Persistence and Vertical Distribution of DDT, Lindane, and Aldrin Residues, 10 and 15 Years after a Single Soil Application

E. Paul Lichtenstein,* Thomas W. Fuhremann, and Kenneth R. Schulz

Agricultural loam soils were treated in 1954 with technical DDT (84% *p,p'*-DDT and 15% *o,p'*-DDT), aldrin, or lindane, and analyzed at intervals over a 15-year period. Data are presented relative to the persistence and metabolism of these insecticides in soils under field conditions. The amount of *p,p'*-DDT plus *o,p'*-DDT recovered 15 years after soil treatment was 10.6% of the combined dosage of 10 lb per acre. Soils that had been treated with DDT at higher dosages showed relatively higher residue levels. DDE was the major metabolite found, but dicofol was also detected. From soils treated with aldrin at 20 lb per acre, 5.8% of the applied dosage was recovered, primarily in the form of dieldrin. Small amounts (0.2% of the applied dosage) of photodieldrin were also detected. Lindane was least persistent and only 0.2% of the insecticide applied at 10 or 100 lb per acre were recovered. In addition to lindane, other compounds were isolated from this soil, but they were nontoxic to houseflies. The effects of soil cultivation on the persistence and vertical distribution of DDT and aldrin are discussed. Although the insecticides had been applied and worked into the soil to a depth of 5 in., the 6–9 in. soil layer 10 years later contained about 30% of the total DDT residues recovered and 18% of the total aldrin residues.

A large amount of data on the persistence, metabolism, and movement of DDT, lindane, and aldrin in soils, and the potential translocation of these insecticides into the edible parts of crops has been accumulated by many research groups in various parts of the world. All data indicate the relatively long persistence of certain insecticidal chemicals which have been in use since 1945. In addition, the effects of some of these chemicals on various components of our environment have been studied, pointing to problems which formerly we were not aware of.

In recent years the term half life has frequently been employed to indicate the stability or persistence of agricultural chemicals in nature. This term, originally used to measure the rate of radioactive decay, can be very misleading when generally applied to pesticides in the environment. Thus, the persistence or metabolism of any pesticide depends on a variety of factors, such as its mode of application (Lichtenstein *et al.*, 1962), the formulation in which it was applied (Lichtenstein *et al.*, 1964), and especially the particular environmental conditions to which it is exposed (Lichtenstein, 1966b). In this respect such factors as the types of soil or water into which a particular pesticide might have been deposited, leaf surfaces, microorganisms, climatic conditions, and the presence of other chemicals such as detergents (Lichtenstein, 1966a) all exhibit a decisive effect on the metabolism and persistence of pesticides. The half life of any given pesticidal chemical, therefore, is a function of its chemical nature and the environmental conditions to which it is exposed. It is for these reasons that this term should only be used if strictly qualified with a specific description of the environmental conditions. In discussing the residual persistence of a pesticide it should also be noted that its rate of decline on or in a given substrate is initially faster than later on (Gunther and Blinn, 1955), thus exhibiting different half lives over a given period. This was also demonstrated by Lichtenstein and Schulz in 1959 when it was reported that half of the DDT applied at 10 lb per acre to a loam soil disappeared within 12 months. This period included 4 to 5 months of freezing conditions, when little or no degradation or loss of the chemical from soil occurred. The decline of the DDT concentration in the loam soil was more rapid during the first 6 months after application than during the following 3 years, and its theoretical half life (T $^1/_2$), extrapolated from the rate of decline prior to freezing, was 7.75 months initially. Thereafter the calculated half life for the remaining DDT soil residues was 24.6 months. Depletion curves obtained for aldrin in soils under laboratory conditions indicated three to four different slopes over a 56-day period.

More information relative to the persistence and fate of insecticides in soils is needed. It was felt, therefore, that the presentation of current data from plots established 10 to 15 years ago for the purpose of answering some of these questions would provide information that could be of value for a general understanding of these problems. Accordingly, insecticide treated soils were reinvestigated in 1965, 1969, and 1970 to determine the persistence and vertical distribution of DDT, lindane, and aldrin residues in soils.

METHOD OF PROCEDURE

Soil Treatment and Sampling. PERSISTENCE OF DDT, LINDANE, AND ALDRIN IN SOILS OVER A 15-YEAR PERIOD. In May of 1954 small plots (29 ft × 100 ft) of an agricultural loam soil (Miami silt loam) in Madison, Wis., were treated as described (Lichtenstein *et al.*, 1960) with DDT or lindane at 10 and 100 lb per acre, and with aldrin at 2 and 20 lb per acre. After application the insecticides were rototilled into the soil to a depth of 4 to 5 in. The DDT formulation used contained 84% of the *p,p'*-isomer and 15% of the *o,p'*-isomer. Since in 1954 analytical methods were not as sensitive as they are today, it was felt that for reliable detection of potential metabolites higher insecticidal application rates would be desirable. These soils were then used for growing crops during each of the following five summer seasons. From 1959 on, however, the experimental plots were overgrown with grass. For the determination of DDT residues, 6-in. deep soil samples were collected in the fall of 1965 and 1969 as described (Lichtenstein and Schulz, 1961).

EFFECT OF SOIL CULTIVATION ON THE PERSISTENCE AND VERTICAL DISTRIBUTION OF DDT AND ALDRIN IN SOIL OVER A

Department of Entomology, University of Wisconsin, Madison, Wis. 53706

Table I. DDT and Lindane Residues in a Silt Loam Soil over a 15-Year Period

Recovered After 11 (1965) and 15 Years (1969) from a 6-in. Soil Layer Treated in 1954 with Insecticides

		10 lb/6 in. acre				100 lb/6 in. acre			
		Lb/acre[a]	% Appl.	Tlc →	Glc[b]	Lb/acre	% Appl.	Tlc →	Glc
			From DDT[c]-treated soils						
A. *p,p'*-DDT	1965	1.27	15.1[d]	+	NA[b]	28.80	34.3[d]	+	NA
	1969	0.89	10.5[d]	+	+	13.01	15.5[d]	+	+
B. *o,p'*-DDT	1965	0.27	18.0[e]	+	NA	9.10	60.5[e]	+	NA
	1969	0.16	10.7[e]	+	+	3.73	24.8[e]	+	+
C. *p,p'*-DDE	1965	1.38	16.5[d]	+	NA	17.12	20.3[d]	+	NA
	1969	0.74	8.8[d]	+	+	7.20	8.6[d]	+	+
Sum: A,B,C	1965	2.92	29.0[f]			55.02	56.0[f]		
	1969	1.79	18.0[f]			23.94	24.0[f]		
p,p'-TDE	1965	ND		ND[b]	NA	ND		ND	NA
	1969	ND		ND	+	ND		+	+
Dicofol	1969	ND		ND	+	0.14		+	+
			From lindane-treated soils						
Lindane	1965	0.05	0.50	+	NA	5.28	5.28	+	NA
	1969	0.02	0.20	ND	+	0.17	0.17	+	+

[a] Lb/acre = determined by gas-liquid chromatography. [b] Tlc → Glc = gas-liquid chromatography of compounds isolated by thin-layer chromatography (tlc). + = isolated compound had same R_f value or retention time as reference grade chemical. ND = compounds were not detected. NA = not analyzed. [c] Technical DDT containing 84% of the *p,p'*-isomer and 15% of the *o,p'*-isomer. [d,e,f] In % of the applied dosage of *p,p'*-DDT[d], *o,p'*-DDT[e] or sum of *p,p'*-DDT, *o,p'*-DDT + *p,p'*-DDE[f].

10-Year Period. In 1960, DDT (84% *p,p'*-isomer and 15% *o,p'*-isomer) or aldrin was applied (Lichtenstein and Schulz, 1961) at the lower rate of 4 lb per acre to a 30 ft × 40 ft area of another agricultural loam soil (Carrington silt loam). After soil treatment the insecticides were rototilled into the soil to a depth of 4 to 5 in. To determine if frequent soil cultivation would enhance the loss of DDT or aldrin residues, one half of each plot (15 ft × 40 ft) was disked to a depth of approximately 5 in. on five consecutive days each week for a period of 3 months during the first growing season. In the fall of 1960 soil samples were collected to a depth of 6 in. and analyzed as described (Lichtenstein and Schulz, 1961). After that no further soil cultivation was performed and the plots were overgrown with grass. In the fall of 1970 both the disked and the nondisked portions of these soils were again sampled to a depth of 9 in. For that purpose 30 cores (0.75 in. in diameter, 9 in. deep) were collected from each plot, and treated with either DDT or aldrin at 4 lb per acre. Each core was divided into three 2-in. layers (0–2 in., 2–4 in., and 4–6 in.) to a depth of 6 in., and one 3-in. layer (6–9 in.) to a depth of 9 in. The comparable soil layers of each plot were combined and frozen for future analyses.

Analytical Procedures. The Miami silt loam samples obtained in 1965 and 1969 and the Carrington silt loam samples obtained in 1970 were prepared, extracted with hexane–acetone (1:1), and analyzed by gas-liquid chromatography (glc) as described (Lichtenstein *et al.*, 1970). In addition, aliquots of air-dried soils were extracted with acetonitrile and analyzed by thin-layer chromatography (tlc) as described (Lichtenstein *et al.*, 1970). For qualitative purposes, compounds detected by this method were then isolated and reanalyzed by glc. Analytical grade chemicals were used as reference materials. These were: *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, *p,p'*-TDE, and dicofol, for analyses of the DDT-treated soils, lindane, 1,2,4-trichlorobenzene, 1,3,5-trichlorobenzene, 1,2,3-trichlorobenzene, 1,2,3,4-tetrachlorobenzene, 1,2,4,5-tetrachlorobenzene, 2,4,5-trichlorophenol, pentachlorobenzene, and γ-pentachlorocyclohexene [prepared according to the method described by Reed and Forgash (1970)] for analyses of lindane-treated soils and aldrin, dieldrin, photoaldrin, and photodieldrin for the analyses of aldrin treated soils.

RESULTS AND DISCUSSION

Persistence of DDT, Lindane, and Aldrin in Soils over a 15-Year Period. Results obtained with soils that had been treated in 1954 with DDT and lindane are presented in Table I. Fifteen years after the application of technical DDT at 10 or 100 lb per acre, 18% and 24%, respectively, of the applied insecticidal dosages were present in the upper 6-in. soil layer in the form of *p,p'*-DDT, *o,p'*-DDT, and *p,p'*-DDE. Residues of *p,p'*-DDT plus *o,p'*-DDT only, recovered in 1969 from the 10 and 100 lb per acre treated plots, amounted to 10.6% (1.05 lb per 6-in. acre) and 17% (16.74 lb per 6-in. acre), respectively, of the dosages applied 15 years previously. This indicated the relatively higher stability of the insecticide in soil which had been treated at the higher dosage. This finding was in agreement with earlier data when it was shown that the relative loss of an insecticide from soils (Lichtenstein and Schulz, 1959) or water (Lichtenstein and Schulz, 1970)—expressed in percent of the applied dosage—was to some extent inversely proportional to the initial insecticide concentration. While 32.5% of the applied *p,p'*-DDT was lost from the soil treated with 10 lb per acre of technical DDT during the first growing season in 1954 (Lichtenstein *et al.*, 1960), a total of 89.5% of this isomer was lost during the 16 growing seasons between 1954 and 1969. The *o,p'*-isomer of DDT was more persistent than the *p,p'*-isomer, as indicated by data obtained from the soil that had been treated with DDT at 100 lb/acre. During the 15-year period only 75.2% of *o,p'*-DDT had been lost, as opposed to 84.5% of the *p,p'*-DDT.

Of the DDT metabolites detected, *p,p'*-DDE was the most prevalent. Its concentration was similar to *p,p'*-DDT in the soil that had been treated at 10 lb per acre, but amounted to about half the concentration of *p,p'*-DDT detected in the soil that was treated at 100 lb per acre. Although during the years 1965 through 1969 DDE presumably was still being formed from DDT, the amount detected in both soil plots in 1969 was only one half of the amount that had been detected 4 years previously. *p,p'*-TDE could not be detected by glc of hexane–acetone extracts, but was isolated in trace amounts by tlc and confirmed by glc. Dicofol, the hydroxylation product of DDT, was present in trace amounts in the 10 lb per acre treated soil and 0.14 lb per acre was found in

Table II. Vertical Distribution of Insecticidal Residues in a Silt Loam, 10 Years after the Application in 1960 of Technical DDT (84% *p,p'*-DDT, 15% *o,p'*-DDT) at 4 lb per Acre to the Upper 5-in. Soil Layer

Soil Layers (In.)	Recovered in Fall of 1970[a]: A. *p,p'*-DDT Lb/acre	A. *p,p'*-DDT % Distr.[b]	B. *o,p'*-DDT Lb/acre	B. *o,p'*-DDT % Distr.	C. *p,p'*-DDE Lb/acre	C. *p,p'*-DDE % Distr.	A + B + C Lb/acre	A + B + C % Distr.
				Nondisked				
0–2	0.141	13	0.030	10	0.051	14	0.222	13
2–4	0.234	21	0.063	21	0.081	23	0.378	22
4–6	0.357	32	0.107	36	0.121	34	0.585	33
Sum: 0–6	0.732		0.200		0.253		1.185	
6–9	0.362	34	0.097	33	0.103	29	0.562	32
Sum: 0–9	1.094	100	0.297	100	0.356	100	1.747	100
% Appl.[c,d]	33[c]		49[d]		11[c]		44[c,d]	
				Disked (1960 only)[e]				
0–2	0.158	21	0.033	15	0.057	21	0.248	20
2–4	0.192	26	0.057	26	0.073	27	.322	26
4–6	0.212	28	0.069	32	0.078	29	0.359	29
Sum: 0–6	0.562		0.159		0.208		0.929	
6–9	0.185	25	0.058	27	0.064	23	0.307	25
Sum: 0–9	0.747	100	0.217	100	0.272	100	1.236	100
% Appl.	22[c]		36[d]		8[c]		31[c,d]	

[a] Data determined by gas–liquid chromatography (glc) and confirmed by glc of compounds that were isolated by thin-layer chromatography. Neither TDE nor dicofol could be detected. [b] Distribution of residues in soil layers in percent of totally recovered chemical in the 0–9 in. soil layer. [c,d] In percent of the applied dosage of [c] *p,p'*-DDT, [d] *o,p'*-DDT, or [c,d] *p,p'*- + *o,p'*-DDT. [e] Soil was disked daily to approximately 5 in. during three summer months in 1960.

Table III. Vertical Distribution of Aldrin and Dieldrin Residues in a Silt Loam 10 Years after the Application of Aldrin at 4 lb per Acre to the Upper 5-in. Soil Layer

Soil Layers (In.)	Recovered in Fall of 1970[a]: A. Aldrin Lb/acre	A. Aldrin % Distr.[b]	B. Dieldrin Lb/acre	B. Dieldrin % Distr.	A + B Lb/acre	A + B % Distr.
			Nondisked			
0–2	0.001	11	0.116	27	0.117	27
2–4	0.003	33	0.130	31	0.133	31
4–6	0.003	33	0.100	24	0.103	24
Sum: 0–6	0.007		0.346		0.353	
6–9	0.002	23	0.075	18	0.077	18
Sum: 0–9	0.009	100	0.421	100	0.430	100
% Appl.[c]	0.22		10.5		10.8	
			Disked (1960 only)[d]			
0–2	0.001	13	0.052	25	0.053	25
2–4	0.002	29	0.061	29	0.063	29
4–6	0.002	29	0.061	29	0.063	29
Sum: 0–6	0.005		0.174		0.179	
6–9	0.002	29	0.035	17	0.037	17
Sum: 0–9	0.007	100	0.209	100	0.216	100
% Appl.	0.18		5.2		5.4	

[a] Data determined by gas–liquid chromatography (glc). Confirmed by glc of compounds that were isolated by thin-layer chromatography. Neither photoaldrin nor photodieldrin could be detected. [b] Distribution of residues in soil layers expressed in percent of totally recovered compounds in the 0–9 in. soil layer. [c] In percent of applied aldrin. [d] Soil was disked daily to approximately 5 in. during three summer months in 1960.

the 100 lb per acre treated soil. Data, therefore, indicate that the concentration of DDT in the soil declined at a relatively slow rate, especially after the first growing season in 1954. Moreover, the insecticide was relatively more persistent in soil to which DDT had been applied at the abnormally high dosage of 100 lb per acre.

Results obtained after analyses of soil treated with lindane in 1954 are included in Table I. This insecticide was the least persistent of the three tested, and it disappeared at a considerably faster rate than did DDT, since a loss of 94.7 and 99.5% of the applied lindane dosages was registered 11 years after the insecticidal application. By the end of the first growing season in 1954, however, 57% of the applied lindane had already been lost from the 10 lb per acre treated soil, and 45% had been lost from the 100 lb per acre treated plot. With the exception of lindane, all compounds isolated by tlc from these soils in 1969 were nontoxic after a topical application of extracts representing 1 g of soil to the abdomen of female houseflies (C.S.M.A. strain). Analyses by glc of these nontoxic isolates from the soil treated at 10 lb per acre resulted in a peak that corresponded to 1,2,3,4-tetrachlorobenzene and three additional peaks which did not correspond with any of the reference materials. Analyses by glc of the tlc isolates from the 100 lb per acre treated soil yielded, in addition to lindane, two peaks with retention times identical to those obtained with 1,2,3,4-tetrachlorobenzene, and γ-pentachlorocyclohexene plus four additional peaks which did not correspond with any of the reference materials.

Aldrin was less persistent than DDT, but was more persistent than lindane, primarily due to the epoxidation of aldrin to dieldrin in soil. Eleven years after soil treatment with aldrin at 2 lb per acre, this insecticide could no longer be detected, but 5.7 and 5.3% of the applied dosage were still recovered in the form of dieldrin in 1965 and 1969, respectively. The application of aldrin at 20 lb per acre in 1954, however, resulted in the detection of measurable amounts of aldrin 15 years later. At that time a total of 5.78% (1.157 lb per acre) of the applied dosage was recovered, of which 0.18% was in the form of aldrin and 5.6% was in the form of dieldrin. In addition, this soil contained 0.048 lb per acre of photodieldrin, corresponding to 0.23% of the applied aldrin.

EFFECT OF SOIL CULTIVATION ON THE PERSISTENCE AND VERTICAL DISTRIBUTION OF DDT AND ALDRIN OVER A 10-YEAR PERIOD. Soils treated in spring of 1960 with DDT or aldrin at 4 lb per acre showed, in the fall of that year, appreciable differences in the level of DDT or aldrin–dieldrin residues between the disked and nondisked portion of the plot (Lichtenstein and Schulz, 1961). While only 26% of the applied DDT was lost during a 4 month period from the nondisked loam plot, a loss of 44% was registered in the soil which had been disked during the first 3 months following soil treatment. This effect was more pronounced when aldrin had been applied to the soil, since in fall of 1960 a loss of 53% and 70% was registered in the nondisked and disked plots, respectively. The mechanism of the effect of frequent soil cultivation on the loss of insecticidal residues is difficult to explain, since a variety of physical, chemical, and biological factors could be involved in this phenomenon.

Results obtained after DDT-treated soils had been sampled in 1970 are presented in Table II. The *o,p'*-isomer of DDT was more persistent than the *p,p'*-isomer. In addition to the originally applied *p,p'*- and *o,p'*-DDT, considerable amounts of *p,p'*-DDE and trace amounts of both *p,p'*-TDE and dicofol could be detected. The effects of soil cultivation by disking 10 years previously were still noticeable, since only 31% of the applied DDT was recovered in the form of *p,p'*-, *o,p'*-DDT, and *p,p'*-DDE from the disked soil, compared to 44% from the nondisked soil. The quantity of *p,p'*- and *o,p'*-DDT amounted to 0.96 lb per acre (24% of the applied dosage) in the disked soil and to 1.39 lb per acre (35% of the applied dosage) in the nondisked soil. Although DDT had been applied and worked into the soil to a depth of 5 in., the 6–9 in. soil layer contained 25% to 32% of the totally recovered DDT residues. Cultivation of the soil in 1960 resulted in a more uniform distribution of the insecticidal residues in the soil, in comparison to the noncultivated soil. In this latter case, most (65%) of the residues were located in the 4–9 in. soil layers and least (13%) were located in the upper 0–2 in. layer.

Results obtained from aldrin treated soils are presented in Table III. The major chemical recovered was dieldrin (over 97% of the total residue), the rest being aldrin. No photoisomers of these compounds could be detected. The total insecticide residue recovered from the disked and nondisked soils amounted to 5.4% and 10.8% of the applied aldrin. Since these insecticides volatilize from soils to a larger degree than DDT (Lichtenstein and Schulz, 1970) it was not surprising to find only 17 to 18% of the totally recovered residue in the 6–9 in. soil layer, as opposed to 25 to 32% with DDT. No differences in the distribution of the residues in the four soil layers were noticeable between the disked and nondisked soils.

In conclusion, it should be reemphasized that DDT, lindane, and aldrin are metabolized in soils or volatilize from soils and disappear at relatively slow rates. Fifteen years after soil treatment, 10.6% of the DDT applied at 10 lb per acre, 5.8% as dieldrin of the aldrin applied at 20 lb per acre, and 0.2% of the lindane applied at 10 lb per acre were recovered from these soils. Soil cultivation was just one of the factors that affected the disappearance of these insecticides from soils.

ACKNOWLEDGMENT

We thank A. J. Forgash of Rutgers University for supplying us with the various potential lindane metabolites and T. T. Liang for his technical assistance.

LITERATURE CITED

Gunther, F. A., Blinn, R. C., "Analysis of Insecticides and Acaricides," Interscience, New York, N.Y., 1955, p 131.
Lichtenstein, E. P., *J. Econ. Entomol.* **59**, 985 (1966a).
Lichtenstein, E. P., Scientific Aspects of Pest Control, Nat. Acad. Sci. Publ. No. 1402, 221 (1966b).
Lichtenstein, E. P., Schulz, K. R., *J. Econ. Entomol.* **52**, 124 (1959).
Lichtenstein, E. P., Schulz, K. R., *J. Econ. Entomol.* **54**, 517 (1961).
Lichtenstein, E. P., Schulz, K. R., J. AGR. FOOD CHEM. **18**, 814 (1970).
Lichtenstein, E. P., Myrdal, G. R., Schulz, K. R., *J. Econ. Entomol.* **57**, 133 (1964).
Lichtenstein, E. P., DePew, L. J., Eshbaugh, E. L., Sleesman, J. P., *J. Econ. Entomol.* **53**, 136 (1960).
Lichtenstein, E. P., Mueller, C. H., Myrdal, G. R., Schulz, K. R., *J. Econ. Entomol.* **55**, 215 (1962).
Lichtenstein, E. P., Schulz, K. R., Fuhremann, T. W., Liang, T. T., J. AGR. FOOD CHEM. **18**, 100 (1970).
Reed, W. T., Forgash, A. J., J. AGR. FOOD CHEM. **18**, 475 (1970).

Received for review December 11, 1970. Accepted February 1, 1971. Published with the approval of the Director of the Research Division, College of Agricultural and Life Sciences. Research supported in part by a grant from the U.S. Public Health Service (FD-00258). Contribution by Project 1387 from the Wisconsin Agricultural Experiment Station as a collaborator under North Central Regional Cooperative Research Project 96 entitled "Environmental Implications of Pesticide Usage."