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# DDT residue in terrestrial environment in the Mount Darwin — Rushinga area: Zimbabwe

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**DDT and its metabolites were analysed in a Mount Darwin terrestrial environment. River sediments, water, soil, house dust, cow dung, grass, leaves, treebark and fish were taken from accessible sites along the Mount Darwin-Rushinga highway during the period March to November 1999. Results obtained indicate widespread contamination from trace residues of DDT. River sediments and house dust had high levels of DDT varying from 0.14 ng/g to 506.2 ng/g and 2.10 ng/g to 276.5 ng/g respectively. Other matrices had generally low residue levels.**

**Keywords:** DDT, pesticides, pesticide residue.

## Introduction

DDT has been widely used in developing countries in agriculture and public health programmes. Although it was successfully used to ensure production of adequate food supplies (Wessels, 1974, 1978; Mhlanga and Madziva, 1990) most of the pesticides found their way into soils, water and other parts of the terrestrial environment. In Zimbabwe, DDT and dieldrin have been used in tsetse fly and malaria control sprays (Mpofu, 1987). DDT has also been extensively used in agriculture prior to 1983 when its use in agriculture was banned. Continued use of DDT for control of disease insect vectors has remained in Zimbabwe for economic reasons (Mpofu, 1987; Matthiessen, 1983).

In tsetse fly control operations, DDT (75 percent wettable powder) is applied selectively at a concentration of 200 g ai/ha of spray area. Potential tsetse resting areas (tree trunks, holes, grass) are sprayed. After Zimbabwean independence in 1980, use of DDT increased especially in the north eastern border and western lying areas of the Zambezi valley (Figure 1). Spraying of DDT for malaria control also increased during the same time, though this was mainly confined to building interiors (Mpofu, 1987).

Different studies have been carried out to assess the environmental fate of these pesticides sprayed for malaria and tsetse fly control operations. Following tsetse fly

control spraying in 1968 to 1971 with DDT, Phelps and Billings (1972) carried out a study of the sprayed area by analysing animal species namely crocodiles, waterbuck, impala, elephant and flycatcher from the sprayed area. The study covered Kariba area, Mount Darwin (Chesa) and Chipinda pools as well as Harare. The study showed build up of DDT mainly in lakes. This was confirmed by Greichus *et al.* (1978) who did a similar study on Lake Chivero and obtained high levels of PCB's and DDT in the lake. In 1985 Mattheissen reported traces of DDT, DDE and other chlorinated pesticides in birds, bats, soil, tree bark and river sediment in areas along the North Western province of Zimbabwe, where DDT had been previously sprayed against tsetsefly and mosquito. Phelps *et al.* (1989) showed that DDT had moved up the food chain to fish whose consumption led to high levels of DDT in milk of breastfeeding mothers. Work by Chikuni *et al.* (1997) confirmed the presence of DDT and PCBs in breastfeeding mothers from various parts of the country. These pesticide contaminations of the aquatic ecosystem have been attributed to agricultural use (before its ban) and hence the environmental bioaccumulations along the food chain. Zaranyika *et al.* (1994) carried out a study of seven river bays on the Zimbabwean side of Lake Kariba. This area had received regular sprays of insecticides for malaria and tsetse fly control in 1984, (Mpofu, 1987). The results confirmed contamination of the bays by DDT and its metabolites.

In this paper a study of the terrestrial environment in Mount Darwin Rushinga area in the north eastern part of the country on the border with Mozambique, was carried out to assess the environmental impact of DDT and its metabolites in relation to previous tsetsefly and malaria control operations. Previous studies mainly focussed on lakes (Greichus *et al.*, 1978; Mhlanga and Madziva, 1990; Zaranyika *et al.*, 1994), in this paper we report analysis of sediments from rivers and dams, fish, soil, cow dung, grass and house dust collected along the accessible parts of Mount Darwin-Rushinga highway near the border with Mozambique. The last DDT spray in the area was carried out in 1986 (Figure 1).

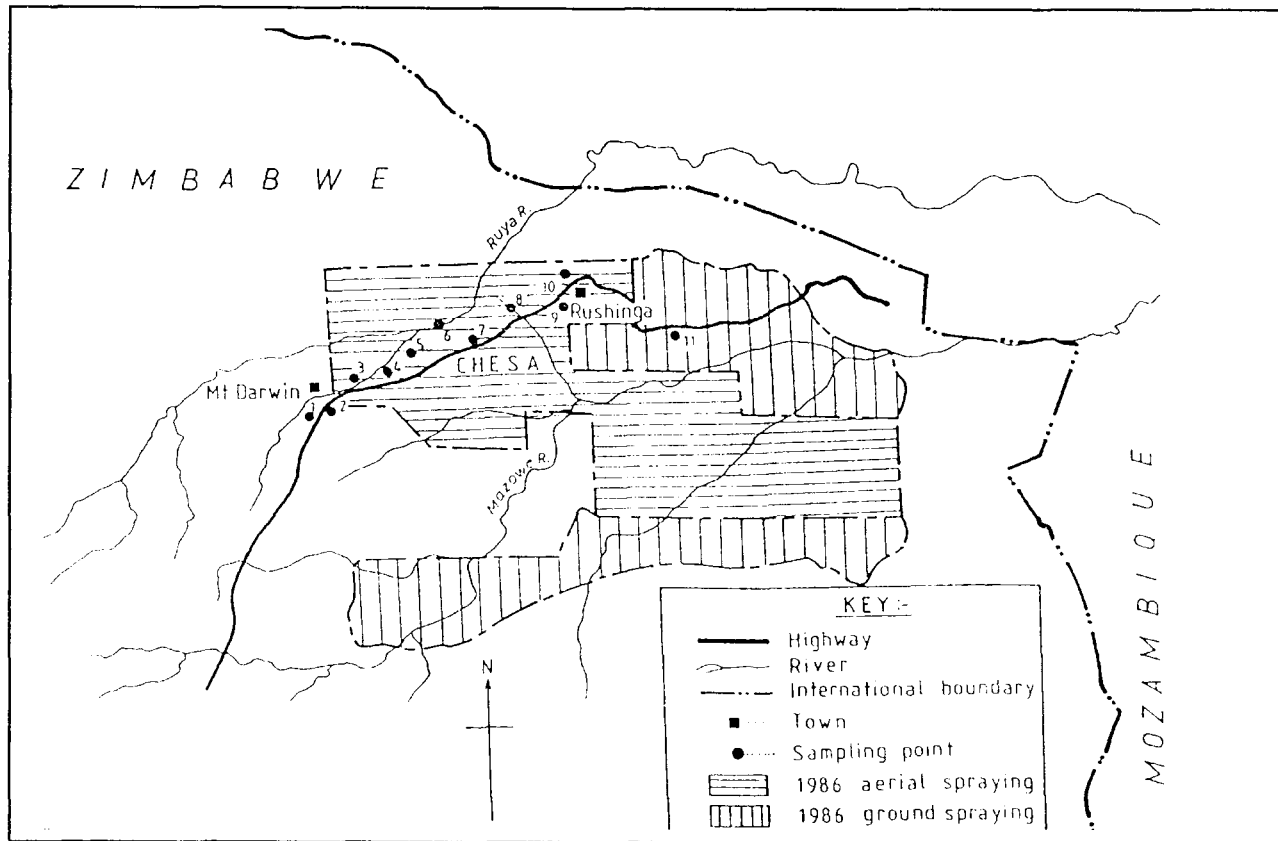
## Methods and Materials

### *Sample collection*

Figure 1 shows the sampling sites from which different samples were collected along the accessible Mount Darwin-Rushinga highway. The area covered lies in the Chesa Communal land (Figure 1). The sites comprised of homesteads, rivers, dams and bushes.

A total of 84 samples were taken twice during 1999 in March and August. Soil samples were taken at random from the bush and fields along the highway. These soil samples were cored at 60 cm depth into clean polythene plastic bags, tightly sealed and stored in a deep freezer (-18° C) prior to analysis.

Housedust was taken from homesteads by sweeping the interior walls and floor of the grass thatched huts. The housedust was put in polythene plastic bags and stored in a deep freezer prior to analysis. The homesteads were situated at different



**Figure 1: Map of Mount Darwin area showing areas sprayed with DDT in 1986 as well as sampling points from which samples were collected.**

locations from Mount Darwin business centre. The first homestead was at 7.2 km from the business centre, the second at 57 km away, the third at 65 km away and the fourth at 71 km from the business centre. Cowdung was taken from the banks of Mupfure river, banks of Tondwe river which was located near a dip tank, the banks of Nyakasikana dam as well as from a kraal close to a homestead, located at 65 km from Mount Darwin business centre. The samples were put in polythene plastic bags and stored in a deep freezer ( $-18^{\circ}\text{C}$ ) prior to analysis. Sediments and riverine silt were collected from accessible dams, rivers and streams. Sediments were taken from edges of rivers and dams, at 20 cm depth, into polythene plastic bags and stored in a deep freezer ( $-18^{\circ}\text{C}$ ) prior to analysis.

Water samples were collected from rivers and dams direct into two litre polythene bottles. The bottles were previously soaked in 15 percent nitric acid, washed with soap solution and rinsed with distilled water. The samples were acidified to pH 2.5 to eliminate any biological activity.

Tree bark and foliage were stripped off fairly large trees at random in the bush. The randomisation was done by taking samples from different types of trees at different locations along the highway, with the sampled tree being the dominant tree at that particular location. Grass was cut underneath the trees. All samples were put in clean polythene plastic bags and stored in a deep freezer ( $-18^{\circ}\text{C}$ ) prior to analysis. Fish were bought from local fisherman at identified dams and rivers, and put in clean polythene plastic bags and stored in a deep freezer ( $-18^{\circ}\text{C}$ ) prior to analysis.

### *Extraction and Clean Up*

#### *Extraction of river sediment, soil, house dust and cowdung*

Samples were allowed to thaw and then ground using mortar and pestle and sieved through a 0.25 cm sieve. A mass (equivalent to one g dry weight) was weighed into a 10 mL beaker. Acetic acid (0.5 ml) was added and the mixture stirred. Nonane (0.5 ml) was added to the slurry and the mixture sonicated for 30 minutes. The mixture was allowed to stand and pesticide grade silica gel (Merck, five) was added and then stirred. Extraction was carried out with 150 ml hexane: benzene (2:1) mixture in a soxhlet extractor and concentrated to one ml using a Kuderna Danish apparatus according to the procedure described by Japenga *et al.* (1987). Cleanup was carried out by passing the extract through a chromatographic column containing florisil and eluted with 20 ml hexane (Fraction 1) followed by 20 ml methanol and hexane (0.5:9.5) — ( Fraction 2). These fractions were separately concentrated to one ml using a Kuderna — Danish concentrator and stored in glass vials at  $-4^{\circ}\text{C}$  until analysis by GC-ECD.

#### *Extraction of grass, tree bark and foliage*

Samples were crushed using a Phillips grinder. The sample (one g) was accurately weighed into a Phillips homogeniser containing acetonitrile (50 ml) and homogenised

to a slurry. The slurry was centrifuged (~ 5 000 rpm) and the supernatant combined into a Kuderna-Danish concentration tube. The combined extract was concentrated to one ml on a water bath and passed through a chromatographic column containing florisil. Elution was done with 15 ml hexane (Fraction 1) followed by 15 ml acetonitrile and hexane (0,5:9,5) — (Fraction 2). The fractions were separately concentrated in a Kuderna- Danish flask to one ml and stored in glass vials at 4° C until analysis by GC-ECD.

#### *Extraction of fish*

Fish samples were allowed to thaw and crushed into fine particles using mortar and pestle. A one g sample was weighed into a 10 ml beaker and anhydrous sodium sulphate (10g) was added. The mixture was mixed thoroughly and acetonitrile (15ml) was added and the mixture sonicated for 10 minutes. The mixture was later centrifuged and the supernatant kept. The residue was further extracted with acetonitrile (five ml) and the supernatants combined. The extract was concentrated to one ml on a Kuderna-Danish and then passed through a chromatographic column containing florisil and eluted with 20 ml hexane (Fraction 1) followed by 20 ml acetonitrile and hexane (0,5:9,5) — (Fraction 2). The two fractions were separately concentrated as explained above and analysed by GC-ECD.

#### *Extraction of water samples*

Two hundred ml of water samples were placed in separating funnels and extracted three times with hexane/ acetone (2:1 volume — 100 mls). The organic layers were washed with two percent sodium sulphate and concentrated on a rotary evaporator. The samples were cleaned up with florisil as above. The extracts were eluted with 20 ml hexane (Fraction 1) followed by 20 ml acetone and hexane (0,5:9,5) — (Fraction 2). The fractions were later concentrated to one ml using the Kuderna-Danish apparatus as explained above and kept at 4° C prior to GC-ECD analysis.

#### *Gas chromatography*

The combined extracts were analysed separately using a Varian Model 3300 gas chromatograph (Varian AB, Solna, Sweden) fitted with a microprocessor, a split/ splitless capillary injector, a Varian model 4400 integrator and a Ni<sup>63</sup>electron capture detector (ECD).

Separations were done using a DB 1701 (30m x 0.25 mm) refined silica capillary column for pesticides and chlorinated aromatics (J and W, Scientific, CA, USA). Ultrapure nitrogen carrier gas was used at a flow rate of five ml/ min (make up nitrogen at 25 ml/ min). The injector was maintained at 200° C and the ECD detector maintained at 300° C. Other GC conditions were as follows:

Initial column temperature	150° C hold for 5 min.
Final column temperature	230° C hold for 15 min.
Temperature programme rate	4° C / min.

Injector temperature	200° C
Detector temperature	300° C
ECD attenuation	8 on autozero
ECD range	10 mV
Initial relay	-1 to +1 after 0.5 min.

Validation of the results was done by spiking samples with a standard solution containing a mixture of 4,4 DDT; 4,4 DDE and 4,4 DDD. The fortified samples were left to stand in the dark for 24 hours to allow for equilibration before extraction, cleanup, concentration and analysis as described above. The unspiked samples were used as blank. The percent recovered was calculated by deducting the unspiked level from the spiked level and dividing by the spike level.

## Results

Tables 1 to 3 show the results obtained for the various samples analysed. The total DDT values (tDDT) are the sum of 4,4 DDE, 4,4 DDD and 4,4 DDT results. Figure 2 shows typical chromatograms for 4,4 DDE, 4,4 DDD and 4,4 DDT in a standard solution (a) as well as in an extracted sediment sample (b).

Recoveries greater than 70 percent were obtained for most samples (Table 4) with the exception of leaves, grass and tree bark for which average recoveries of 56 percent were obtained. The limit of detection using the ECD detector was 0.16 ng/g for 4,4 DDE, 1.0 ng/g for 4,4 DDD and 4,4 DDT. (Tables 1 to 4 and Figure 2 are at the end of the article).

## Discussion

Tables 1 to 3 show the results of the various samples analysed in the Mount Darwin-Rushing area, expressed as total DDT. Sediments showed a widespread contamination though at low levels except at Mupfure river (Point 2) and Nyakasikana (Point 1). The sampling point at Mupfure river (1, see Figure 1) was at the beginning of the river and hence DDT levels tend to be high at this point and decrease along the course of the river. Nyakasikana sampling point (6, see Figure 1) was after the confluence of two rivers, and hence the high level of DDT found was possibly due to the deposition of DDT from the two rivers. This widespread contamination is reflective of the fact that sediments act as a residue sink as noted by Zaranyika et al. (1994). The high DDE/tDDT ratios for most sediments (> 30 percent) implies a significant chemical conversion of the applied DDT to its metabolites (Zaranyika and Mugari, 1997). Since chemically, DDT degrades extremely slowly, this suggests that the DDT was applied a long time ago. There was a general decline in the amount of DDT found in sediments sampled in August



as opposed to March. This was probably as a result of reduced river flow during the dry season (April to September). Water samples had generally low DDT levels with the highest being Mupfure river with 6.26 ng/g and this correlated with high residue levels of DDT in sediments at the same point in that river.

All the fish samples analysed had DDT, DDD and DDE, though in some cases DDE alone was found. The percent fat content of the fish analysed ranged from 9.94 percent at Nyamahokobo to 12.74 percent at Nyakasikana. Fish forms a significant protein for most people in Zimbabwe. The presence of DDT and its metabolites in fish would lead to pollution in humans and other fish eating birds and animals. Previous studies have revealed the presence of DDT and its metabolites in fish eating birds and animals (Phelps and Billings, 1972; Phelps *et al.*, 1974; Greichus *et al.*, 1978; Phelps *et al.*, 1989). Chikuni *et al.* (1997) found DDT, DDE and PCB's in human milk of mothers. This was attributed to the mothers' diet, which was comprised mainly of fish. Further work is required to assess the extent of human and animal pollution due to the presence of DDT in fish.

House dust samples analysed were contaminated with levels ranging from 2.10 ng/g to 276.5 ng/g. This contamination has been attributed to indoor spraying for malaria control and use of cow dung as a floor polish. Levels of tDDT ranging from not detectable to 6.83 ng/g were detected in cow dung samples (no statistical difference was noted in DDT levels for March and August samples). Cow dung is used as a floor polish in most huts in Africa, hence it can be a source of pollution in houses using cow dung as polish. The presence of DDT in housedust would lead to deposition of DDT in the lungs of humans who breath the dust, hence close monitoring of the situation is required.

Leaves, tree bark and grass had generally low DDT levels. Tree leaves had tDDT values ranging from not detectable to 3.70 ng/g. Tree bark on the other hand had values from not detectable to 1.79 ng/g with the exception of muunze (*Brachystegia glauceslens*) bark with 178.8 ng/g. Muunze and mopane were sampled from the same area. Although DDT was detected in soil and grass within the vicinity of both trees, no DDT was detected in mopane bark and high DDT levels were detected in muunze bark. It is also noted that the DDE/ tDDT ratio is very small (0.004) for the muunze bark suggesting little chemical conversion of the applied DDT. The fact that DDT was found in muunze bark and not mopane, may suggest that muunze bark accumulates DDT. Further work is required to confirm this. Grass had tDDT levels ranging from not detectable to 2.26 ng/g. The presence of DDT in grass and tree leaves would lead to contamination of domestic and herbivorous wild animals. There is need therefore to monitor meat and milk for DDT contamination. Soil samples had varied tDDT levels ranging from 0.36 ng/g to 179.0 ng/g.

Comparison of tDDT levels for samples collected from aerial sprayed and ground sprayed areas, as well as samples collected outside the sprayed areas (Figure 1) showed no discernible distribution patter. This is attributed to dispersal of DDT from the sprayed areas through wind, evaporation and rainfall run-off.

## Conclusion

From the foregoing discussion we conclude that the Mour<sup>t</sup> Darwin-Rushinga terrestrial environment still shows widespread trace contamination from DDT residues in spite of DDT having been officially sprayed in the area about 13 years ago, to combat malaria and tsetse fly.

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**Table 1: Results of river water, sediments and fish samples analysed for DDT in Mount Darwin area.**

<b>Sediments</b>					<b>Water Fish (wet mass)</b>					
Sample	Sam. pt*	Total DDT ng/g	DDE ng/g	DDE/tDDT	Total DDT ng/g	DDE ng/g	DDE/tDDT	Total DDT ng/g	DDE ng/g	DDE/tDDT
Mupfure: 1	1									
March		14.91± 0.46	2.45± 0.72	0.16	ND	ND	ND			
August		13.27± 0.07	3.68±0.07	0.27	ND	ND	ND			
Mupfure: 2	1									
March		506.2±14.3	210.48±20.4	0.42	6.26±0.08	1.03±0.10	0.16			
August		492.9±12.3	206.0±10.4	0.42	4.38±0.20	1.64± 0.17	0.37			
Nyakasikana: 1	6									
March		450.3±4.30	157.6±0.08	0.35	ND	ND	ND	0.74±0.19	0.74±0.19	1.00
August		0.34± 0.02	0.34±0.02	1.00	2.78±0.09	1.72± 0.12	0.62	15.56±0.06	14.02±0.03	0.90
Nyakasikana: 2	6									
March		5.16± 0.53	1.25±0.03	0.24	ND	ND	ND			
August		2.26±0.16	0.72±0.18	0.21	ND	ND	ND			
Nyakasikana: 3	6									
March		0.63 ! 0.06	0.30±0.04	0.50	6.10±0.15	2.02±0.17	0.30			
August		ND	ND	ND	ND	ND	ND			
Nyamahokobo: 1	8									
March		0.14±0.05	ND		2.06± 0.17	1.07±0.24	0.52	1.06± 0.46	1.06±0.46	1.00
August		2.02±0.09!	0.49±0.27	0.24	0.94±0.05	0.94±0.05	1.00	2.54± 0.12	0.96±0.17	0.38
Nyamahokobo: 2	8									
March		1.88±0.10	1.21±0.13	0.64	0.31 ! 0.06	0.31 ! 0.06	1.00			
August		1.91±0.25	0.39±0.09	0.20	2.06 ! 0.16	1.07 ! 0.09	0.52			
Nyamahokobo: 3	8									
March		0.74±0.25	0.74±0.25	1.00	ND	ND	ND			
August		61.45±0.11	0.66±0.16	0.01	3.52±0.58	0.54± 0.13	0.15			
Tondwe	3									
March					ND	ND	ND			
August		2.18±0.07	0.50±0.12	0.23	ND	ND	ND			
<b>Mean</b>		<b>91.55±0.19</b>	<b>34.52±22.91</b>	<b>0.35</b>	<b>1.58!0.69</b>	<b>0.57±0.41</b>	<b>0.25</b>	<b>4.98±0.52</b>	<b>4.20!0.53</b>	<b>0.82</b>

ND= not detected = sample not collected Sam pt\* = sampling point, see Figure 1. tDDT = total DDT.

**Table 2: Total DDT, DDE and DDE/tDDT levels in housedust and cowdung samples.**

Sample	Samp pt *	Housedust			Cowdung		
		Total DDT ng/g	DDE ng/g	DDE/ tDDT	Total DDT ng/g	DDE ng/g	DDE/ tDDT
7.2 Homestead	2						
March		276.5±5.0	51.2±1.20	0.19			
August		119.8±2.0	51.23±0.12	0.43			
57 Homestead	9						
March		31.71±0.73	1.45±0.56	0.05			
August		5.72±0.16	0.99±0.23	0.17			
65 Homestead	10						
March		13.1±0.20	12.31±0.52	0.94	2.43±0.06	0.57±0.08	0.56
August		4.20±0.03	0.98±0.05	0.23	2.36±0.07	0.79±0.09	0.33
71 Homestead	11						
March							
August		2.10±0.05	0.48±0.03	0.22			
Mupfure River	1						
March					6.83±1.02	6.83±1.02	1.00
August					3.38±0.16	0.81±0.21	0.24
Tondwe River	3						
March					ND	ND	ND
August					5.53±0.34	1.32±0.43	0.24
Nyakasikana River	6						
March					ND	ND	ND
August					2.22±0.15	0.54±0.18	0.24
<b>Mean</b>		<b>64.73±5.44</b>	<b>16.95±1.45</b>	<b>0.32</b>	<b>2.84±1.10</b>	<b>1.36±1.15</b>	<b>0.33</b>

ND = not detected.



= sample not collected.

Samp pt \* = sampling point, see Figure 1.

tDDT = total DDT.

**Table 3: Total DDT, DDE and DDE/ tDDT levels in tree leaves, tree bark as well as soil and grass from the vicinity of the tree sampled.**

Sample	Date	Samp pt *	Tree leaves			Tree bark			Soil			Grass		
			Total DDT ng/g	DDE ng/g	DDE/ tDDT	Total DDT ng/g	DDE ng/g	DDE/ tDDT	Total DDT ng/g	DDE ng/g	DDE/ tDDT	Total DDT ng/g	DDE ng/g	DDE/ tDDT
Mopane 19 <i>Colaphospermum mopane</i>	March August	4	ND	ND	ND	ND	ND	ND	0.70±0.08	0.10±0.05	0.14	0.15±0.04	0.15±0.04	1.00
Mupfuti 26 <i>Brachystegia boehmii</i>	March August	5	ND	ND	ND	ND	ND	ND	179.0±6.6	0.78±0.10	0.004	0.34±0.16	0.34±0.16	1.00
Munzvuru 26 <i>Vangueria infausta</i>	March August	5	ND	ND	ND	ND	ND	ND	10.2±0.70	10.2±0.70	1.00	ND	ND	ND
Mutohwe 34 <i>Azanza garckeana</i>	March August	7			ND	ND	ND	ND	ND	ND	ND	ND	ND	
Muunze 19 <i>Brachystegia glaucescens</i>	March August	4	3.70±0.16	0.95±0.17	0.26	178.8±3.10	0.73±0.18	0.004	1.86±0.17	0.34±0.05	0.18	0.71±0.08	ND	
Musasa 26 <i>Brachystegia Spiciformis</i>	March August	5	2.26±0.15	0.56±0.01	0.25	1.79±0.09	0.11±0.05	0.06	0.36±0.11	0.36±0.11	1.00	ND	ND	ND
Mutsuma 26 <i>Diospyros mespiliformis</i>	March August	5	ND	ND	ND	0.49±0.13	0.49±0.13	1.00	2.06±0.21	0.35±0.01	0.17	2.26±0.40	0.56±0.13	0.25
Arable 34 Macheka farm	March August	7							15.17±0.15	3.06±0.35	0.20	1.90±0.33	0.38±0.03	0.20
Dead wood	March August					ND	ND	ND						
Mean			0.99±0.22	0.25±0.17	0.09	22.64±3.10	0.17±0.23	0.13	26.17±6.65	1.90±0.80	0.34	0.67±0.55	0.18±0.21	0.31

ND = not detected.

□ = sample not collected.

Samp pt \* = sampling point, see Figure 1.

tDDT = total DDT.

**Table 4: Extraction recoveries percent of 4,4 DDE; 4,4 DDD and 4,4 DDT added to various samples at various fortification levels.**

Sample	4,4 DDE			4,4 DDD			4,4 DDT		
	Spike level ng/g	Rec. n =2 ng/g	% Rec.	Spike level ng/g	Rec. n =2 ng/g	% Rec.	Spike level ng/g	Rec. n =2 ng/g	% Rec.
Housedust:									
57 Km	5.6	4.32	<b>77.1</b>	1.4	1.03	<b>73.6</b>	2.85	2.40	<b>84.2</b>
Sediment:									
Mupfure	1.1	1.08	<b>98.2</b>	2.35	2.00	<b>85.1</b>	1.55	1.58	<b>102</b>
Water:									
Nyamahokobo	1.55	1.20	<b>77.4</b>	2.60	1.38	<b>72.3</b>	0.75	0.51	<b>68.0</b>
Fish:									
Nyamahokobo	2.4	1.62	<b>67.5</b>	2.55	1.88	<b>73.4</b>	2.85	1.78	<b>62.5</b>
Soil:									
34 Km	2.1	2.09	<b>99.5</b>	2.50	2.13	<b>84.0</b>	4.50	4.56	<b>101</b>
Cowdung:									
65 km	3.5	2.60	<b>74.4</b>	2.50	1.73	<b>69.2</b>	1.10	0.61	<b>55.4</b>
Grass:									
34 Km	2.0	1.51	<b>75.5</b>	2.30	1.95	<b>84.7</b>	1.50	1.20	<b>80.0</b>
Leaves:									
19 Km	2.65	2.35	<b>88.7</b>	3.40	2.11	<b>62.0</b>	4.00	2.24	<b>56.0</b>
Treebark	1.35	0.34	<b>62.2</b>	1.45	1.15	<b>79.3</b>	2.00	1.24	<b>62.0</b>

Rec = Recovery.



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