# Detection and Movement of Dichlorodiphenyl Trichloroethane (DDT) Residues in Soil Pedons at Different Locations of Dhaka City in Bangladesh

<sup>1</sup>M.A. Zaman, <sup>2</sup>S.M. Ullah, <sup>3</sup>M.A.Z. Chowdhury and <sup>4</sup>M.A.U. Mridha

<sup>1</sup>Department of Soil Science, University of Chittagong, Chittagong, Bangladesh <sup>2</sup>Department of Soil, Water and Environment, University of Dhaka, Dhaka, Bangladesh <sup>3</sup>Institute of Food and Radiation Biology, Atomic Energy Research Establishment, Ganakbari, Savar, Dhaka, Bangladesh,

<sup>4</sup>Department of Botany, University of Chittagong, Chittagong, Bangladesh

Abstract: Soil samples were collected at different depths of soil pedons from five locations of Dhaka city to study the DDT residues using ELISA (Enzymed-Linked Immuno Sorbent Assay) method during pre-monsoon transition and rainy seasons. The result showed that surface layers of all pedons contain DDT while all the selected depths of two pedons at Taltala and Hazaribag were contaminated with DDT. The unaltered plough pan was found to resist the downward movement of DDT in the cultivated lands at Aminbazar, Vaturia and Ainkha, whereas at Hazaribag the leaching of DDT through soil profile occurred without any resistance along with water soluble fat rather than organic matter and clay. In Taltala, DDT moved downward by organic matter and soil colloid within the cracked plough layer during dry fallow period. Correlation between DDT and some physico-chemical properties of Taltala and Hazaribag soils showed that positive relationships of DDT with clay and organic carbon occurred at significant levels, although a negative correlation was found between DDT and sand of Taltala soil. No significant relations of DDT with silt, moisture, pH, total nitrogen and cation exchange capacity were found in the tested soils.

**Key words:** DDT • soil profile • Dhaka city • ELISA

### INTRODUCTION

Dichlorodiphenyl Trichloroethane (DDT) is an organochlorine insecticide used throughout Bangladesh mainly to control mosquito-borne malaria in 1970s. In Bangladesh, pesticide use started in mid 1950s and gained momentum in late 1960s with the introduction of green revolution through the use of HYV rice in the country. The pesticide consumption in the country has reached 151600 tons of formulated product and 2443.11 tons of active ingredients in 1990-2000 [1]. The use of pesticides started with Endrin, Benzene Hexachloride (BHC), DDT etc. all belonging to chlorinated hydrocarbons. The use of chlorinated hydrocarbons declines due to their long persistence, danger to terrestrial and aquatic lives and ecosystems.

DDT was unrestricted for public health purposes until it was banned in 1993. In Dhaka, like other parts of Bangladesh, pesticides are used for agricultural and other purposes. In metropolitan areas of Dhaka city, organochlorine (DDT) is used mainly for mosquito control.

DDT is very persistent in the environment, with a reported half-life between 2-25 years [2, 3]. The solubility of DDT in water is 0.003 mg/L [4]. Due to its extremely low solubility in water, DDT is retained to a greater degree by soils and soil fractions with higher proportions of soil organic matter [5]. These properties make DDT dispersed ubiquitously in the environment. When applied on a field, DDT may be taken up by plants, retained by soil or degraded. It may also be carried away long distance from its site of application or may reach surface waters by runoff, atmospheric transport, drift etc. [6]. Over very long periods of time, DDT may leach into groundwater, especially through soils with little soil organic matter. Consequently, considerable levels of organochlorine have been detected in air, soil, surface water and groundwater [7-13] where it may be available to organisms.

Immunological methods for detecting insecticide residues in food and environmental samples offer several advantages over gas chromatographic methods. While maintaining comparable sensitivity, they are simple, cost effective and can be adapted for field use [14, 15].

The purpose of the present study was to obtain baseline data on DDT concentration in soil profiles by ELISA which may pose an environmental concern because of the possibility that it will reach the water table and contaminate the ground water.

#### MATERIALS AND METHODS

Collection and storage of soil: Soil samples were collected from different depths (upper depth of 0-15 cm-U, middle depth of 15-30 cm-M, lower depth of 30-45 cm-L) of each pedon by the core sampler from five locations (Aminbazar, Vaturia, Ainkha, Taltala and Hazaribag) of Dhaka district during May-June period which covers both the pre-monsoon transition and rainy seasons. Soil series were described with the help of Soil Resources Development Institute in Dhaka. Samples were brought to the laboratory in cleaned poly bag and stored in refrigerator prior to analyses.

Analytical methods: Particle size distribution of the soils was determined by hydrometer method [16]. The textural classes were made by following Marshall's triangular co-ordinates as devised by the USDA [17]. Soil moisture was determined by constant weight drying in an oven at 110°C and Cation Exchange Capacity (CEC) was determined after extraction of the cations with ammonium acetate [18], pH was measured in soil-water suspension (1:2.5) using a corning glass electrode pH-meter. Organic carbon and total nitrogen were determined by wetoxidation method of Walkley - Black and micro-kjeldahl's method respectively [19]. C/N ratio was calculated by dividing the percentage of total carbon by the percentage of total nitrogen present in the soil. The extraction, analysis, calculation and recovery of DDT were made by the methodology of EnviroLogix Inc. [20]. The Envirologix DDT plate kit is a competitive enzyme-linked immunosorbent assay (ELISA) and used for detecting DDT residues in soil samples.

DDT extraction procedure: Soil sample (20 g each) was taken into 250 ml conical flask, 100 ml of double-distilled hexane and acetone (50:50-v:v) was added and shaken in the mechanical shaker (EYELA Multi-Shaker Mms) for 10 hours at 180 rpm. The extract was separated and the supernatant liquid phase was collected in the evaporating flask. Two further extractions with 50 ml hexane-acetone were done. The extract was concentrated to 2-5 ml using vacuum rotary evaporator and transferred to a glass vial followed by the complete evaporation of solvent under a mild stream of N<sub>2</sub>. The residue in the vial was ready for ELISA test.

Analysis: One twenty microliter of DDT assay diluent was added to each well. Twenty microliter of negative control, 20 μl of each calibrator, 20 μl of each soil sample extract were added to their respective wells in a plate layout which was setup previously. This same order of addition for all reagents was followed. The contents of the wells were mixed thoroughly by moving the strip holder. Sixty microliter of working DDT-enzyme conjugate was added to each well and mixed uniformly for 30 sec. The wells were covered with parafilm and kept for 1 hour. Finally, the covering was removed and the wells were flooded with water and shaken to empty. Hundred microliter of substrate (Acetyl Thiocholine) was added to each well and incubated for 30 min. Hundred microliter of stop solution (1.0 N HCl acid) was added to each well and mixed thoroughly. The well contents turned yellow and the plate was read within 30 min. A group of blank readings (200 µl water in each well) was also run. The Optical Density (OD) of the content of each well was measured by spectrophotometer at 450 nm wavelength.

**Calculation:** Mean Optical Density (OD) values were calculated for each standard or sample by subtracting OD values of negative control. The absorbances were converted to % Bo values by the following formula:

$$\% B_o = \frac{OD \text{ of standard or sample}}{OD \text{ of negative control}} \times 100$$

Standard curve (Fig. 1) was prepared by plotting % Bo of each calibrator against its DDT concentration ( $\mu g \ l^{-1}$ ) on a semi log scale. The total DDT content of each sample was determined by finding its % Bo value and the corresponding concentration level on the graph. The detection limit of Envirologix DDT plate kit was 0.025-0.875 ppm.

For the sample having % Bo higher than that of lowest calibrator, sample concentration is <0.025 ppm. For the sample having % Bo lower than that of higher calibrator, sample concentration is >0.875 ppm

**Recovery:** Soil samples (20 g each) were separately spiked with known doses of DDT standard mixed thoroughly and uniformly and then kept at 25°C for 2 days. The samples were extracted in the same way of unspiked samples as mentioned above. Mean recoveries found for soils were 84.67 and 83.33% (Table 1) for the first and second day respectively indicating the suitability of the methodology.

## RESULTS AND DISCUSSION

**Translocation of DDT into soil profile:** Physico-chemical characteristics of the tested soils are presented in Table 2.

Table 1: Recovery analysis by ELISA of soil samples spiked with different levels of DDT Day 1 (First day)

					Concentration			Mean
	Optical				determined by	Concentration	Recovery	Recover
Sample	density	Mean OD (±SD)	% CV	$\%\mathrm{B}_{\!\scriptscriptstyle 0}$	ELISA (ppm)	added (ppm)	(%)	(%)
Negative Control (NC)	1.195	1.209 (±0.020)	1.65	100.00	-	-	-	-
'0.00 ppm'	1.223							
Calibrator1 (C1)	0.860	0.840 (±0.028)	3.33	69.48	-	-	-	-
'0.025 ppm'	0.820							
Calibrator2 (C2)	0.615	$0.602 (\pm 0.018)$	2.99	49.79	-	-	-	-
'0.1 ppm'	0.589							
Calibrator3 (C3)	0.255	0.272 (±0.024)	3.82	22.50	-	-	-	-
'0.875 ppm'	0.289							
Soil-Blank	0.000	00	0.00	00.00	00.00	00.00	-	
Soil 1	0.420	0.404 (±0.023)	5.69	32.00	92.06	108.30	85	84.67
	0.388							
Soil 2	0.601	0.619 (±0.025)	4.04	49.00	17.76	21.66	82	
	0.637							
Soil 3	0.803	$0.827 (\pm 0.034)$	4.11	65.50	7.32	8.42	87	
	0.851							
Day 2 (Second day)								
Soil Blank	0.000	00	00	00.00	00.00	00.00	-	
Soil 1	0.445	0.435 (±0.014)	3.22	36.00	84.47	108.30	78	83.33
	0.425							
Soil 2	0.618	0.629 (±0.016)	2.54	52.03	17.55	21.66	81	
	0.640							
Soil 3	0.770	0.792 (±0.031)	3.91	65.50	7.66	8.42	91	
	0.814							

The soil samples at middle (M) and lower (L) depths of Aminbazar, Vaturia and Ainkha profiles contained higher amounts of clays than that of the upper (U) depths (Table 2). It may be due to the enrichment of clays rather than their downward movement. The same reason may be applied to organic matter, upon which soil is being developed, particularly in the areas near Hazaribag tannery factories. Soil pH ranged from 4.2 to 6.9, with relatively higher pH (6.9) in the upper depth of Taltala soil and lower pH (4.2) in the lower depth of Hazaribag soil. Total nitrogen varied from 0.02% in Vaturia soil to 0.20% in Hazaribag soil. The main source of total nitrogen in Hazaribag soil was the animal wastes derived from the nearby tannery factories. The C/N ratio ranged from 2.81 in Taltala soil to 20.40 in Ainkha soil. The most important differences among the soil profiles studied were in their Organic Carbon (OC) content and Cation Exchange Capacity (CEC). Taltala soil had the highest OC of 1.76% in the lower depth and Vaturia soil had the lowest OC of 0.2% in the middle depth. Cation exchange capacity varied from 8.91 to 48.51 meq/100 g

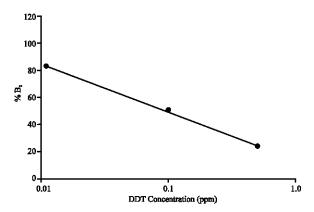


Fig. 1: Standard curve of DDT for soil

soil. The variations in CEC in soils of different profiles depend on the amount of both organic matter and clay contents in the soils.

DDT concentrations in different soil profiles are shown in Table 3. DDT was present throughout profiles of two locations namely at Taltala (Karail series) and

Table 2: Physico-chemical characteristics of the soils used in the investigation

	Depth		Particle size distribution (%)					Total	Cation Exchange		
Location	of soil	Moisture				Textural		Carbon	Nitrogen	C/N	Capacity (CEC)
(Soil Series)	(cm)	(%)	Sand	Silt	Clay	class	pН	(%)	(%)	ratio	meq/100 g Soil
Aminbazar	U: 0-15	23.0	42.3	44.6	13.1	Lo	5.6	0.96	0.14	6.85	8.91
(Khilgaon Series)	M:15-30	22.8	2.5	18.4	79.1	C	5.0	1.21	0.12	10.08	26.87
	L:30-45	21.9	3.2	18.5	78.3	C	5.0	0.73	0.09	8.11	25.36
Vaturia, Dakkhinkhan	U:0-15	22.0	17.2	53.8	29.0	SiCL	6.8	0.77	0.08	9.63	12.43
(Kalma Series)	M:15-30	20.1	11.4	59.8	28.8	SiCL	5.6	0.20	0.02	10.00	10.71
	L:30-45	21.0	13.0	58.0	29.0	SiCL	5.5	0.25	0.02	12.50	9.10
Ainkha, Keraniganj	U: 0-15	22.2	26.3	53.7	20.0	SiL	4.5	0.66	0.08	1.44	18.75
(Pagla Series)	M:15-30	22.5	2.4	54.0	43.6	SiC	6.4	1.02	0.05	20.40	17.48
	L:30-45	22.8	1.9	47.5	50.6	C	6.4	0.65	0.05	13.00	15.42
Taltala, Uttara	U: 0-15	22.6	19.6	39.7	40.7	CL	6.9	0.81	0.10	8.20	26.05
(Karail Series)	M:15-30	22.9	29.6	34.9	35.5	CL	5.2	0.45	0.16	2.81	33.48
	L:30-45	23.0	11.1	38.4	50.5	SiCL	5.4	1.76	0.17	10.35	48.51
Hazaribag	U: 0-15	21.3	14.9	64.4	20.7	SiL	6.2	1.23	0.20	6.15	10.13
(Disturbed soil)	M:15-30	20.8	30.2	60.2	9.6	SiL	4.5	0.37	0.10	3.37	9.39
	L:30-45	19.4	10.6	70.9	18.5	SiL	4.2	1.11	0.12	9.25	12.05

U-Upper, M-Middle, L-Lower, Lo-Loam; C-Clay; SiCL-Silty Clay Loam; SiC-Silty Clay; CL-Clay Loam; SiL-Silt Loam

Hazaribag (disturbed soils), whereas, the other three locations contained no DDT in the lower horizons except in surface horizons.

The distribution of DDT in the profiles at five locations is also shown in Fig. 2. The DDT is mainly concentrated at the surface of the soil profiles. The first three soils of Aminbazar, Vaturia and Ainkha contained 0.0012, 0.009 and 0.0052 ppm of DDT respectively at the surface soils (depth: 0-15 cm) only. The residues decreased with depth from the surface up to 15-30 cm and increased thereafter with depth in case of Taltala and Hazaribag soils (Fig. 2).

Three rice crops were grown frequently in the investigated area of Aminbazar, Vaturia and Ainkha. Consequently, the soil was kept wet for cultivation throughout growing seasons and there is no scope to dry up the soil and to crack down the plough pan formed at a depth of few cm below the surface due to intensive rice cultivation. Rice land is usually prepared by puddling under wet condition with the country plough. Due to the presence of compact plough pan, the vertical movement of pesticides like DDT, soil particles and organic matter with soil fluid in the lower horizons will be very limited, although lateral movement of soil fluid occurs in the soil naturally.

Another reason might be the migration of clay fractions and organic matter from the higher topographic position to the nearby lower position and thereafter accumulation occurred over the migrated clay and organic

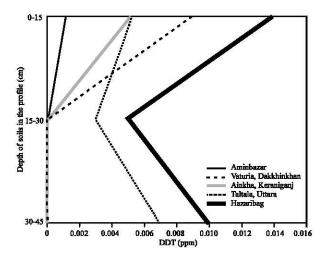


Fig. 2: The distribution of DDT residues in soil profiles at the five tested locations

fractions. Under wet conditions, enhanced accumulation of organic matter might occur rather than decomposition [21].

Soil colloids and dissolved organic matter led DDT to move down from the upper horizons to lower ones through the profiles of Taltala and Hazaribag areas. In the case of Taltala, the agricultural lands remain flooded during late June to late August and after receding the flood, rice is grown in a year. The land is kept fallow during part of dry season. Such condition favours heavy clayey surface soil or compact plough

Table 3: Concentrations of DDT residues in different depths of soil profiles

Location	Depth (cm)	Optical density (OD)	Mean OD values (±SD)	%CV	$^{\circ}\!\!/_{\!\!\mathrm{B}_{\mathrm{o}}}$	DDT concentration (ppm)
Aminbazar soil	U:0-15	0.748	0.737 (±0.016)	2.17	58.35	0.0012
		0.726				
	M:15-30	1.201	1.181 (±0.028)	2.33	93.51	ND
		1.161				
	L:30-45	1.214	1.201 (±0.018)	1.50	95.09	ND
		1.188				
Vaturia soil	U:0-15	0.394	0.404 (±0.014)	3.47	31.99	0.009
		0.414				
	M:15-30	1.180	1.163 (±0.024)	2.06	92.08	ND
		1.146				
	L:30-45	1.235	1.225 (±0.014)	1.14	97.00	ND
		1.215				
Ainkha soil	U:0-15	0.492	0.485 (±0.009)	1.86	38.40	0.0052
		0.478				
	M:15-30	1.207	1.195 (±0.017)	1.42	94.62	ND
		1.183				
	L:30-45	1.215	1.200 (±0.021)	1.75	95.01	ND
		1.185				
Taltala soil	U:0-15	0.494	0.485 (±0.014)	2.89	38.40	0.0052
		0.474				
	M:15-30	0.527	$0.537 (\pm 0.014)$	2.61	42.52	0.003
		0.547				
	L:30-45	0.463	0.448 (±0.021)	4.69	35.47	0.007
		0.433				
Hazaribag soil	U:0-15	0.281	0.303 (±0.031)	10.23	24.00	0.014
(Disturbed soil)		0.325				
	M:15-30	0.482	0.493 (±0.016)	3.25	39.00	0.005
		0.504				
	L:30-45	0.399	0.385 (±0.020)	5.20	30.50	0.01
		0.371				

layer to crack down. At this stage, the organic matter and soil colloids enhance the translocation of pesticides to the lower horizons with rainwater or irrigation water during latter agricultural practices.

Kan and Tomson [22] investigated the effects of Dissolved Organic Matter (DOM) on the transport of hydrophobic organic compounds like DDT in soil columns. They reported that DOM increased the movement of highly hydrophobic compounds such as DDT by a factor of a thousand or more.

It is known from the local inhabitants that the investigated site of Hazaribag was a depression area, in the past and was filled up with sediments transported from the nearby lands. The location is situated at the endpoint of drainage systems of Hazaribag where the entire discharged tannery wastes of leather factories are being deposited that make soils loose and friable.

One of the important characteristics of Hazaribag soil is that each layer contains a considerable amount of water soluble fat, which is visible during the soil extraction for pesticide analysis. The fat originated from raw skins of animals (i.e. cow, goat, sheep etc) during processing.

At Hazaribag, DDT was found distributed throughout the profile like that of Taltala (Table 3). Here, the transporting agents for DDT-leaching to the lower depth of soil are likely somewhat different from Taltala soil. The investigated area at Hazaribag is not used for agricultural purposes at all from the beginning and it has no plough pan below the surface soil. So, DDT residues are being translocated down through the loose friable soil profile by water soluble fat rather than by organic matter or soil colloids.

At Taltala and Hazaribag, the soils of lower horizons contained more DDT, although the middle horizons having low contents of DDT residues (Table 3). It is likely that the lower horizons are free from microbial degradation and volatilization and ideal zone for the DDT accumulation. Spencer *et al.* [23] stated that the concentrations of DDT in the deeper layer of soil profile

were greater where it is more protective from microbial degradation and volatilization.

The surface soils of all profiles contained low amounts of DDT ranging from 0.009 to 0.014 ppm. In the surface horizon, disappearance of DDT is largely due to adequate soil moisture and microbial activity, in addition to volatilization, hastened by heat and solar radiation [24].

**Correlation of DDT with physico-chemical characteristics of soils:** Soils of Taltala and Hazaribag profiles showed positive correlations (r = +0.97; p-0.05 for Taltala and r = +0.96; p-0.05 for Hazaribag) between DDT and clay particle, although a negative relationship (r = -0.99; p-0.01) existed between DDT and sand for Taltala soil (Table 4). Li [25] found that DDT and BHC increased with decreasing particle size. The amounts of DDT and BHC in <0.25 mm soil particles were 41-89% and 30-81%, respectively.

The adsorption of DDT on clay particles may be related to complexation of humic fraction with clay particle. In soil, the positive relationship of DDT with clay was found as these components favour the adsorption and persistence of non polar pesticides like DDT in soil. No significant relationships of DDT were observed with silt, soil moisture, pH, total nitrogen and CEC.

The studied soil moisture was adsorbed by soil components like clay particle and organic matter. Such kind of moisture usually has no function to change the DDT content in soil like loose water, because adsorbed moisture evaporates to the limited extent.

The effect of soil water content (saturated and unsaturated soil) on the fate of <sup>14</sup>c labelled DDT and DDE were studied by Boul [26]. He stated that <0.7% of the added DDT and DDE were mineralized in unsaturated soil and virtually none in saturated soil. The slow rate of DDT degradation in saturated soil is due to the soil conditions, rather than to a lack of degradative capacity within the soil microorganims. Willett *et al.* [27] reported that irrigating the soil with a small amount of water dramatically increased the volatilization flux over that from a unirrigated soil and reduced the amounts of DDT and metabolites due to continuous volatilization.

No significant relations of DDT and soil pH were found in soils of Taltala (r = +0.17) and Hazaribag (r = +0.75). However, Alam *et al.* [28] observed an inverse relationship between DDT and pH in soil.

DDT and organic carbon also showed positive correlations for soils of Taltala and Hazaribag (r = +0.95, p 0.05) (Table 4) because organic carbon content is directly related to organic matter content in soil.

Table 4: Correlation between DDT and physico-chemical characteristics of soils of Taltala and Hazaribag

	Correlation Co-efficient (r)				
Soil characteristics	Taltala	Hazaribag			
Sand	-0.99**	-0.78ns			
Silt	+0.75ns	+0.45ns			
Clay	+0.97*	+0.96*			
Moisture	+0.18 ns	+0.19ns			
pH	$+0.17\mathrm{ns}$	+0.75ns			
Organic carbon	+0.95*	+0.95*			
Total nitrogen	+0.075ns	+0.92ns			
C/N ratio	+0.98*	+0.50ns			
Cation exchange capacity (CEC)	+0.61ns	+0.33ns			

\*, \*\* Significant at  $P \le 0.05$ ,  $P \le 0.001$  probability levels respectively; nsnon significant

Guzzella [29] observed a similar significant correlation of DDT in lake sediment, Orta in Italy with organic carbon content.

Adsorption of non polar chlorinated hydrocarbon pesticides by lipid in organic matter has been described by Khan [30]. It has been suggested that the adsorption of non-polar pesticides on organic matter is mainly due to pesticide-lipid interaction. Quite generally, adsorption by soils of the non-ionic pesticide is positively correlated with soil organic matter content [31]. No significant relationships were found between DDT and total nitrogen or between DDT and CEC for both soils of Taltala and Hazaribag profiles (Table 4). Taltala soil presented a positive relationship (r = +0.98; p 0.05) between DDT and C/N ratio (Table 4), while Hazaribag profiles showed no significant relationship.

# CONCLUSIONS

DDT residues tightly sorbed by clay and organic matter fractions of soil matrix in the studied areas are negligible and their downward movement was restricted due to plough pan especially in agricultural lands while DDT is likely to be leached below through soils having no pans. Further investigation is required in different environmental matrices because DDT residues may be threaten to public health or living organisms by entering into food chain if their concentrations are high.

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