were the same as those from India, with a range of 0.85–2,200 ng g^{-1} and a mean of 99 ng g^{-1} (Kawano *et al*., 1992).

In the case of HCHs, the residue levels obtained by our analysis of soils from Vietnamese paddy fields were relatively lower than those from India (range 0.42–280 ng g^{-1}; mean of 34 ng g^{-1}, Kawano *et al*., 1992). Similarly, the concentrations of HCHs in upland and urban soils recorded in this study were lower than those from Indonesia (range 4.9–18 ng g^{-1}; mean of 11 ng g^{-1}, Kuwatsuka *et al*., 1986) and India (range 2.9–86,000 ng g^{-1}; mean of 3,800 ng g^{-1}, Kawano *et al*., 1992).

A wide range of variation in the percentage composition of DDT and its metabolites in the soil samples was observed. In samples from paddy fields from northern and southwestern Vietnam, residue levels of DDTs were found to be in the order of *p*,*p*′-DDE > *p*,*p*′DDD > *p*,*p*′-DDT > *o*,*p*′-DDT meanwhile those from the central and southern region were in the order of *p*,*p*′-DDD > *p*,*p*′-DDE > *p*,*p*′-DDT > *o*,*p*-DDT. This is significant by the fact that under aerobic conditions, high formation rate of *p*,*p*′-DDE from *p*,*p*′-DDT takes place whereas the turnover of *p*,*p*′-DDT into *p*,*p*′-DDD is more rapid under anaerobic conditions (Richard, 1976). During the dry season in which samples were taken in northern and southwestern Vietnam, with aerobic conditions occurring in the soil the active oxidative transformation of *p*,*p*′-DDT to *p*,*p*′-DDE was facilitated. On the other hand, in central and southern areas of Vietnam, the predominance of *p*,*p*′-DDD was found perhaps because of constant rains during the wet season coupled with anaerobic conditions in the soil. Climate appears to be important factor for the transformation of DDT to its metabolites. In addition, a relatively high rate of *p*,*p*′-DDT was found in some samples probably suggesting that technical grade DDT was deposited to the soils just a short time prior to the sampling date.

HCHs exhibited markedly different behavior, judging from the specific percentage composition of HCH isomers. Some samples from paddy fields

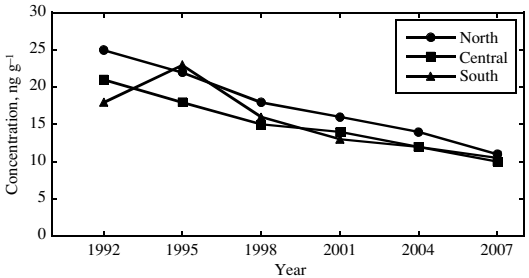

Fig. 2. Median concentration of ∑HCH vs. time.

showed higher levels of β-HCH. Among the isomers, β-HCH has the lowest water solubility and vapour pressure, but is the most stable and relatively resistant to microbial degradation (Bachmann *et al.*, 1988). These physico-chemical properties of β-HCH may explain its higher levels found in some samples. A larger proportion of α-HCH and γ-HCH isomers was found in some samples suggesting the effects of technical grade HCH, which contains large amounts of α and γ isomers, sprayed or drifted into the soils. A relatively higher percentage of γ-HCH in some other samples was observed, probably due to the usage of lindane (γ-HCH) for vector control.

PCBs

Concentrations of PCBs in paddy fields, upland soils and street dust samples from Vietnam ranged from 0.61 to 320 ng g^{-1}, from 1.1 to 6.2 ng g^{-1} and from 1.6 to 9960 ng g^{-1}, respectively. Levels of PCB were highest in street dust samples, followed by soils from paddy fields. Since threshold values of PCBs in the soil environment have not yet been defined (Pal *et al.*, 1980), it would be difficult to make a specific evaluation. However, PCB residues could be detected in all soil samples which indicate that PCB contamination of soil from Vietnam has been taking place.

The assumption that there exists an even and equal PCB distribution in soils may not be correct. Higher levels of PBCs are expected in the industrialized zones where the production and utilization of PCBs are more intensive. Also, the levels would be higher in soils from and downwind of urban areas, compared to those of rural and agricultural areas. Pal *et al.* (1980) reported a mean PCB level of 100 ng g^{-1} in surface soils from the U.S. and Canada in the order of a few μg g^{-1} to 100 μg g^{-1} in urban areas, but less than a few ng g^{-1} in rural forest and agricultural areas.

Data obtained from the present study seems to be significant in the light of the above remarks. In Vietnam soils, high levels of PCBs were observed in some samples from suburban areas of the cities of Hanoi and Ho Chi Minh. This is apparently due to the fact that the sampling sites were close to industrial areas

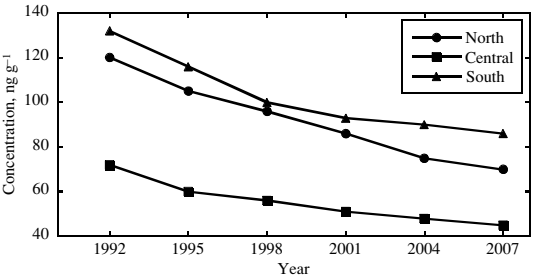

Fig. 3. Median concentration of ∑DDT vs. time.

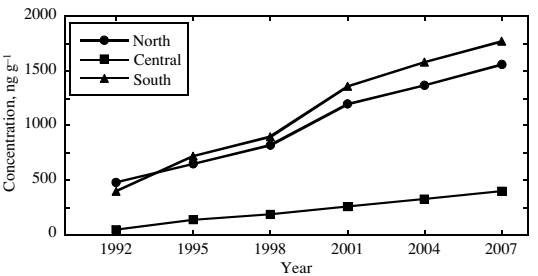

Fig. 4. Median concentration of ∑PCB vs. time.

with factories for textile-dying, wood processing, and transformer and chemical manufacturing. It may be suggested that even in an underdeveloped country with small industries, soil contamination by PCB can occur in some point sources for PCB storage and usage, and their ambient areas, especially in the case where facilities for proper use and disposal of PCB are lacking.

In big cities like Hanoi and Ho Chi Minh, surprisingly high concentrations of PCB were detected, reaching the values of 8817 and 9960 ng g^{-1}, respectively. This indicates that PCB has been intensively used in these cities and the chemicals have found their way to the environment.

It is worth to noting that the concentrations of HCHs and DDTs have gradually decreased over time from 1990 to the present. A decrease in ∑DDT and ∑HCH contamination can be expected over time as long as these substances are not used anymore. Concerning the levels of PCB in soil, an increasing trend can clearly be seen, which is further confirmed by high PCB residues detected in recently collected (2005–2007) soil samples. Therefore, adequate management and disposal of PCB from sources would help to prevent further PCB release to the environment. Temporal variation of residual levels of the above mentioned chemicals are clearly seen in Figs. 2–4.

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