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Organochlorine Insecticide Residues in Some Agricultural Soils on the North Coast Region of New South Wales

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Organochlorine (OC) insecticides effectively control many agricultural insect pests. Surveys of agricultural soils in temperate regions have shown that OC residues have accumulated to unacceptably high levels (e.g., Carey 1979; Edwards and Adams 1970; Frank et al. 1974; Harris et al. 1977; Saha and Sumner 1971; Stevens et al. 1970) and their use has been severely curtailed. Elsewhere OC insecticide use is still common - in New South Wales (NSW), they have been used for agricultural purposes until only recently. In contrast to temperate regions, little information is available on the extent to which OC residues have accumulated in subtropical soils as a result of normal insecticide control practices. We wish to report results of a study done to determine levels to which OC residues have accumulated in pasture, sugar cane and banana plantation soils in the NSW North Coast Region.

MATERIALS AND METHODS

Responsibility for residue analyses was shared between the Board of Tick Control Laboratory, Lismore (pasture soils) and the Biological and Chemical Research Institute, Rydalmere (sugar cane and banana plantation soils).

Five dairy farms in the Kempsey district were sampled intensively in 1983-84, with soil samples being collected from individual paddocks. Thirty, 2.5 x 7.5 cm cores were taken in a zig-zag fashion diagonally across a paddock and were composited. To determine vertical distribution of the residues, a 2.5 x 45 cm piece of conduit was tapped into the soil, removed and divided into 4, 7.5 cm segments and 1, 15 cm segment. Soil from equivalent segments from 3 cores/paddock was removed and composited. After evaporating excess moisture when necessary, the soil sample was sieved and was thoroughly mixed. A 10-50 g subsample was dried for 24 h at 105°C for moisture determination.

To extract insecticide residues from pasture soils, a 10 g subsample was soaked with acetone in a 250 ml glass stoppered flask for 16-24 h, with 3 vigorous 30 second shakings. A 10 ml aliquot

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of the extract was removed, the acetone was evaporated and replaced with petroleum ether, and the extract was dried with anhydrous sodium sulphate. Samples were analysed without further treatment using a Varian model 2100 gas chromatograph (GC) fitted with 4, 180 cm x 2 mm ID glass columns packed with 2% OV 17; 4% OV 210; and 3.5% OV 210 + 1.5% SE 30 (2 columns) all on Chrom W-DMCS-AW at 205°C. Detection was by ^{3}H electron capture detector at 260°C, with injector at 220°C. This combination of columns allowed analysis of all insecticides under investigation with quantitation by peak ht compared with external composite standards.

Sugar cane soils were sampled in the Tweed (32 fields), Clarence (25) and Richmond (12) River districts in 1985 taking 50, 2.5 x 15 cm cores as described for pasture soils. To determine vertical distribution of the residues, a 5 cm diam auger was used to collect consecutive 0-15, 15-30, 30-45 and 45-60 cm segments through the soil profile. Soil from equivalent segments from 3 cores/field was composited.

Banana plantations were sampled in the Tweed River (9 plantations), Macksville (5) and Coffs Harbour (5) districts in 1985. Cores, 2.5 x 15 cm were taken within 30 cm of the base of 10 plants in each of rows 2 and 3 at the top of the plantation and were composited. Ten cores also were taken between each of rows 2 and 3 and 3 and 4 and were composited. Sampling was done in a similar fashion at the bottom of the plantation. An additional composite sample comprising 20 cores was collected 5-10 metres below the plantation.

Sugar cane and banana plantation soils were sieved and mixed, and moisture content was determined as described earlier. A 25 g subsample was extracted by the method of Miles and Harris (1971) and an aliquot of the concentrated extract was cleaned on deactivated alumina (de Fauber Maunder et al. 1964). Samples were analysed using a Varian model 3700 GC equipped with a ^{63}Ni electron capture detector and a 183 cm x 3 mm ID stainless steel column packed with a 1:1 mixture of 20% OV 17 and 3% DC 200 on 80-100 mesh Gas Chrom Q. The injector block was lined with a replaceable glass liner. Operating conditions were: column 240°C, injector 190°C, detector 360°C, nitrogen flow 50 ml min^{-1}. A more polar 2.1% PEGS column (122 cm x 3 mm at 140°C) was used for confirmation.

Soil from all locations was examined for heptachlor (H), heptachlor epoxide (HE), aldrin (A), dieldrin (D), α, β, γ-BHC, p,p'-DDT, p,p'-DDE and p,p'-TDE. Results are expressed in ppm (µg dry soil) with levels <0.01 ppm being indicated by T (trace); limit of sensitivity was 0.005 ppm. ND = None detected; NS = Not sampled.

RESULTS AND DISCUSSION

NSW dairy farms are typically divided into paddocks, each comprising a few hectares. Maize or other field crops are often rotated with pasture. Pesticide use histories indicated that H was the

preferred insecticide used to control African black beetle, Heteronychus arator and the white fringed weevil, Graphognathus leucoloma. Detailed results are summarised in Table 1 for only 1 farm - 9 of 16 paddocks sampled contained H and/or HE residues; 7 paddocks contained D residues. Data for the remaining farms are

Table 1. OC insecticide residues (ppm) in pasture soils, Kempsey district

Farm No	Paddock No(s)	H	HE	D
1	1	ND	ND	ND
	2	0.02	0.14	ND
	3	ND	0.28	0.14
	4	ND	ND	0.16
	5	ND	ND	0.25
	6	ND	ND	0.16
	7	ND	ND	ND
	8	ND	0.22	0.02
	9	0.23	ND	ND
	10	ND	ND	ND
	11	0.15	0.26	0.07
	12	ND	0.31	ND
	13	0.03	0.48	ND
	14	ND	0.16	ND
	15	0.09	0.21	0.05
	16	ND	ND	ND
	1-16	0.03(ND-0.23)	0.13(ND-0.48)	0.05(ND-0.25)
2	1-28	0.03(ND-0.31)	0.07(ND-0.30)	0.01(ND-0.24)
3	1-12	T (ND-0.06)	0.03(ND-0.10)	ND
4	1-2	0.04(ND-0.08)	0.05(0.01-0.09)	ND
5	1-5	0.38(ND-1.8)	0.35(0.03-1.4)	ND

summarised in terms of number of paddocks sampled, average, and range of residue levels detected. With exception of farm 5, where residue levels were substantially higher, H residues in the soil generally averaged <0.1 ppm; HE residues were usually slightly higher. Total H + HE residues were, with exception of farm 3, usually close to, or >0.1 ppm. D residues were present in only 2 of the 5 farm soils. No other OC insecticide residues were identified in the pasture soils.

Vertical distribution of the insecticide residues was determined in a number of paddocks on different farms. Results were very similar; data from 2 of the paddocks sampled are summarised in Table 2. H and HE residues were generally highest in the top 22.5 cm of soil. D residues were more uniformly distributed through the entire 45 cm layer sampled.

Table 2. Vertical distribution of OC insecticide residues (ppm) in pasture (Kempsey district) and sugar cane (Richmond River district) soils.

Farm (Paddock) No	Depth (cm)	H	HE	D	α-BHC	β-BHC	γ-BHC
			Pasture				
1(8)	0.0- 7.5	0.06	0.14	0.02	ND	ND	ND
	7.5-15.0	0.04	0.12	0.03	ND	ND	ND
	15.0-22.5	0.05	0.12	0.03	ND	ND	ND
	22.5-30.0	0.02	0.09	0.03	ND	ND	ND
	30.0-45.0	ND	ND	0.06	ND	ND	ND
2(12)	0.0- 7.5	0.04	0.10	0.03	ND	ND	ND
	7.5-15.0	0.04	0.10	0.02	ND	ND	ND
	15.0-22.5	ND	0.03	0.02	ND	ND	ND
	22.5-30.0	ND	ND	0.01	ND	ND	ND
	30.0-45.0	ND	ND	0.01	ND	ND	ND
			Sugar Cane				
5	0-15	ND	ND	0.07	0.01	0.03	0.01
	15-30	ND	ND	0.24	0.02	0.92	ND
	30-45	ND	ND	0.06	T	0.03	ND
	45-60	ND	ND	0.04	T	0.02	ND
11	0-15	ND	ND	0.25	ND	ND	ND
	15-30	ND	ND	0.16	ND	ND	ND
	30-45	ND	ND	0.11	ND	ND	ND
	45-60	ND	ND	0.08	ND	ND	ND

Pesticide treatment histories from sugar cane growers indicated that BHC, D or A, had been applied 1 to >3 times over the years to control Lepidiota, Dermolepida and Antitrogus spp. grubs and the soldier fly, Inopus rubriceps. In the Tweed River district (Table 3), BHC residues were present in ca. one third of the fields sampled. D residues were detected in soils from 29 of 32 (91%) farms sampled; average D level in soil was 0.13 ppm (range ND-0.94 ppm). DDT residues of 0.43, 0.18, and 0.20 ppm were detected at sites 30, 31, and 32 respectively. Those fields were currently being used for purposes other than sugar cane production. In the Clarence River district (Table 4), no residues of BHC or DDT were detected in any of the farm soils. Although pesticide use histories suggested that A (1-3 applications) was used in a preference to D, conversion to the latter was almost complete, with A residues being present on only 5 farms; 0.01, 0.02, 0.01, 0.01, and <0.01 ppm on farms 2, 12, 14, 24, and 25 respectively. D residues were detected in all 25 fields sampled; average D residue in the soil was 0.11 ppm (range 0.05-0.29 ppm). In the Richmond River district (Table 4), no DDT, H, or A residues were detected in any of the farm soils;

Table 3. OC insecticide residues (ppm) in sugar cane soils, Tweed River district.

Farm No	α-BHC	β-BHC	γ-BHC	A	D
1	T	T	T	ND	0.06
2	ND	ND	ND	ND	0.28
3	ND	ND	ND	T	0.02
4	T	0.01	T	T	0.03
5	T	T	T	T	T
6	0.08	0.45	0.14	ND	0.14
7	T	ND	T	ND	0.94
8	0.09	0.38	0.22	T	0.04
9	0.01	0.02	0.02	ND	0.05
10	T	0.01	T	ND	0.15
11	0.06	0.07	0.08	T	0.65
12	T	T	T	T	0.24
13	ND	ND	ND	ND	0.07
14	ND	ND	ND	ND	0.08
15	ND	ND	ND	ND	0.01
16	ND	ND	ND	ND	0.10
17	ND	ND	ND	ND	0.11
18	ND	ND	ND	ND	0.20
19	ND	ND	ND	ND	0.09
20	ND	ND	ND	0.01	0.03
21	ND	ND	ND	ND	0.03
22	ND	ND	ND	0.01	0.09
23	ND	ND	ND	0.01	0.18
24	ND	ND	ND	ND	0.07
25	ND	ND	ND	ND	ND
26	ND	ND	ND	ND	ND
27	ND	ND	ND	ND	0.22
28	ND	ND	ND	ND	ND
29	ND	ND	ND	ND	0.04
30	ND	ND	ND	ND	0.07
31	ND	ND	ND	ND	0.04
32	ND	ND	ND	ND	0.04

α-, β- and γ-BHC residues of 0.06, 0.55, and 0.18; and 0.01, 0.03 and 0.01 ppm, respectively, were detected on farms 4 and 5. D residues were present at all locations; average D residue in the soil was 0.13 ppm (range 0.02-0.25 ppm).

Results of vertical distribution studies on 2 caneland farms, which are typical of all 12 Richmond River district farms sampled, are summarised in Table 2. When present, e.g., farm 5, BHC residues were distributed through the soil to at least 60 cm. In general, residues were highest in the upper 30 cm soil. D residues, as exemplified by data obtained on farms 5 and 11, showed a similar

Table 4. D residues (ppm) in sugar cane soils, Clarence River and Richmond River districts

Farm No	D	Farm No	D	Farm No	D
		Clarence River district			
1	0.05	9	0.06	17	0.09
2	0.29	10	0.08	18	0.08
3	0.08	11	0.09	19	0.09
4	0.08	12	0.21	20	0.13
5	0.06	13	0.05	21	0.05
6	0.12	14	0.26	22	0.08
7	0.09	15	0.09	23	0.14
8	0.09	16	0.08	24	0.23
				25	0.17
		Richmond River district			
1	0.09	5	0.07	9	0.15
2	0.12	6	0.08	10	0.13
3	0.02	7	0.15	11	0.25
4	0.06	8	0.22	12	0.19

distribution pattern.

In banana plantations, D had been used to suppress banana weevil borer, Cosmopolites sordidus, with treatments being applied to, and around the base of the plant. In the Tweed River district, where a deliberate effort was made to select plantations which reportedly had received no D teatments since the early 1970s, D residues were, with one exception (Farm 2 - top), generally <0.1 ppm (Table 5). Residues in the Macksville and Coffs Harbour soils, where D reportedly had been used as recently as 1984, were substantially higher (Table 5). In general, D levels in-row were higher than those found inter-row reflecting the application method. Residues in soil samples taken 10 m below the plantations were substantially lower than levels detected in the bottom plantation row. DDT residues of 0.04 and 0.02 ppm were detected in bottom and below sampling sites on farm 6.

As the data indicate, cyclodiene insecticides were commonly detected in areas of reported use, with residue levels being similar to or less than those found in temperate regions. As reported by many authors (e.g., Cliath and Spencer 1971; Harris and Sans 1970; Lichtenstein et al. 1971; Voerman and Besemer 1975), vertical mobility of the insecticides in soil was limited, with residues tending to be concentrated in the plough layer. D appeared to be slightly more mobile than HE. Data obtained by Gilbert and Lewis (1982) suggest that H and HE or D residues >0.1 ppm in mineral soils in NSW could result in residues in milk above the maximum residue

Table 5. D residues (ppm) in soil collected in, or adjacent to, banana plantations.

Farm No	Top	Top inter-row	Bottom	Bottom inter-row	Below (10m)
		Tweed River district			
1	0.03	NS	0.02	NS	NS
2	0.24	NS	ND	NS	NS
3	0.02	NS	0.07	NS	NS
4	0.06	NS	0.03	NS	NS
5	0.03	NS	0.04	NS	0.04
6	0.04	NS	0.04	NS	0.03
7	0.05	NS	0.05	NS	0.05
8	ND	T	ND	T	1.5
9	0.13	0.04	0.04	0.02	0.03
		Macksville district			
1	0.24	0.10	0.33	0.27	NS
2	0.38	0.98	0.88	0.11	NS
3	0.42	0.53	0.42	0.56	0.09
4	0.44	0.10	0.24	0.15	0.12
5	3.4	0.63	4.1	2.3	0.08
		Coffs Harbour district			
1	0.59	0.25	0.26	0.26	NS
2	0.33	0.36	0.08	0.08	NS
3	0.58	0.48	0.36	0.52	0.03
4	14.	1.2	3.1	0.90	0.55
5	0.28	0.06	0.16	0.04	0.03

limit (MRL) (0.15 mg/kg in butterfat). Average H and HE levels in soils from the dairy farms tested were, with 1 exception, close to 0.1 ppm with levels in some paddocks being substantially higher. Milk samples from these farms have all, at one time or another, exceeded the MRL. D residues in sugar cane soils also averaged about 0.1 ppm with individual fields being substantially higher, suggesting that conversion of caneland soils to pasture should be approached with caution.

Total OC residues in pasture, sugar cane, and banana plantation soils were generally much lower than levels reported in many of the surveys of pasture, field crop, and orchard soils referred to earlier. This was due primarily to the fact that, in contrast to other countries, DDT residues were seldom detected. Thus, environmental contamination to the extent encountered in other countries is unlikely in the North Coast region of New South Wales.

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REFERENCES

Carey AE (1979) Monitoring pesticides in agricultural and urban soils of the United States. Pestic Monit J 13:23-27

Cliath MM, Spencer WF (1971) Movement and persistence of dieldrin and lindane in soil as influenced by placement and irrigation. Soil Sci Soc Amer Proc 35:791-795

Edwards CA, Adams RS (1970) Persistent pesticides in the environment. In: Critical reviews in environmental control, Vol 1. Chemical Rubber Co, Cleveland, p 7-67

Frank R, Montgomery K, Braun HE, Berst AH, Loftus K (1974) DDT and dieldrin in watersheds draining the tobacco belt of southern Ontario. Pestic Monit J 8:184-201

de Fauber Maunder MJ, Egan H, Godley EW, Hammond EW, Roburn J, Thomson J (1964) Clean-up of animal fats and dairy products for the analysis of chlorinated pesticide residues. Analyst 89:168-174

Gilbert WS, Lewis CE (1982) Residues in soil, pasture and grazing dairy cattle after the incorporation of dieldrin and heptachlor into soil before sowing. Aust J Exp Agric Anim Husb 22:106-115

Harris CR, Sans WW (1970) Vertical distribution of residues of organochlorine insecticides in soils collected from six farms in southwestern Ontario. 1969 Proc Ent Soc Ont 100:156-164

Harris CR, Chapman RA, Miles JRW (1977) Insecticide residues in soils on fifteen farms in southwestern Ontario, 1964-74. J Environ Sci Hlth. B12:163-177

Lichtenstein EP, Schulz KR, Fuhremann TW (1971) Effects of crop cover versus soil cultivation on the fate and vertical distribution of insecticide residues in soil 7 to 11 years after soil treatment. Pestic Monit J 5:218-222

Miles JRW, Harris CR (1971) Insecticide residues in a stream and a controlled drainage system in agricultural areas of southwestern Ontario 1970. Pestic Monit J 5:289-294

Saha J, Sumner AK (1971) Organochlorine insecticide residues in soil from vegetable farms in Saskatchewan. Pestic Monit J 5:28-31

Stevens LJ, Collier CW, Woodham DW (1970) Monitoring pesticides in soils from areas of regular, limited and no pesticide use. Pestic Monit J 4:145-166

Voerman S, Besemer AFH (1975) Persistence of dieldrin, lindane, and DDT in a light sandy soil and their uptake by grass. Bull Environ Contam Toxicol 13:501-505

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