Bull. Environ. Contam. Toxicol. (1993) 51:178–184

Environmental Contamination and Toxicology

Soil Persistence of DDT, Dieldrin, and Lindane over a Long Period

A. Martijn, H. Bakker, and R. H. Schreuder

Plant Protection Service, P.O. Box 9102, 6700 HC Wageningen, The Netherlands

The great persistence of chlorinated hydrocarbon pesticides has been recognised since soon after their introduction into agricultural practice. Though originally this property was considered as being favourable, because it yielded long protection against all sorts of pests, the detrimental effects soon became apparent. Numerous studies have been undertaken to investigate these effects.

Voerman and Besemer (1970) reported a study in which among others DDT, dieldrin and lindane had been applied to crops and soil over a period as long as 15 years in which figures of residues of the applied chemicals were given. Five years after termination of this study they (Voerman and Besemer 1975) re-examined the soil layers of the treated fields for residues. A considerable fraction of the orginally applied amounts were still present. Since then, the experimental field was kept intact and left relatively undisturbed, while no pesticides were applied. The long well-documented treatment history, the long period elapsed since the last treatment, and the uncertainty about the future use of the field led in 1989 to the decision to have another check on the soil residue situation of this unique experimental field. The residue levels and changes of the vertical distribution after so many years were thought to be of interest. The long-term effects of the presence of DDT, dieldrin and lindane on the soil fauna will be reported elswhere (*Van de Bund, in preparation*).

MATERIALS AND METHODS

Details of the experimental design, the lay-out of the trials and the treatments have been reported earlier (Voerman and Besemer, 1970). Essentially the pesticides DDT, dieldrin and lindane were applied on separate plots of 12 m^2 at 3 different dosage regimes i.e. an

Send reprint requests to A. Martijn at the above address.

annual crop treatment at normal dosages, an annual crop treatment at a double dosage rate and an annual soil treatment, treatments B, C and D respectively. For each pesticide an untreated plot was present. Experiments were started in 1953 and continued until 1968. The field was then left fallow until 1970.
Rye was sown in 1971 and from 1972-1976 the field was covered with perennial rye grass. Sugar beet grew in 1977 and spring barley in 1978. From 1979 to 1986 the site was again covered by perennial rye grass. From 1987 to 1989 the field was used for experiments with yellow nutsedge (*Cyperus esculentus*). No pesticides were applied. At all times care was taken to prevent cross-contamination of the soil from adjacent plots. This was facilitated by the fact that the plots were separated by sunken concrete slabs which emerged 10 cm above soil level. The soil was a light sandy soil with 3% organic matter.

The plots were sampled mid December 1989. Samples were taken to a depth of 60 cm with a half-open-faced auger (2.5 cm diam.). From each plot 6 samples were taken. Each sample was divided into 6 sub-samples, corresponding to layers of 10 cm. The six sub-samples per layer of 10 cm from one plot were combined to one sample per layer, and mixed. Samples were stored at -18°C until analysis.

The soil samples were air dried, mixed thoroughly and sieved over a 1 mm sieve. Portions of 25 g were transferred to 250 ml flasks and extracted by shaking for 12 hours with 115 ml of a acetone-hexane-water (10 + 8 + 5) mixture to which 7.5 g sodium chloride had been added. Extracts were filtered over a Büchner filter with Whatman nr. 5 filter paper. The flasks were washed twice with 5 ml acetone. The acetone washings were also filtered over the Büchner filter. The combined filtrates were then transferred to a 250 ml separating funnel with two 5 ml portions of hexane. After separation the hexane layer was transferred to a thick-wall 250 ml round-bottom flask through a funnel provided with a cotton pad and filled with 20 g sodium sulphate. The extraction was repeated twice with 15 ml hexane. The dried and combined hexane layers were than concentrated to about 2 ml in a rotating evaporator at 40°C. The concentrated extract was transferred quantitatively to a pre-packed Bakerbond-SPE Florisil (art. nr. 7213-07) column placed over a calibrated 10 ml tube. Pesticide residues were eluted with 5 ml hexane/acetone (9 + 1) and concentrated to the required volume with a stream of dry nitrogen.

Capillary gaschromatography was carried out on a Hewlett-Packard 5880 model gas chromatograph equipped

Table 1. Residues (mg/kg) found in soil after application of DDT, dieldrin and lindane

Compound	Plot	Year	Soil layer(cm)					
			0-10	10-20	20-30	30-40	40-50	50-60
Dieldrin	B	1969	1.25	0.23	0.02	0.01	-	-
		1973	0.77	0.71	0.17	0.03	0.02	0.04
		1989	0.43[a]	0.43[a]	0.17	0.03	< 0.01	< 0.01
	C	1969	2.29	0.86	0.02	0.01	-	-
		1973	1.73	1.72	0.33	0.04	0.02	0.02
		1989	1.46[a]	1.46[a]	0.68	0.12	0.02	0.01
	D	1969	7.33	2.50	0.05	0.03	0.02	0.01
		1973	7.3	8.0	1.3	0.25	0.10	0.11
		1989	4.83[a]	4.83[a]	3.11	0.45	0.10	0.04
Lindane	D	1969	1.30	0.23	-	-	-	-
		1973	0.32	0.35	0.10	0.11	-	0.01
		1989	0.01[a]	0.01[a]	0.01	< 0.005	< 0.005	< 0.005
p,p' DDT	B	1969	3.53	0.56	0.01	0.01	-	-
		1973	2.45	2.36	0.36	0.07	0.04	0.01
		1989	1.35[a]	1.35[a]	0.30	0.17	< 0.01	< 0.01
	C	1969	8.94	1.05	0.02	0.01	0.01	-
		1973	5.8	5.6	0.98	0.13	0.08	0.06
		1989	2.5[a]	2.5[a]	0.55	0.05	0.02	0.01
	D	1969	59.6	9.22	0.15	0.10	0.11	0.13
		1973	42.5	37.3	2.7	0.36	1.9	0.19
		1989	22.2[a]	22.2[a]	6.6	1.2	0.10	0.08

[a] Soil layers of 0-10 cm and 10-20 cm were combined.

Table 1 (continued).

Compound	Plot	Year	Soil layer(cm)					
			0-10	10-20	20-30	30-40	40-50	50-60
p,p' DDE	B	1969	0.39	0.07	-	-	-	-
		1973	0.44	0.42	0.10	0.02	0.01	0.01
		1989	0.51[a]	0.51[a]	0.17	0.09	0.01	< 0.01
	C	1969	0.77	0.11	0.01	0.01	-	-
		1973	1.2	1.1	0.21	0.04	0.02	0.02
		1989	1.1[a]	1.1[a]	0.35	0.07	0.03	0.02
	D	1969	3.33	0.51	0.02	0.02	0.02	0.02
		1973	4.5	4.4	0.42	0.06	0.16	0.03
		1989	7.0[a]	7.0[a]	2.2	0.54	0.08	0.08
o,p' DDT	B	1969	0.78	0.07	-	-	-	-
		1973	0.63	0.58	0.08	0.02	-	-
		1989	0.23[a]	0.23[a]	0.07	0.04	-	-
	C	1969	1.77	0.22	-	-	-	-
		1973	1.5	1.4	0.24	0.03	-	0.01
		1989	0.61[a]	0.61[a]	0.14	0.01	< 0.01	< 0.01
	D	1969	12.9	2.02	0.04	0.02	0.02	0.02
		1973	12.2	11.0	0.83	0.10	0.46	0.05
		1989	5.8[a]	5.8[a]	1.8	0.36	0.02	0.02
o,p' DDE	B	1989	0.01[a]	0.01[a]	-	-	-	-
	C	1989	0.01[a]	0.01[a]	-	-	-	-
	D	1989	-	-	-	-	-	-

[a] Soil layers of 0-10 cm and 10-20 cm were combined.

Table 2. Recoveries at different levels (%)

Compound	0.01 mg/kg	0.1 mg/kg	1 mg/kg
p,p' DDT	97	86	83
o,p' DDT	102	85	84
p,p' DDE	96	82	84
o,p' DDE	96	82	86
Dieldrin	96	80	83
Lindane	165	98	85

Table 3. Climatic conditions during the 1973-1989 period

	Av.temp. (°C)	St.dev.*	Av.rainfall (mm)	St.dev.*
Winter	2.6	1.7	-	-
Spring	8.3	0.8	-	-
Summer	16.3	0.9	-	-
Autumn	10.1	0.7	-	-
Total	-	-	718	102

* n = 17

Table 4. Total amounts of DDT, dieldrin and lindane

Compound	Plot	Applied ($g/12m^2$)	Recovered ($g/12m^2$)		
			1969	1973	1989
Dieldrin	B*	16	2.4	2.7	1.6
	C*	33	5.0	6.0	5.8
	D	83	15.5	26.6	20.8 (25)
Lindane	D	30	2.3	1.4	0.05 (0.2)
p,p' DDT	B*	32	6.4	8.2	4.9
	C*	64	15.6	19.7	8.7
	D	305	108	132.5	81.7 (28)
o,p' DDT	B*	11	1.3	2.0	0.9
	C*	22	3.1	4.9	2.1
	D	102	23	38.4	21.6 (21)
p,p' DDE	B*	-	0.7	1.6	2.0
	C*	-	1.4	4.0	4.2
	D	-	6.1	14.9	26.2 (9.6)

* Application to crops.
Between brackets, % of applied (DDE calculated as DDT).

with a Ni-63, electron capture detector, a 25 m x 0.32 mm capillary OV-1701 column, and an automatic injector. The following operating conditions were used: column temperature: 225°C; injection temperature: 250°C; detector temperature: 300°C; carrier gas: nitrogen at 30 kPa; injection volume: 3 µl; split ratio 10 : 1. External standardization was used. Standard solutions of 0.05 and 0.5 mg/l were used. The detector was linear in the 0.2 to 2 mg/l range.

RESULTS AND DISCUSSION

The results are presented in Table 1. The figures are the averages of two determinations and are based on air dried soil. No corrections for recovery were applied. Because the levels of the 0-10 cm and the 10-20 cm layers in the 1973 experiment differed very little, this time the samples from the two layers were combined. The lindane levels of 1973 were already low and were expected to have decreased further. Therefore samples were taken only from the lindane plot that had received the highest dose (treatment D). Only traces of o,p' DDE could be detected in the upper layer of the DDT plot (plot D). Recovery experiments were carried out at 0.01, 0.1 and 1.0 mg/kg levels. Results are given in Table 2. Climatic conditions recorded by the Meteorological Department of the Agricultural University at Wageningen are given in Table 3. In Table 4 the total amounts still present were calculated assuming a soil density of 1.3 (i.e. 1560 kg soil per layer of 10 cm). For comparison the results from 1969 (Voerman and Besemer, 1970) which have been calculated in the same way are added, as are the re-calculated data from the 1975 report. For the calculation of the applied amounts of p,p' DDT and o,p' DDT it was assumed that the technical DDT had contained 75% p,p' DDT and 25% o,p' DDT. In comparing the figures one should keep in mind that the amounts given as applied do not represent the actual amounts that have reached the soil. Only in the D series of experiments have the pesticides been applied directly to the soil. In the other two cases they have been applied to the crops and only a part of the dose will have reached the soil. The levels of 1973 are, contrary to what might be expected, higher than the ones from 1969. This was probably caused by a change of the analytical method. As the method used in 1989 differed also from the ones used before, the results cannot be used to calculate the half lifes of the applied pesticides. A rough estimate however yields half lifes of about 20 years for p,p'DDT and about 15-20 years for o,p' DDT. Dieldrin seems to be more persistent. The half life of lindane may be in the order of 3 to 4 years. Very little movement to lower soil layers has occurred. It is clear from the

figures presented that the top 20 cm of the soil column have been thoroughly mixed by the soil tillage. Even the transport to the 20 to 40 cm layers may have been caused, at least to a considerable extent, by mechanical disturbance rather than by leaching.

Acknowledgments. The authors are indebted to Mr J. Smelt of the Winand Staring Centre of Integrated Land Soil and Water Research for his advice and assistence with the sampling and to Dr L.J.M. Kroon of the Meteorological Department of the Agricultural University Wageningen for supplying the climatological data.

REFERENCES

Voerman, S, Besemer, AFH (1970) Residues of dieldrin, lindane, DDT and parathion in a light sandy soil after application throughout a period of 15 years. J Agr Fd Chem 18:717-719

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

Received August 26, 1992; accepted February 7, 1993.