

# Installment Application Effects upon Insecticide Residue Content of a California Soil

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From 1953 to 1957 annual applications of eight organochlorine insecticides were worked into Holtville sandy clay. One pound of lindane per acre per year resulted in little total organically bound chloride in the soil during 1953 to 1962. Measurable enduring soil insecticide residues occurred in soil receiving DDT, toxaphene, chlordane, dieldrin, endrin, heptachlor, and aldrin at the respective rates of 20, 20, 10,

5, 5, 5, and 5 lb/acre/year. A rank of decreasing persistence (persistence index: 1.00 = no degradation or other disappearance during first year) over an 11-year period places the treatments as DDT (0.26), dieldrin (0.22), endrin (0.20), toxaphene (0.18), heptachlor (0.14), chlordane (0.13), and aldrin (0.09).

The man-made accumulation of often undesirable chemicals in the soil is not a new problem in agriculture. The use of agricultural chemicals has insured satisfactory production of high quality food products and protected the health of animals, including man. Because of certain undesirable side effects ranging from loss of economic return on pesticide expense to the creation of undesirable persisting residues, unrestrained use of most pest control and other treatments has not been recommended. Application of excessive rates of nitrogenous materials, such as the use by prehistoric man of fish as fertilizer, has caused ammonium toxicity to plants. Soon after the turn of the century repeated heavy treatments of certain insecticides to fruit orchards were applied to control insect pests. These resulted in accumulations of arsenic in the soil to a point where many trees, as well as subsequent vegetable and field crop plantings, became stunted and sometimes died.

With the development of the organochlorine insecticides in the early 1940's, man made tremendous progress in controlling his insect pests and increasing yields beyond all expectations. But again this was not accomplished without causing some losses and creating hazards. Off-flavors often appeared in root crops, particularly potatoes, that were grown in soils treated with benzenehexachloride. Plants in general absorb insecticide residues from the soil and translocate them into the edible portion (Ebeling, 1963, and subsequently many others). One of the most important aspects which must be understood to evaluate this source of food contamination involves understanding the manner in which pesticide residues accumulate in and dissipate from the soil.

The major factors influencing insecticide residue levels in soils have been discussed (Edwards, 1966). Ebeling (1963) discusses the fact that: "The disappearance of most pesticide residues appears to depend on first-order reaction kinetics and accordingly may be plotted as a straight-line relationship of the logarithm of the quantity of residue retained and the period of exposure..." Earlier Gunther and Blinn (1955) indicated that some of these curves consisted of at least two components, so it was clear that the promising mathematical model would provide for a number of components. Each of these components should be associated with a mechanism of loss as done with soil insecticide residues by Edwards (1966; Fig. 4).

Chisholm (Fig. 1 in Foster *et al.*, 1956) calculated the

amounts of insecticide accumulated in soil following applications of 10 lb/acre/year, assuming 40, 60, 80, and 90% persistence annually. These insecticide residue persistence curves seem to represent the sums of infinite geometric series.

The purpose of this paper is to report on an 11-year experiment designed to determine the effect of installment applications of eight insecticides on the insecticide residue contents of soil.

## EXPERIMENTAL

**Materials and Methods.** In 1953 a randomized complete block field experiment was established at the University of California Field Station in Imperial Valley, 6 miles east of El Centro, Calif. The plots were 21 × 104 ft; 6-ft border strips were arranged between plots, separated by dikes 1 ft high. Five replicates were furnished as blocks. The properties of the Holtville sandy clay involved are given in Table I.

The eight organochlorine insecticides were applied according to times shown in Table II at the rates of active ingredients shown in Table III. The preparations were sprayed onto the soil surface and disked the same day into the upper 6 in. of the soil.

Row crops were grown yearly. Before soil samples were taken, the soil was irrigated and cultivated. Samples were collected by obtaining thirty 1.125-in. cores from each replicate for each treatment over the 0 to 6-in. soil horizon. Within 1 week the moist samples were passed through 0.25-in. mesh hardware cloth. Sieved samples were either extracted immediately or stored at 5° C. Samples in storage were extracted within 1 month; no effect of storage time was apparent in the final data.

The method of chemical analysis determined the total organically bound chloride content of the soil samples. To 1 kg of field moist soil was added 500 ml of isopropyl alcohol. One hour later, 1 l. of *n*-hexane was added and the sample was tumbled at 58 rpm for another hour in a drum-tumbler stripper (Gunther and Blinn, 1955). Subsequent handling followed Gunther and Blinn (1955). The resulting extracts were washed free of inorganic chloride 9× with 250-ml portions of distilled water, followed by 2× with 250-ml portions of conductivity water, and then stored over 5 g of anhydrous sodium sulfate at 5° C until analyzed within 2 months. The total organically bound chloride present was determined by the direct potentiometric method after combustion in a stream of oxygen (Gunther and Blinn, 1955). The resulting residue data were calculated as parent compound and corrected to oven-dry weight of the soil at 105° C.

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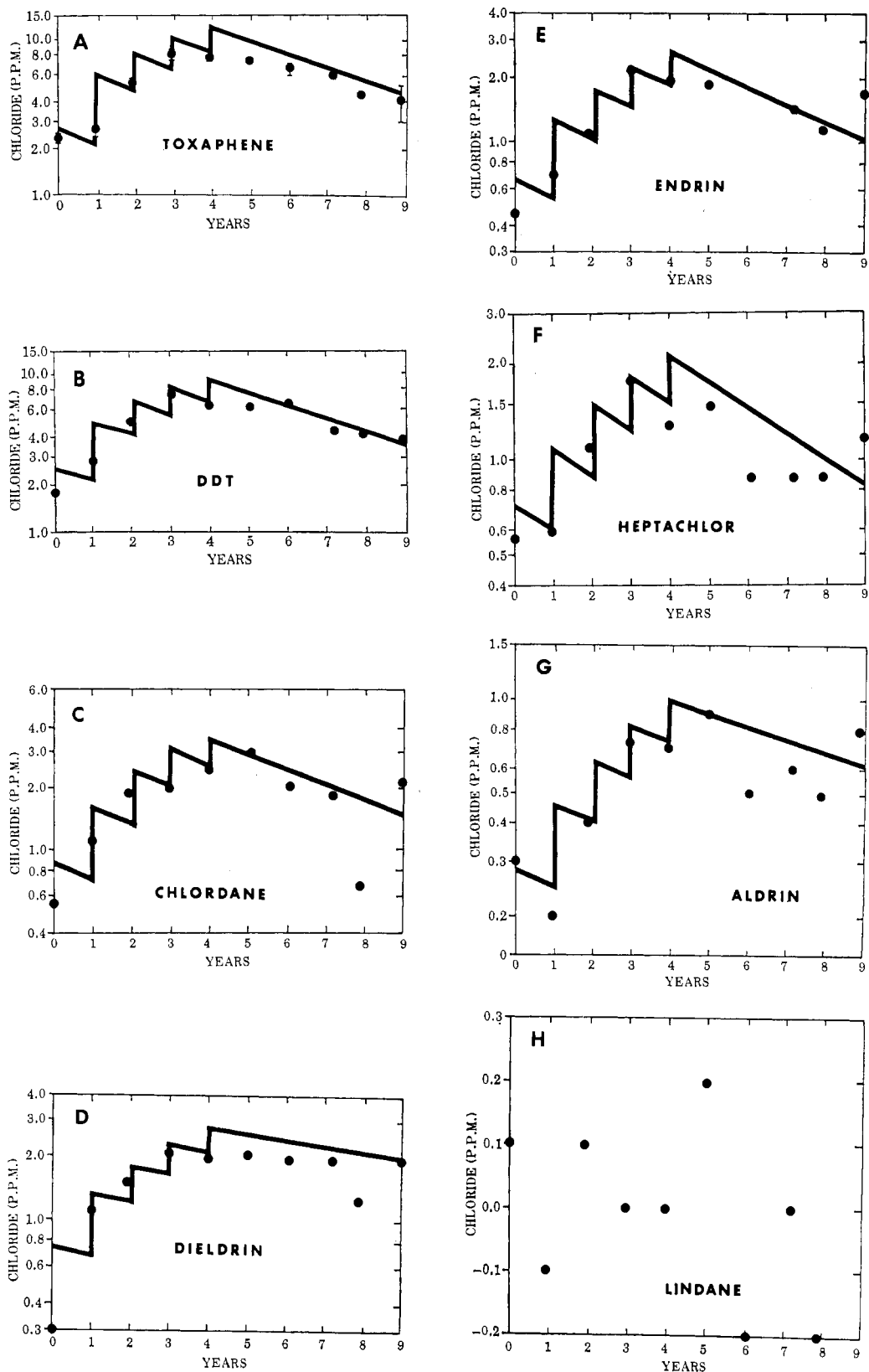


Figure 1. Variations in the total organically bound chloride contents of soil samples, expressed as persistence curves. The vertical lines through the circles in the toxaphene curve (A) represent the standard deviations which apply to all the curves. A curve cannot be fitted to the lindane (H) data

#### EXPERIMENTAL RESULTS

Figure 1 shows the total organically bound chloride contents of the soil samples. Each point represents the mean for the five field replicates. Each mean has been reduced by that from the untreated soil. Because of this correction, negative values reflecting experimental error occur for lindane in Figure 1. Each year the disagreement among replicates furnished an estimate of experimental error. These errors were used to

estimate the standard deviation of the difference between two means. The vertical lines through the toxaphene means in Figure 1 represent these standard deviations, which also apply to the other treatment means at the corresponding sampling times.

#### DISCUSSION

All insecticide applications except that of lindane resulted

**Table I. Chemical and Physical Properties of Holtville Sandy Clay in 1953**

Soil Property	Measurement
Mechanical analysis	
sand	48%
silt	16%
clay	36%
Soil pH	7.5 ± 0.1
Electrical conductivity (25° C)	3.8 ± 0.6 mmho/cm
Composition of saturation extract	
Ca plus Mg	36 ± 8 mequiv/l.
Na	11 ± 2 mequiv/l.
K	0.6 ± 0.1 mequiv/l.
SO <sub>4</sub>	34 ± 5 mequiv/l.
NO <sub>3</sub>	5.0 ± 0.6 mequiv/l.
Cl	5.2 ± 1.0 mequiv/l.
HCO <sub>3</sub>	2.5 ± 0.2 mequiv/l.

**Table II. Time Table for Soil Insecticide Field Work**

Time	Action
10/20/53	Initial soil sampling
10/20/53	Initial treatments
10/5/54	2nd soil sampling
10/6/54	2nd treatment
9/14/55	3rd soil sampling
10/25/55	3rd treatment
9/17/56	4th soil sampling
9/18/56	4th treatment
9/18/57	5th soil sampling
9/18/57	5th treatment
10/1/58	6th soil sampling
10/1/59	7th soil sampling
11/10/60	8th soil sampling
7/31/61	9th soil sampling
8/9/62	10th soil sampling
10/1/63	11th soil sampling

**Table III. Amounts (lb/Acre) of Active Ingredients Annually Applied to Soil**

Insecticide	Time					Mean	Total
	1953	1954	1955	1956	1957		
Toxaphene	19.6	20.0	20.8	20.8	22.4	20.7	103.6
DDT	19.5	20.0	20.8	23.2	20.0	20.7	103.5
Chlordane	9.4	10.0	11.4	11.6	9.7	10.4	52.1
Endrin	4.9	5.0	5.2	5.3	5.4	5.16	25.8
Dieldrin	6.0	5.2	4.4	5.1	5.1	5.16	25.8
Heptachlor	5.0	5.2	5.0	4.8	4.9	4.98	24.9
Aldrin	4.7	4.6	4.6	5.4	5.0	4.86	24.3
Lindane	0.9	1.0	1.0	1.0	1.0	1.00	4.9
Untreated	0.0	0.0	0.0	0.0	0.0	0.00	0.0

in significant ( $P < 0.05$ ) residues in the soil. As the other insecticides were added annually to the soil, the pesticide residue found in the soil climbed from 0 toward an apparent limit. When periodic addition ceased, each pesticide residue level fell off from the maximum value in a typical persistence curve. Due to experimental error, the overall trends for the insecticides applied at lower rates are expected to be less clear. Observation of Figure 1 shows that the trend in the residues was nearly ideal for both toxaphene and DDT treatments which involved 20 lb/acre annually. The other insecticides were applied at lower rates so that contamination or other error would cause greater percent deviation from the ideal trend line.

**Mathematical Model.** It was desirable to carry the quantitative character of this data toward the summary so that

more detailed aspects became evident and that greater confidence existed for certain conclusions. Nonlinear regression analysis was practiced. The purpose was to relate insecticide rate, application time, and residence time to soil insecticide residue levels. The following general mathematical model is hypothesized.

$$\log \text{Cl} = \sum_{j=1}^n f_j(b_{j0} + b_{j1}t) = f_1(b_{10} + b_{11}t) + f_2(b_{20} + b_{21}t) + \dots + f_n(b_{n0} + b_{n1}t) \quad (1)$$

Here log Cl is the common logarithm of the ppm of total organically bound chloride content of the soil at time  $t$  after insecticide was placed in the soil. Each value of  $j$  is associated with a mechanism of loss. Let  $j = 1$  implicate application losses and  $j = 2$  to  $n$  relate to losses from the soil sampling volume. The weight function,  $f_j$ , indicates the portion of the total residue subject to loss by the  $j$ th phenomenon. These functions are determined by pesticide properties, soil properties, time, biological factors, cultural practices, etc. Constants are designated by  $b_{ji}$ . This model is a modified formulation of the situation hypothesized by Edwards (1966; Fig. 4). It represents the dissipation of pesticide residue after a single soil treatment which occurred at  $t = 0$ . Needed modifications of the model will occur when pesticide behavior in the environment is studied quantitatively.

We must adapt this model to our experimental conditions. Assume that losses from the soil sampling volume can be completely explained by the effects of the following factors: vaporization ( $j = 2$ ); leaching (3); chemical degradation (4); and enzymatic degradation (5). Therefore, the model (1) is reduced to the case of  $n = 5$ .

Application losses were assumed equal for the five applications. The weight function,  $f_1$ , went to 0 shortly after application because little pesticide residue is blown from the plot after incorporation; thus, the application loss  $f_1(b_{10} + b_{11}t)$  is a constant,  $b_{12}$ .

Edwards (1966) points out that volatilization loss of organochlorine insecticides from soil is most evident immediately after application. This must be partly due to geometric factors because later it is slower and depends upon soil type. It is reasonable to assume that the vaporization weight function,  $f_2$ , approaches its long-term value by the end of 1 year. Therefore, if the soil is not sampled within 10 months after application time, the vaporization component,  $f_2(b_{20} + b_{21}t)$ , may not be distinguished from other long-term effects. The effect can be partitioned to provide  $b_{22} + f_2(b_{20} + b_{21}t)$ . The leaching of organochlorine insecticides is a very minor loss so that  $f_3 = 0$  and the term  $f_3(b_{30} + b_{31}t)$  drop out. Chemical degradation of organochlorine insecticides in soil has not been partitioned from that of enzymatic degradation. These two factors were assumed to act over a comparable time period.

The reduction of eq 1 follows.

$$\log \text{Cl} = (b_{12} + b_{22} + f_2b_{20} + f_4b_{40} + f_5b_{50}) + (f_2b_{21} + f_4b_{41} + f_5b_{51})t \quad (2)$$

Let

$$\log A = b_{12} + b_{22} + f_2b_{20} + f_4b_{40} + f_5b_{50} \quad (3)$$

$$b = f_2b_{21} + f_4b_{41} + f_5b_{51} \quad (4)$$

Then

$$\log \text{Cl} = \log A + bt \quad (5)$$

So the general mathematical model capable of relating total

organically bound chloride to residence time of a single insecticide dose is exponential.

$$Cl = A 10^{bt} \quad (6)$$

In practice, however, a series of insecticide applications occur. In the case of equal installments under the same conditions  $A$  would remain the same for each application. The total organic chloride content of the soil would then vary according to the sum of a geometric series. The following series was formulated for the case when the soil was sampled just prior to the next annual insecticide application.

$$Cl_i = A \sum_{j=0}^{(i-1)} 10^{(s_i - a_j)b} + e_i \quad (7)$$

where  $Cl_i$  = the amount (ppm) of total organic chloride in the soil sample collected during year  $i$ ;  $A$  = a constant which reflects the depreciated amount (ppm) of organic chloride annually added to the soil sampling volume;  $s_i$  = the soil sampling time (years);  $a_j$  = the insecticide application time (years) for the  $j$  year treatment;  $b$  = the dissipation constant; and  $e_i$  = the disagreement between observed and calculated total organic chloride.

**Regression Problem.** Adapt this model to the experimental conditions. Termination of the periodic application of insecticide is reflected by a partition of the geometric series. The bracketed term in eq 8 reflects the ascending level of soil insecticide residue as each addition of pesticide provides a depreciated residue increment to the total soil insecticide residue reservoir. The exponential term to the right of this reflects the loss from the total residue reservoir after periodic treatment has ceased.

$$Cl_m = A \left[ \sum_{j=0}^{i-1} 10^{(s_i - a_j)b} \right] 10^{(s_k - s_5)b} + e_m \quad (8)$$

$$\begin{aligned} i &= 0, 1, 2, \dots, 5 & m &= i \text{ or } k \\ j &= 1, 2, \dots, i-1 & (s_k - s_5) &> 0 \text{ if } k > 5 \\ k &= 6, 7, \dots, 9 & (s_k - s_5) &= 0 \text{ if } k < 5 \end{aligned}$$

Solve this for the error terms. Provide an equation representing each year's data. The formulation found in Table IV results.

This set was studied by approximation methods including group relaxation. Other methods (Henrici, 1964; Hart and Motzkin, 1956) would work as efficiently. The aim was to reduce the sum of squares for  $e_m$  to a minimum by an appropriate choice of  $A$  and  $b$ . A good estimate for the DDT data in Figure 1 is  $A = 2.74$  ppm and  $b = -0.084$  year<sup>-1</sup>. Less ideal solutions of the systems for toxaphene, chlordane, dieldrin, endrin, heptachlor, and aldrin appear in Table V.

Using these constants the persistence curves were drawn for Figure 1. The vertical portions of the curves indicate the five annual insecticide applications. The heights of the left face of the saw-teeth depend upon the amounts of persistent insecticide actually added to the soil sampling volume; this is determined by the application rate minus application and early vaporization losses in this case.

The analysis of variance in Table VI shows that the regression associates 96% of the DDT treatment sums of squares with the treatment parameters. Similar success was obtained for the other six treatments. When total (10 d.f.) and regression (2 d.f.) mean squares were tested against their associated lack of fit (8 d.f.) mean square, they were all significant at  $P$  smaller than 0.05. Comparison of the lack of fit mean squares with a long-term, among replicates, error mean square (360 d.f.) of 0.15 indicates no significant lack of fit at  $P = 0.05$ . This shows that the regression equations developed do predict insecticide residue content of soil which had been given the described treatment.

Table IV. Formulation for the Insecticide Residues in Soil

$$\begin{aligned} e_0 &= Cl_0 \\ e_1 &= Cl_1 - A 10^{(s_1 - a_0)b} \\ e_2 &= Cl_2 - A [10^{(s_2 - a_0)b} + 10^{(s_2 - a_1)b}] \\ e_3 &= Cl_3 - A [10^{(s_3 - a_0)b} + 10^{(s_3 - a_1)b} + 10^{(s_3 - a_2)b}] \\ e_4 &= Cl_4 - A [10^{(s_4 - a_0)b} + 10^{(s_4 - a_1)b} + 10^{(s_4 - a_2)b} + 10^{(s_4 - a_3)b}] \\ e_5 &= Cl_5 - A [10^{(s_5 - a_0)b} + 10^{(s_5 - a_1)b} + 10^{(s_5 - a_2)b} + 10^{(s_5 - a_3)b} + \\ &\quad 10^{(s_5 - a_4)b}] = Cl_5 - \Gamma_5 \\ e_6 &= Cl_6 - \Gamma_5 10^{(s_6 - s_5)b} \\ e_7 &= Cl_7 - \Gamma_5 10^{(s_7 - s_5)b} \\ e_8 &= Cl_8 - \Gamma_5 10^{(s_8 - s_5)b} \\ e_9 &= Cl_9 - \Gamma_5 10^{(s_9 - s_5)b} \end{aligned}$$

Table V. Estimates of the Dissipation Constant,  $b$ , and Constant  $A$  for Regression Equations

Insecticide	$b$ (Year <sup>-1</sup> )	$A$ (ppm)
DDT	-0.084	2.7
Chlordane	-0.072	0.94
Heptachlor	-0.072	0.46
Toxaphene	-0.072	2.5
Endrin	-0.072	0.57
Aldrin	-0.048	0.26
Dieldrin	-0.024	0.62

Table VI. Analysis of Variance to Partition Individual Soil Insecticide Treatment Sums of Squares for Regression Considerations

Insecticide Treatment	Coefficient of Determination	Mean Square for Indicated Source of Variation		
		Total	Regression	Lack of Fit
Toxaphene	0.97	6.75	32.66	0.27
DDT	0.96	5.16	25.75	0.27
Chlordane	0.97	0.75	3.63	0.03
Endrin	0.91	0.44	2.00	0.05
Dieldrin	0.93	0.66	2.80	0.05
Heptachlor	0.93	0.25	1.17	0.02
Aldrin	0.78	0.11	0.42	0.03

**Implications.** It is felt that these equations fit well enough to consider their implications. First it should be pointed out that total organically bound chloride determinations are non-specific for the individual insecticide residue, so the regression equations provide upper limits for the corresponding specific residue functions. Since total organic chloride content is provided, multiplication of the function by the chloride conversion factor for the most toxic known residue provides an upper limit for the hazard. For example, multiplication of the heptachlor equation by 1.57 will scale it to heptachlor epoxide hazard.

While in its installment phase, function 8 is a specific case of the infinite geometric series

$$\begin{aligned} Cl_{n-1} &= A (10^{0b} + 10^{1b} + 10^{2b} + \dots + 10^{(n-1)b}) - A \\ &= A (1 - 10^{nb}) / (1 - 10^b) - A \quad (9) \end{aligned}$$

Here  $n - 1$  is the number of years from the initial treatment.  $Cl_{n-1}$  is the total organic chloride content of the soil at  $n - 1$  years and just before the succeeding application. As  $n$  becomes infinite, the summation term converges since  $10^b$  is less than 1. A limit can be calculated

$$\lim_{n \rightarrow \infty} Cl_{n-1} = A / (1 - 10^b) - A \quad (10)$$

For the DDT treatment the residue measured 1 year after the last treatment approaches the limit

$$\lim_{n \rightarrow \infty} Cl_{n-1} = 2.74 / (1 - 10^{-0.084}) - 2.74 = 13 \text{ ppm} \quad (11)$$

Table VII. Consequences Derived from the Regression Analysis

Treatment	Maximum Organic Cl Level (ppm)	Persistence Half-life (Years)	Insecticide Cl Content (%)	Persistent Cl Index	Vapor Pressure <sup>a</sup> (mm)
Toxaphene	14	4	68	0.18	$3 \times 10^{-1}$
DDT	13	4	50	0.26	$3 \times 10^{-7}$
Chlordane	5	4	69	0.13	$1 \times 10^{-5}$
Endrin	3	4	56	0.20	$2 \times 10^{-7}$
Dieldrin	10	13	56	0.22	$1 \times 10^{-6}$
Heptachlor	3	4	66	0.14	$3 \times 10^{-4}$
Aldrin	2	~6 <sup>b</sup>	58	0.09	$3 \times 10^{-5}$

<sup>a</sup> From Zweig (1964). Refers to the chemical at 25° C. <sup>b</sup> Aldrin plus conversion product dieldrin.

The corresponding limits for the other treatments are given in Table VII as maximum organic chloride levels. The most striking thing about them is that they increase with insecticide application rate.

The time,  $\beta$ , to half of maximum buildup can be ascertained. It shall be defined by the relationship

$$1/2 \left( \lim_{n \rightarrow \infty} Cl_{n-1} \right) = Cl_{\beta} \quad (12)$$

Here  $\beta$  is analogous to  $n - 1$  except that it is a dependent variable instead of an independent one. Thus, it can replace  $n - 1$  in eq 9 which can be used with eq 10 to derive the function

$$\beta = -(\log 2)/b \quad (13)$$

The relationship can be used to estimate that annual application of 20.7 lb of DDT per acre will raise the soil insecticide residue level to over half its maximum possible level in 4 years.

$$\beta = -(\log 2)/(-0.084) = 3.6 \text{ years} \quad (14)$$

Our mathematical model constrains  $\beta$  to be the same as the persistence half-life. The persistence half-lives appear in Table VII. These correspond to the buildup half-lives when soil samples are taken immediately after application. Dieldrin residues in soil are highly persistent under the conditions of this study.

Division of  $A$  by the average annual amount of organically bound chloride which left the sprayer produces the persistent chloride index in Table VII. These indices appear beside the vapor pressure data for the insecticides at 25° C. Persistence is, therefore, slightly related to the vapor pressures of the chemicals; the agreement is as good as that reported by Edwards (1966). Slightly volatile compounds get into and remain in the sampling volume to a greater extent than moderately volatile and volatile ones. For these latter compounds it is especially clear that azeotropic and soil adsorption effects on vapor pressure must be considered to rationalize short-term soil insecticide losses.

**Generalizations.** Further work is required to establish whether or not pesticide residues in soil build up according to a geometric series. In case this relationship is supported, a 3-year study could produce necessary data for legal applications or for recommendation of safe local practices. The key relationship would be the summation eq 9. The data for two application periods would be the minimum requirement to estimate the unknowns  $A$  and  $b$ . In case of legal interests these should agree within experimental error with the claims made.

Observation of Figure 1 and the analysis of variance in Table VI show that the suggested general mathematical model

$$Cl_i = A \sum_{j=0}^{(i-1)} 10^{(s_i - a_j)b} \quad (15)$$

fits the insecticide residue data well. Here the variables are defined as in eq 7. Since none of the lack of fit mean squares is significant, the distances of the observed points from the curve largely represent the individual  $e_i$ . This experimental error includes the bias of an over-simple model, the effect of soil moisture content on extraction efficiency, and the effect of environmental variation at the field location. A modification of the suggested model can be used to help account in a quantitative manner for the field variation of these additional variables; this operation would bring the points into even better agreement with the ideal curve.

In addition to representing these residue data satisfactorily, the model can be used as a tool to study pesticide residue degradation factors. Its good potential here rests on the fact that it has a rational development. Its rationality arises from the fact that it can be derived from a modification of Edward's soil insecticide degradation relationship. The lower envelope to the buildup curve looks like those suggested by Chisholm (Fig. 1 in Foster *et al.*, 1956); thus a means has been furnished to cope with the buildup and decline of pesticide residues in nature.

#### NOMENCLATURE

Aldrin	1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo,exo-5,8-dimethanonaphthalene
Chlordane	1,2,4,5,6,7,8,8-octachloro-2,3,3a,4,7,7a-hexahydro-4,7-methanoindene
DDT	1,1,1-trichloro-2,2-bis( <i>p</i> -chlorophenyl)ethane
Dieldrin	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-endo,exo-5,8-dimethanonaphthalene
Endrin	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-endo,endo-5,8-dimethanonaphthalene
Heptachlor	1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-endo-methanoindene
Heptachlor epoxide	1,4,5,6,7,8,8-heptachloro-2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindene
Toxaphene	Octachlorocamphene

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